20 December 2023—Revision 1—American Mineralogist (MS #9004R) 1 2 An evolutionary system of mineralogy, Part VIII: 3 The evolution of metamorphic minerals 4 5 SHAUNNA M. MORRISON,<sup>1</sup> ANIRUDH PRABHU,<sup>1</sup> AND ROBERT M. HAZEN<sup>1,\*</sup> 6 7 <sup>1</sup>Earth and Planets Laboratory, Carnegie Institution for Science, 8 9 5251 Broad Branch Road NW, Washington DC 20015, U. S. A. 10 11 12 ABSTRACT 13 Part VIII of the evolutionary system of mineralogy focuses on 1220 metamorphic mineral 14 species, which correspond to 755 root mineral kinds associated with varied metamorphic rock 15 types, most of which likely formed prior to the Phanerozoic Eon. A catalog of the mineral 16 modes of 2785 metamorphic rocks from around the world reveals that 94 mineral kinds often 17 occur as major phases. Of these common metamorphic minerals, 66 are silicates, 14 are oxides 18 or hydroxides, 8 are carbonates or phosphates, 4 are sulfides, and 2 are polymorphs of carbon. 19 Collectively, these 94 minerals incorporate 23 different essential chemical elements. 20 Patterns of coexistence among these 94 minerals, as revealed by network analysis and 21 Louvain community detection, point to six major communities of metamorphic phases, three of 22 which correspond to different pressure-temperature (P-T) regimes of metamorphosed siliceous 23 igneous and sedimentary rocks, while three represent thermally altered carbonate and calc-24 silicate lithologies. 25 Metamorphic rocks display characteristics of an evolving chemical system, with significant

26 increases in mineral diversity and chemical complexity through billions of years of Earth history.

27 Earth's first metamorphic minerals formed in thermally altered xenoliths and contact zones 28 (hornfels and sanidinite facies) associated with early Hadean igneous activity (> 4.5 Ga). The 29 appearance of new Hadean lithologies, including clay-rich sediments, arkosic sandstones, and 30 carbonates, provided additional protoliths for thermal metamorphism prior to 4.0 Ga. 31 Orogenesis and erosion exposed extensive regional metamorphic terrains, with lithologies 32 corresponding to the Barrovian sequence of index mineral metamorphic zones appearing by 33 the Mesoarchean Era (> 2.8 Ga). More recently, rapid subduction and rebound of crustal 34 wedges, coupled with a shallowing geothermal gradient, has produced distinctive suites of 35 blueschist, eclogite, and ultrahigh pressure metamorphic suites (< 1.0 Ga). The evolution of 36 metamorphic minerals thus exemplifies changes in physical and chemical processes in Earth's 37 crust and upper mantle. 38

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- 41 **Keywords:** metamorphism; Barrovian sequence; philosophy of mineralogy; classification;
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43 analysis

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## INTRODUCTION

46 *Metamorphism*: the word, itself, means change through time—"evolution" by that 47 contentious word's most basic definition. Every metamorphic mineral assemblage derives from 48 prior minerals. Each metamorphic rock has a history revealed in the varied attributes of its 49 phases—deep-time stories of changing crustal and mantle environments that epitomize mineral 50 evolution.

And yet, paradoxically, metamorphic minerals are among the most difficult to place into an unambiguous, historical narrative. Each new suite of metamorphic phases—generation after generation of prograde followed by retrograde transformations—may partially mask what came before. Unlike the holocrystalline igneous assemblages featured in Part VII of this series (Hazen et al. 2023), coexisting metamorphic minerals are often in disequilibrium. In this contribution, though not fully resolving those ambiguities, we attempt to add metamorphic minerals to the larger context of the evolving mineral kingdom.

58 The evolutionary system of mineralogy is an effort to place all of the more than 6000 mineral 59 species approved by the International Mineralogical Association's Commission on New 60 Minerals, Nomenclature, and Classification (IMA-CNMNC; https://rruff.info/ima, accessed 17 61 December 2023) into their historical and paragenetic contexts (Hazen 2019; Cleland et al. 2021; 62 Hazen and Morrison 2022; Hazen et al. 2022). The initial five parts of the system cataloged 63 almost 300 mineral kinds that occur as primary and secondary phases in meteorites (Hazen and Morrison 2020, 2021; Morrison and Hazen 2020, 2021; Hazen et al. 2021). Part VI focused on 64 65 Earth's earliest mineralogy, including 262 species formed via a variety of igneous, hydrothermal, aqueous alteration, and other near-surface processes (Morrison et al. 2023). 66

67 Part VII on "The evolution of the igneous minerals" documented 1665 primary species that

68 crystallized from a melt, with special attention to associations and antipathies among the most

- 69 common 115 kinds of igneous minerals (Hazen et al. 2023).
- 70 Our exploration of the evolution of metamorphic minerals is organized into five main 71 sections.
- 721. The first section considers the unusual character of metamorphic mineral evolution

as a historical pursuit, while outlining four significant challenges to this effort.

- Section 2 reviews the sources of mineralogical data employed in this study, while
   introducing eight major groups of metamorphic minerals that represent different
   formational environments and/or compositional ranges.
- 77 3. As in prior parts of the evolutionary system, extensive tabulations of metamorphic 78 mineral co-existence data are particularly ammenable to mineral network analysis 79 (Newman 2010; Morrison et al. 2017) and community structure analysis (Girvan and 80 Newman 2002; Fortunato 2010). Therefore, the third section applies these methods 81 to explore patterns of mineral coexistence in metamorphic rocks. We present a data 82 resource that records patterns of coexistence among 73 relatively common 83 metamorphic minerals in 2785 diverse metamorphic rocks, with a consideration of 84 potential preservational and anthropogenic biases in the literature of metamorphic 85 petrology.
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  4. Section 4, addressing the evolution of metamorphic minerals at a planetary scale,
  87
  87 considers if there have been significant changes in the nature of metamorphism
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This is the peer-reviewed, final accepted version for American Mineralogist, published by the Mineralogical Society of America. The published version is subject to change. Cite as Authors (Year) Title. American Mineralogist, in press. DOI: https://doi.org/10.2138/am-2023-9004. http://www.minsocam.org/ 89 5. Finally, Appendix I details the nature and distribution of 94 of the most common 90 minerals in metamorphic rocks, as well as the modified nomenclature employed in 91 this contribution for some mineral kinds. 92 93 **1.** ON THE NATURE OF "METAMORPHIC MINERAL EVOLUTION" 94 In Part VIII of the evolutionary system we consider metamorphic minerals, many of which 95 first appeared prior to 2.5 Ga, though some high-pressure metamorphic lithologies appear to 96 be confined to the past billion years (Carswell and Compagnoni 2003; Palin and White 2016; 97 Brown and Johnson 2019). The strategy, as outlined in previous contributions, is to identify 98 historical processes that produced minerals with diagnostic combinations of physical and 99 chemical attributes—what we suggest are "historical natural kinds" (Boyd 1991, 1999; Hawley

and Bird 2011; Magnus 2012; Khalidi 2013; Ereshevsky 2014; Godman 2019; Cleland et al. 2021;

101 Hazen et al. 2022). By so doing, the evolutionary system complements standard taxonomic

102 protocols of the IMA-CNMNC, which differentiate each mineral "species" based on its unique

103 combination of major element chemistry and idealized atomic structure (e.g., Burke 2006; Mills

104 et al. 2009; Schertl et al. 2018; Hatert et al. 2021; Hazen 2021; Hawthorne et al. 2021).

Here we consider the diversity, distribution, and ages of 1220 metamorphic minerals, all but five of which (*Fe-dolomite, olivine, phengite, plagioclase,* and *silicate glass*) are species approved by the IMA-CNMNC (https:rruff.info/ima; accessed 17 March 2023). We define a metamorphic mineral as a naturally occurring solid phase that forms by the transformation of one or more prior solid phases through the sustained action of temperature and/or pressure. This definition is consistent with that of Philpotts and Ague (2009): "Metamorphism is the sum

111 of all changes that take place in a rock as a result of changes in the rock's environment; that is, 112 changes in temperature, pressure (directed as well as lithostatic), and composition of fluids. 113 The changes in a rock may be textural, mineralogical, chemical, or isotopic." Note, however, 114 that we restrict the focus to mineralogical changes resulting from T >  $\sim$ 150 °C up to 115 temperatures that cause melting and magma genesis. By this definition, we include regional 116 metamorphism from zeolite to ultrahigh temperature (UHT) facies; contact (i.e., thermal) 117 metamorphism from hornfels and sanidinite facies; and ultrahigh pressure metamorphism from 118 blueschist, ecologite, and ultrahigh pressure (UHP facies). However, we do not consider 119 alteration by impacts, lightning, or other transient processes that are considered in other parts 120 of this series (e.g., Hazen et al. 2023; Morrison et al. 2023). Furthermore, alterations that result 121 from biological sources, notably pyrometamorphism by hydrocarbon combustion, as well as 122 metamorphism of biomediated protoliths such as coal and phosphorites, will be considered in a 123 subsequent part of this series.

124 Metamorphism might seem the quintessential example of mineral evolution because 125 metamorphic assemblages by definition require sequential mineralogical changes through time. 126 However, the conventions of the evolutionary system of mineralogy result in four inherent 127 difficulties related to: (1) determining the timing of metamorphism; (2) documenting the 128 sequence and environment of metamorphic mineral paragenesis; (3) dealing with the critical 129 importance of many mineral's variable compositions owing to solid solution; and (4) 130 deciphering the important roles of fluids in metamorphism. In this contribution we only 131 partially resolve these four challenges.

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1331. The timing of metamorphic mineralization: Mineral evolution, as originally presented by134Hazen et al. (2008), considers the near-surface (< 3 km) mineralogy of terrestrial planets</td>135and moons. Three factors were cited to justify this depth criterion: (1) these are the136minerals most easily documented on Earth, (2) they are the minerals we are most likely to137observe on other terrestrial worlds, and (3) interactions with cellular life occur primarily138in this near-surface domain. Any consideration of the mineral evolution of much deeper

139 regimes must inevitably lead to increased degrees of speculation and uncertainty.

140 Complications may arise because Earth's near-surface mineral inventory originates 141 both from near-surface paragenesis (e.g., volcanism, evaporation, biomineralization), and 142 from much deeper processes (igneous intrusion, deep hydrothermal mineralization, 143 regional and ultrahigh pressure metamorphism). In the latter instances, a mineral is 144 included in our tabulations only if it subsequently appears in the shallow crust, usually 145 through some combination of volcanic, tectonic, and/or erosional processes. 146 Consequently, the timing of a metamorphic mineral's appearance in a relatively shallow 147 crustal environment likely postdates its formation age by tens to as much as hundreds of 148 millions of years (e.g., Ganade et al. 2023)—a vast interval during which retrograde 149 compositional and structural alterations are common.

Evidence for this protracted history is provided by the observation that the volume of metasedimentary rocks <u>at Earth's surface</u> for different geological ages has significantly decreased over the past 600 Myr. An average of > 40 vol % of the exposed lithologies of Neoproterozoic age (1000 to 541 Ma) are metamorphic rocks, compared to < 20 vol % of surface lithologies from the subsequent Phanerozoic Eon (Bluth and Kump 1991; Peters

155 and Husson 2017; Peters et al. 2018; Lipp et al. 2021; https://macrostrat.org, accessed 31 156 December 2022). In spite of this disparity, we suggest that the total volume of 157 metasediments, including those still buried deep within the crust, has likely not 158 diminished significantly over the past billion years. The occurrences, extents, and ages of 159 near-surface metamorphic minerals are as much functions of the rates of tectonic and 160 erosional processes as they are of the deep transformation of protoliths at pressure and 161 temperature. Therefore, somewhat paradoxically, the evolution of metamorphic minerals 162 at once provides some of the clearest examples of an evolutionary sequence of 163 mineralogical states, while defying any simple statistical evaluation of the changing 164 diversity and distribution of metamorphic minerals through deep time.

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166 2. The sequence of metamorphic mineralization: The inevitable extended time intervals 167 required for a protolith's burial, metamorphism, and uplift are particularly challenging in 168 any attempt to document the exact sequence of a metamorphic mineral's evolution. On 169 the one hand, metamorphic minerals, in contrast to those from most other paragenetic 170 processes, represent a true *sequential* and *congruent* evolutionary pathway. Every 171 metamorphic mineral assemblage was derived by the stepwise alteration of an igneous, 172 sedimentary, or metamorphic protolith (Goldschmidt 2011), often with multiple 173 transformations that reflect changes in the mineralizing temperature, pressure, and/or 174 fluid composition (Barrow 1893; Eskola 1920; Bowen 1940; Tilley 1951). One can thus 175 visualize an evolutionary metamorphic pathway with successive prograde and retrograde 176 chemical reactions, illustrated for example on pressure-temperature-time diagrams

177 (Philpott and Ague 2009, their Figure 16-6, and references therein) or through a series of

tree or network graphs (e.g., Heaney 2016; Morrison et al. 2017).

On the other hand, the temporal details of a metamorphic rock's history may be scrambled and difficult to interpret. Many metamorphic processes occur deep in the crust or upper mantle; hundreds of millions of years may be required for tectonic processes and erosion to expose highly altered metamorphic terrains.

183 An additional conundrum relates to metamorphic rocks, particularly lower grade 184 contact and regional metamorphic formations, with biologically-derived protoliths. The 185 processes of contact and regional metamorphism likely commenced long before the first 186 living cells, and many metamorphic formations are unambiguously abiotic—i.e., arising 187 via purely chemical and physical processes. However, the burial and alteration of coal, 188 phosphorites, and a wide range of fossil-bearing limestone, marl, shales, and other 189 biogenic deposits must postdate their Phanerozoic origins. Furthermore, the resultant 190 metamorphic minerals, even if representing the same IMA-CNMNC-approved species as 191 those in much more ancient abiotic rocks, may preserve diagnostic biosignatures in their 192 trace elements, isotopes, morphologies, and other attributes. If so, then by the 193 conventions of the evolutionary system these metamorphic phases represent distinct and 194 relatively recent biologically mediated mineral kinds. Consequently, we defer discussion 195 of the metamorphism of biotic protoliths until the final part of this series.

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*3. Mineral solid solution*: Most diagnostic metamorphic minerals display significant solid
 solution, ranging from binary systems [e.g., olivine (Mg,Fe)<sub>2</sub>SiO<sub>4</sub>], to minerals with 4 or

of

"hornblende"

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199	more	chemical	degrees	of	freedom	[e.g.,	"hornblende"
200	(Na,K)Ca	2(Mg,Fe <sup>2+</sup> ,Al,Fe	<sup>3+</sup> ) <sub>5</sub> (Si,Al) <sub>8</sub> O <sub>22</sub> (C	OH,F,Cl)₂]	. Continuous c	omposition	al variations with
201	changes	in temperature	e, pressure, and	d/or fluid	composition a	are central t	to understanding
202	metamo	rphic processe	es, especially	teasing	out the evo	olution fro	m one mineral
203	assembla	age to the next.	However, the	nomencl	ature adopted	by the IMA	-CNMNC, as well
204	as in the	evolutionary s	ystem in its pr	resent pro	eliminary form	n, are ill-sui	ted to describing
205	such sub	tle yet critical s	hifts in elemen	t ratios.			

206 For example, both the IMA and evolutionary systems employ the end-member olivine 207 minerals forsterite (Mg<sub>2</sub>SiO<sub>4</sub>) and fayalite (Fe<sub>2</sub>SiO<sub>4</sub>), neither of which is particularly useful 208 when dealing with the variable intermediate Mg-Fe compositions of olivine in 209 metamorphosed mafic and ultramafic rocks. Consequently, we add *olivine* as a mineral 210 kind defined as  $(Mg,Fe)_2SiO_4$  with 0.3 < Fe/(Fe + Mg) < 0.7. Similarly, while the IMA 211 system names only end-member compositions in the ternary of Ca-Na-K feldspars as valid 212 mineral species, most natural specimens have intermediate compositions along either the 213 Ca-Na (plagioclase) or Na-K (alkali feldspar) binaries and, in the extreme case of ultrahigh 214 temperature facies, feldspar compositions may lie well within the ternary region (e.g., 215 Harley 2021, their Figure 20).

216 This situation is especially concerning when comparing the mineralogy of very 217 different protoliths. For example, near end-member forsterite formed during the contact 218 metamorphism of dolomite differs significantly from intermediate olivine in metabasalt 219 with Mg > Fe. Both minerals are classified as "forsterite" in the IMA formalism, though we 220 call intermediate Mg-Fe olivine compositions olivine. Until we have much more

information on the compositional idiosyncrasies associated with minerals from different
 metamorphic environments (i.e., the "metamorphic mineral kinds"), these ambiguities in
 nomenclature will limit our efforts.

224 An additional challenge with respect to mineral nomenclature is the use of some 225 names in the metamorphic petrology literature that do not coincide with approved 226 mineral species. For example, "breunnerite" is not an approved mineral name, yet it is 227 commonly applied to intermediate compositions of the magnesite—siderite [(Mg,Fe)CO<sub>3</sub>] 228 solid solution, where 0.1 > Fe/(Mg+Fe) > 0.3. In our study, all such occurrences are 229 reported as the Mg end-member, magnesite. Ankerite, by contrast, is an IMA-approved 230 name for minerals in which Fe > Mg on the dolomite—ankerite [Ca(Mg,Fe)(CO<sub>3</sub>)<sub>2</sub>] binary. 231 However, "ankerite" has been traditionally employed in the metamorphic petrology 232 literature for a range of ferroan dolomites, including the majority with Fe/(Mg + Fe) < 0.5233 (e.g., Chang et al. 1996; Ferry 1996; Ferry et al. 2015)—all occurrences that IMA would 234 designate as dolomite. Consequently, important information related to intermediate 235 compositions is lost through standardized mineral nomenclature.

Accordingly, in this study we employ several names for solid solutions. *Augite* and *pigeonite* are anomalous IMA-approved names for broad solid solutions among Ca-Mg-Fe clinopyroxenes. In this contribution we adopt the non-approved names "*Fe-dolomite*" for *dolomite* with 0.15 < Fe/(Fe + Mg) < 0.50, "*olivine*" for examples with 0.3 < Fe/(Fe + Mg) < 0.70, and "*plagioclase*" for intermediate Ca-Na feldspar compositions with 0.15 < Ca/(Ca + Na) < 0.85. We also employ a number of group names such as *biotite*, *hornblende*, *scapolite*, *serpentine*, and *tourmaline* to lump several closely related species—a

243 convention that is especially useful for mineral groups for which the exact species is rarely

reported in the petrologic literature (see Appendix 1).

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246 4. The role of fluids: Another ambiguity, as yet imperfectly resolved in our treatment, relates 247 to the role of fluid alteration during metamorphic processes. Hydration/dehydration, 248 carbonation/decarbonation, and other fluid-rock interactions are integral to many 249 metamorphic reactions. However, metasomatic alteration, by which a protolith reacts 250 with external fluids, introduces complexities. We define metamorphism as mineralogical 251 changes induced by changes in temperature and/or pressure, whether or not a fluid of 252 differing composition is also involved. By contrast, in metasomatism mineralogical 253 changes are caused by an influx of fluids of differing composition, whether or not changes 254 in temperature and/or pressure are involved.

255 The processes of metamorphism and metasomatism thus differ conceptually; 256 however, sharp boundaries do not exist between the two (Ramberg 1952). For example, 257 Joplin (1968) notes that their "division of contact metamorphism and contact 258 metasomatism into separate chapters is an artificial one since the two processes are very 259 closely associated and cannot readily be separated." Consequently, in some cases 260 outlined below we attempt to include minerals that have likely been compositionally 261 transformed by proximal fluids (e.g., boron-fluorine metasomatism in some skarns; Tilley 262 1951; Marincea and Dumitras 2019), whereas in other instances we exclude minerals for 263 which the alteration to a new phase appears to be principally related to fluids derived 264 from sources more distant in space and/or time. Note that weathering, lithification, and

This is the peer-reviewed, final accepted version for American Mineralogist, published by the Mineralogical Society of America. The published version is subject to change. Cite as Authors (Year) Title. American Mineralogist, in press. DOI: https://doi.org/10.2138/am-2023-9004. http://www.minsocam.org/ 265 diagenesis, as well as seafloor alteration by serpentinization, will be considered in Part IX, 266 even if temperatures may be significantly greater than 150 °C in some instances. 267 268 In spite of these four challenges, we adopt the strategy employed in Part VII (igneous 269 mineral evolution; Hazen et al. 2023) by tabulating the modes of diverse metamorphic rocks 270 and investigating observed patterns of mineral coexistence, both their associations and their 271 antipathies. 272 273 2. ON THE DISTRIBUTION OF MINERALS IN METAMORPHIC ROCKS 274 An examination of the varied formation processes of all mineral species approved by the 275 IMA-CNMNC (Hazen and Morrison 2022; https://rruff.info/ima, accessed 17 January 2022) 276 reveals 1220 minerals that have been reported as phases in metamorphic rocks. Accordingly. 277 we tabulate 1215 metamorphic mineral species approved by the IMA-CNMNC, as well as Fe-278 dolomite, olivine, phengite, plagioclase, and silicate glass [see Supplementary Table 1 and 279 associated Supplementary Read-Me File 1; see also Hazen and Morrison (2022; their 280 Supplementary Table 1 and additions)]. Table 1 catalogs 5 Supplementary Tables and an 281 interactive graphical figure associated with this contribution.

- 283 **Table 1.** Catalog of 5 Supplementary Tables (see also 5 associated Supplementary Read-Me
- Files) and an Interactive Graphical Figure (see figure caption for links and instructions).
- 285
- 286 Supplementary Table 1. A list of 1220 metamorphic mineral species (including Fe-dolomite,
- 287 *plagioclase*, and *silicate glass*), and their correspondence to 755 root mineral kinds, with
- compositions and distributions among 8 metamorphic rock types.
- 289 Supplementary Table 2. A list matching 652 IMA-approved species with their associated 187
- 290 mineral kinds, which are defined by lumping two or more species.
- 291 <u>Supplementary Table 3</u>. A matrix listing the modes of 2785 metamorphic rocks. We record the
- distribution of 94 of the most common metamorphic minerals among these rocks.
- 293 Supplementary Table 4. A 73 x 73 symmetrical matrix that records the numbers of co-
- 294 occurrences among 73 of the minerals in Supplementary Table 3. We do not include 21 295 minerals that occur commonly in multiple types of metamorphic environments.
- 296 Supplementary Table 5. A 73 x 73 symmetrical matrix (derived from Supplementary Table 4)
- 297 that records the percentage of the less common mineral that co-occurs with the more 298 common mineral.
- 299 <u>Interactive Figure 1.</u> A unipartite network of coexistence among 73 of the most common
   300 metamorphic minerals, based on data in Supplementary Tables 3, 4, and 5. The nodes are
   301 colored according to 6 communities, based on Louvain Community Detection.
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304 Mineral natural kinds: The list of 1220 metamorphic mineral species is significantly modifed in 305 the evolutionary system of mineralogy by rules for "lumping and splitting" (Hazen et al. 2022). 306 We have thus consolidated the list of 1220 minerals to recognize 755 "natural kinds" (Hawley 307 and Bird 2011; Magnus 2012; Bird and Tobin 2015; Hazen 2019; Cleland et al. 2021) of metamorphic minerals [Appendix I; Supplementary Table 2 and associated Supplementary 308 309 **Read-Me File 2;** see also Hazen et al. 2022, their Supplementary Table 1 and additions)]. 310 Of the 1220 metamorphic mineral species, 568 correspond exactly to 568 root mineral kinds, 311 whereas the other 652 species are lumped into 187 root mineral kinds. The majority of the 312 resulting 755 metamorphic root mineral kind names (each iltalicized) are the same as the 313 corresponding IMA mineral species name; thus, the IMA species albite is equivalent to the root 314 mineral kind *albite*. In 26 instances, we adopt the IMA-approved mineral name, minus its suffix 315 or prefix, as the root mineral name for groups of two or more closely-related species; thus, 316 chabazite is the root mineral name for the four lumped species chabazite-Ca, chabazite-K, 317 chabazite-Mg, and chabazite-Na, whereas *apatite* is the root mineral name for fluorapatite and 318 hydroxylapatite. In 18 instances (androsite, apophyllite, biotite, chlorite, ellestadite, hogbomite, 319 hornblende, kspar, leakeite, melilite, orthoenstatite, Os-Ru alloy, Pd-Pt-Rh alloy, scapolite, 320 serpentine, taaffeite, tourmaline, and wolframite), we employ an unapproved mineral kind 321 name for a group of closely-related IMA-approved mineral species. Thus, for example, 322 tourmaline is the root mineral kind name for 18 IMA-approved species formed by metamorphic 323 processes in the tourmaline group.

As suggested above, this proposed nomenclature is a first step in developing a much richer mineral taxonomy related to historical mineral kinds. In the case of metamorphic minerals,

numerous additional subdivisions are suggested. For example, our approach has yet to recognize and name many of the intermediate compositions of such important metamorphic mineral groups as amphiboles, carbonates, feldspars, micas, olivines, or pyroxenes. Until large databases of mineral compositions and their petrologic contexts are interrogated with cluster analysis (e.g., Gregory et al. 2019; Boujibar et al. 2021; Hystad et al. 2021), any consideration of mineral associations will be limited.

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333 Metamorphic paragenetic modes: One approach to understanding metamorphic mineral 334 evolution lies in documenting the coexistence of phases in different groups of metamorphic 335 rocks. Supplementary Table 1 lists the distribution of these phases among 8 distinctive groups 336 of metamorphic rocks (see also Table 2), each with characteristic mineral assemblages and each 337 designated by a 3-letter abbreviation, as well as the paragenetic mode number (p##) originally 338 employed by Hazen and Morrison (2022). In the course of this work we have expanded and 339 revised the list of paragenetic modes associated with metamorphic minerals. In Supplementary 340 Table 1, as indicated in red highlights, we have added 273 new combinations of a mineral 341 species and metamorphic paragenetic mode, while removing 7 entries for which no supporting 342 information could be found.

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Table 2. Division of mineral species and "root mineral kinds" among eight metamorphic paragenetic modes

<u>Pa</u>	aragenetic Mode	Code <sup>a</sup>	Age (Ga)	# Species <sup>b</sup>	#Unique <sup>c</sup>	# Kinds <sup>d</sup>
P)	rometamorphism of xenoliths	XEN; p09	> 4.5	173	87	130
Co	ontact metamorphism	CON; p31	> 4.0	424	241	264
M	etamorphic Ba/Mn/Pb/Zn deposits	BAM; p32	> 3.0	449	375	310
O	phiolites	OPH; p38	> 3.0	109	69	96
Hi	gh-pressure metamorphism	HPM; p39	< 1.0	113	29	89
Re	egional metamorphism	REG; p40	< 3.0	351	151	205
M	antle metasomatism	MET; p41	> 4.0	37	5	35
<u>Sh</u>	near-induced minerals	SHE; p43	> 4.0	30	2	29
<sup>a</sup> F	Paragenetic codes from Hazen and N	1orrison (202	22).			
1 <sup>d</sup>	Number of IMA-CNMNC-approved sp	pecies associ	ated with t	his parageneti	c mode.	
۲	Number of IMA-CNMNC-approved sp	pecies unique	e to this par	ragenetic mod	e.	
<mark>4 b</mark>	Number of root mineral kinds associa	ated with thi	is paragene	tic mode.		
In						
ages	the following sections, we su of the eight groups of metam	mmarize t orphic mir	he components	ositions, pro sidered in tl	ocesses, dive his contribut	rsity, and poss ion.
ages <u>1</u> .	of the eight groups of metamon of the eight groups of metamon <u>Pyrometamorphism of xeno</u>	mmarize t orphic mir <u>liths (XEN;</u>	:he compo nerals con : <i>p09</i> ): The	ositions, pro sidered in tl ermally alte	ocesses, dive his contribut red xenolith	rsity, and poss ion. s are derived fi
ages	of the eight groups of metamo of the eight groups of metamo <u>Pyrometamorphism of xeno</u> both crustal and mantle so	orphic mir orphic mir <u>liths (XEN;</u> ources sub	he componentials con perals con <u>pog):</u> The jected to	ositions, pro sidered in tl ermally alte high-tempe	ocesses, dive his contribut red xenolith erature, low-	rsity, and poss ion. s are derived fi -pressure horn
ages <u>1.</u>	of the eight groups of metamo of the eight groups of metamo <u>Pyrometamorphism of xeno</u> both crustal and mantle so and sanidinite facies metamo	orphic mir orphic mir <u>liths (XEN;</u> ources sub <u></u> oorphism b	the componentials con the componentials con the contract contract to the contract contract to the contract to the contract to the contract to the contract to	ositions, pro sidered in tl ermally alte high-tempe s melts (Gra	ocesses, dive his contribut red xenolith erature, low- pes 2006). T	rsity, and poss ion. s are derived fi -pressure horn ypically formed
ages <u>1</u> .	<ul> <li>the following sections, we su</li> <li>of the eight groups of metame</li> <li><u>Pyrometamorphism of xeno</u></li> <li>both crustal and mantle so</li> <li>and sanidinite facies metame</li> <li>temperatures above 900 °C</li> </ul>	orphic mir orphic mir <u>liths (XEN;</u> ources sub <u></u> norphism b C and pres	the components con merals con <u>pog):</u> The jected to by igneous ssures les	ositions, pro sidered in tl ermally alte high-tempe s melts (Gra s than 0.5	ocesses, dive his contribut red xenolith erature, low- pes 2006). T GPa, pyrom	rsity, and poss ion. s are derived fi -pressure horn ypically forme etamorphic su
ages <u>1</u> .	<ul> <li>the following sections, we su</li> <li>of the eight groups of metame</li> <li><u>Pyrometamorphism of xeno</u></li> <li>both crustal and mantle so</li> <li>and sanidinite facies metame</li> <li>temperatures above 900 °C</li> <li>were likely the earliest o</li> </ul>	Immarize t orphic mir <u>liths (XEN;</u> ources sub <u></u> norphism b C and pres	the components con merals con <u>pog):</u> The jected to by igneous ssures les near-sur	ositions, pro sidered in tl ermally alte high-tempe s melts (Gra s than 0.5 face metan	ocesses, dive his contribut red xenolith erature, low- pes 2006). T GPa, pyrom norphic lith	rsity, and poss ion. s are derived fi pressure horn ypically formed etamorphic su ologies. The
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372 conditions.

- 373 We identify 173 IMA-approved mineral species, corresponding to 130 mineral kinds, 374 that formed by xenolith pyrometamorphism. Based on our surveys, 87 of these phases 375 are unique to xenoliths. Of special note are the more than 50 Ca-bearing limestone 376 xenolith minerals from the intensively studied lavas of the Somma-Vesuvius Complex, 377 Naples, Italy (https::/mindat.org; accessed 06 January 2023). 378 Note that under this heading we do not include several groups of relatively recent (< 379 400 Ma) biologically-mediated pyrometamorphic assemblages, including minerals 380 formed in coal mine fires or by hydrocarbon fires, notably from the Hatrurim Basin
- 382 minerals from coal and other carbon-rich lithologies altered as xenoliths in lava or 383 through contact metamorphism are deferred until Part XII of this series.

(Grapes 2006; https://mindat.org, accessed 17 March 2023). Similarly, pyrometamorphic

384

381

385 2. Thermal alteration via contact metamorphism (CON; p31): Contact metamorphism 386 occurs when an igneous intrusive body thermally alters the older host country rock, 387 including sedimentary, igneous, and metamorphic lithologies. Contact metamorphism is 388 similar in many respects to the thermal alteration of xenoliths. However, contact 389 metamorphism often differs by the extent of reactions with proximal aqueous fluids, 390 notably those rich in carbonate and phosphate, as well as sulfate, borate, halogens, and 391 other solutes derived from both the magma and the country rock (Einaudi et al. 1981; 392 Einaudi and Burt 1982; Button 1982; Falkowski et al. 2000; Klein 2005; Kappler et al. 393 2005). We tabulate 424 mineral species (264 mineral kinds) that form through contact 394 metamorphism, including phases that arise through reactions with local fluids. We find

395 that 241 of these species are unique to contact metamorphic environments. Note that 396 continents with deep roots and active hydrological cycles also formed minerals by high-397 temperature aqueous alteration through metasomatism—processes to be considered 398 further in Part IX of this series.

Contact metamorphism is an ancient process that must have commenced as early as a second generation of magma penetrated the first cooling Hadean crust. The subsequent history of contact metamorphism and its expanding mineral diversity parallels the evolution of ever more differentiated crustal lithologies. Of special interest are limestones and dolomites, which are particularly susceptible to transformation by hot fluids emanating from acid intrusions—assemblages known as skarns (Bowen 1940; Harker 1950; Tilley 1951).

406

407 3. Metamorphic Ba/Mn/Pb/Zn deposits (BAM; p32): We distinguish metamorphic phases 408 rich in otherwise minor metal elements, including Ba, Mn, Pb, and/or Zn, because they 409 feature distinctive mineral assemblages in both contact metamorphic and regional 410 metamorphic contexts (Post 1999; Leach et al. 2005). With 449 species (310 mineral 411 kinds), 375 of which are unique to these environments, these deposits are the most 412 mineralogically diverse of any metamorphic group. Several classic localities, including 413 Broken Hill, Australia (Spry et al. 2008); Franklin, Sussex County, New Jersey (Peters et al. 414 1983; Frondel 1990); Fresno County, California (Alfors et al. 1965); and the Wessel 415 manganese mine, South Africa (Cairncross and Beukes 2013), highlight the role of a few 416 mineral-rich localities in enhancing Earth's mineral diversity. Note, however, that these

deposits are volumetrically insignificant compared to regional and contact metamorphic
rocks of more common igneous and sedimentary protoliths. Consequently, these
uncommon phases do not appear in our lists of metamorphic mineral modes
(Supplementary Table 3 and associated Supplementary Read-Me File 3).

421

422 4. Ophiolites (OPH; p38): Ophiolites incorporate highly altered sequences of mafic and 423 ultramafic rocks from the deep oceanic lithosphere—phases that have been 424 subsequently obducted onto crustal formations. As such, they provide important insights 425 to the mineralogy and petrology of the crust-mantle boundary (Moores 2002; Dilek 2003; 426 Kusky 2004). Ophiolites are documented to hold at least 109 mineral species (96 mineral 427 kinds), some of which resemble those of ocean floor igneous rocks altered by 428 serpentinization. However, we consider them as a separate metamorphic group for their 429 unique subaerial exposures of mantle lithologies, because of the occurrence of the 430 mineralogically distinctive and enigmatic Luobusha ophiolite from the Shannan 431 Prefecture of Tibet (references in Litasov et al. 2019), as well as minealogically diverse 432 ophiolite localities in the Urals and other localities (references in Litasov et al. 2019). 433 Luobusha is of special interest for its puzzling suite of ultrahigh pressure minerals (e.g., 434 diamond and moissanite) that co-occur with dozens of highly reduced phases, including 435 native elements (Al, Cr, Cu, Fe, Ti, W), carbides, nitrides, and phosphides that are unique 436 to this locality (Bai et al. 2011). In addition, the Luobusha occurrence features many PGE 437 metal alloys (including possibly as many as 30 undescribed metal phases) that are 438 associated with chromitite zones reminiscent of assemblages in layered intrusions

(references in Litasov et al. 2019; Hazen et al. 2023). We include these diverse, rare
minerals under ophiolites, though details of their paragenesis, and especially their
assignments to metamorphic processes, remain uncertain (Ballhaus et al. 2017; Litasov
et al. 2019).

Extensive alteration of Earth's oldest rocks obscures the identity of the oldest ophiolites, which may have predated the Proterozoic Eon, significantly before 2.5 Ga (Kusky et al. 2001; Zhai et al. 2002; Furnes et al. 2007; Nutman and Friend 2007). Confident identifications of several altered ophiolites support an origin by at least 2.5 Ga (Kusky 2004)—an observation consistent with the establishment of some form of plate tectonics by the late Archean Eon.

449

450 5. High-pressure metamorphism (HPM; p39): So-called "high-pressure" metamorphic 451 assemblages are distinguished by their formation in the deep crust and upper mantle 452 under unusually low geothermal gradients of less than 10 °C/km. Such conditions are 453 only possible during transient subduction and subsequent rapid buoyant uplift of crustal 454 rocks (Chopin 1984; Carswell and Compagnoni 2003; Hacker 2006; Palin and White 2016; 455 Zheng and Chen 2017). High-pressure metamorphic rocks include blueschist facies, 456 typically with *glaucophane*, *jadeite*, and/or *lawsonite* (formed at depths to 30 km); 457 eclogite with omphacite and pyrope (> 45 km); and ultrahigh pressure (UHP) formations, 458 featuring the dense *coesite* form of SiO<sub>2</sub> (> 80 km), micro-*diamond*, and other dense 459 phases that point to origins approaching depths of 200 km. We list 113 high-pressure 460 metamorphic mineral species (89 mineral kinds), of which 29 species, including

461 *barioperovskite* (BaTiO<sub>3</sub>), *ellenbergerite*  $[Mg_6(Mg,Ti^{4+},Zr^{4+},\Box)_2(Al,Mg)_6Si_8O_{28}(OH)_{10}]$ , and

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462 trinepheline (NaAlSiO<sub>4</sub>), are unique to these metamorphic environments.
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463 The ages of the oldest known high-pressure metamorphic rocks (< 850 Ma), and even younger ages for the oldest UHP occurrences (Ganade et al. 2023), reveal much about 464 465 Earth's evolving lower crust and upper mantle (Jahn et al. 2001; Brown 2007; Stern 466 2018). Thus, for example, several of the oldest recorded *lawsonite* occurrences are from 467 the Paleozoic Era (De Roever 1956). These Neoproterozoic and later developments are in 468 sharp contrast to ultrahigh temperature metamorphic rocks from the Archean and 469 Proterozoic Eons (Brown 2006; Harley 2021). In particular, there are no known examples 470 of blueschist or ultrahigh pressure metamorphism until after 700 Ma (Ernst 1972; Palin 471 and White 2016; Brown and Johnson 2019; Holder et al. 2019). These temporal 472 differences apparently reflect the cooling average geotherm over billions of years of 473 Earth history.

474

<u>6. Regional metamorphism (REG; *p40*):</u> Most metamorphic rocks fall under the broad
heading of regional metamorphism, which is associated with burial, alteration, and uplift
of thick accumulations of sediments, volcanic rocks, and intrusive igneous lithologies. We
list 351 regional metamorphic mineral species, representing 205 mineral kinds, with 151
species unique to this paragenesis. Most regional metamorphic minerals derive from
common sedimentary and igneous protoliths.

## 481 Regional metamorphic rocks, which typically formed under an average geothermal 482 gradient of 15 to 30 °C/km (Vernon 2008; Philpotts and Ague 2009), are subdivided into

483 Barrovian index mineral zones based on the appearance of new minerals with increasing 484 temperature and/or pressure: chlorite, biotite, garnet, staurolite, kyanite, and sillimanite, 485 as first outlined by Barrow (1893). Mineral zones also extend to lower pressures, though 486 higher than shallow contact/thermal metamorphism (Miyashiro 1961), as well as to 487 ultrahigh temperature environments (> 900 °C at P < 1.5 GPa; Harley 2021). Goldschmidt 488 (1911) and Eskola (1920) proposed a series of facies of increasing metamorphic grades, 489 originally based on the mineralogy of metabasalt, including greenschist facies (typically 490 with chlorite, epidote, and serpentine), amphibolite facies (hornblende and plagioclase), 491 and granulite facies (a pyroxene group mineral and *plagioclase*). Subsequent research 492 extended metamorphism to the UHT facies (often with orthopyroxene, osumilite, and/or 493 sapphirine). In this overview we have lumped these regional metamorphic zones, though 494 future research may warrant splitting into multiple groups based on ranges of 495 temperature, pressure, and/or composition. Regional metamorphism, unlike high-496 pressure metamorphism, was well established by the Neoarchean Era (> 2.5 Ga).

497 Regional metamorphic mineral zones may form by either prograde metamorphism
498 during heating and/or burial, or retrograde metamorphism during cooling and/or
499 unburial. If close to equilibrium conditions, metamorphic rocks rarely contain more than
500 4 or 5 major minerals, following Gibbs' phase rule. However, many metamorphic mineral
501 assemblages have more than 6 coexisting major phases and therefore may not represent
502 equilibrium mineral assemblages.

503

504	<u>7.</u>	Mantle metasomatism (MET; p41): Deep-seated metasomatism, including processes in
505		both the lower crust and upper mantle (Luth 2003; O'Reilly and Griffin 2012), is a high-
506		pressure process that combines aspects of metamorphism and metasomatism, thereby
507		altering the chemistry of existing ultramafic/mafic minerals and producing new phases
508		through interactions with deep C-O-H fluids (Manning and Frezzotti 2020). We identify
509		37 oxide and silicate mineral species (35 kinds) formed through mantle metasomatism,
510		all but 5 of which (including <i>nixonite</i> , $Na_2Ti_6O_{13}$ ; Anzolini et al. 2019) are familiar minerals
511		in other environments.

512

<u>8.</u> Shear-induced minerals (SHE; *p43*): Minerals that form while experiencing significant
 shear strain represent a distinct metamorphic paragenetic mode in our system. Shear induced mineralization is most commonly associated with the polished fault surfaces
 known as slickensides that represent zones of mylonitization, typically with pyroxene
 converted to amphibole and plagioclase much reduced in grain size and spread out into
 layers (Harker 1950; Passchier and Trouw 2005; Trouw et al. 2009).

519 Such shear zones can also act as fluid conduits (Gates and Speer 2022). Minerals 520 produced by strain/shear can be formed by purely mechanical action, or they may 521 involve chemical changes. Shear phenomena include twinning, as commonly observed 522 with *calcite* in marble; strain bending, for example of mica; inclusion trains; cracks; and 523 recrystallization (Harker 1950).

524 In Supplemental Table 1 we tabulate 30 mineral species (29 kinds) known to form in 525 shear zones, most of which are common oxides and silicates that recrystallize in silicate

526 rocks during shearing. The "sericite" variety of muscovite is among the most common 527 mylonite minerals, often in association with *albite*. In other reactions, *biotite* shears to 528 form chlorite, at times with magnetite; augite or hornblende shears to chlorite plus 529 epidote or calcite; forsterite shears to tremolite or anthophyllite plus talc; andradite 530 transforms in part to *titanite* and *magnetite*; and the antigorite form of *serpentine* arises 531 through shearing of other *serpentine* polymorphs. While most of these phases probably 532 first appeared early in Earth's history, the rare chlorite group mineral donbassite 533  $[Al_2(Si_3Al)O_{10}(OH)_2 \cdot Al_{2,33}(OH)_6]$  has been reported uniquely from slickensides of coal 534 (Anthony et al. 1990–2003), and therefore must have formed within the past 350 million 535 years as a biologically-mediated phase.

536

537 These eight proposed groups of metamorphic rocks, though useful in the larger context of 538 the evolutionary system and its consideration of paragenetic modes writ large, are subjective 539 and fail to properly represent the diversity of metamorphic P-T-X environments. Inevitably, 540 significant overlaps occur in the compositional, environmental, mineralogical, and temporal 541 ranges of the eight broad categories of metamorphic rocks outlined above. Nevertheless, we 542 suggest that each group is associated with its own characteristic mineral assemblages and 543 environmental contexts (e.g., Deer et al. 1982-2013; Anthony et al. 1990-2003). Therefore, each 544 of the eight processes plays its own distinctive role in the evolutionary system of mineralogy.

545

## 3. METAMORPHIC MINERAL ASSOCIATIONS AND NETWORK ANALYSIS

Bowen (1928) employed patterns of "mineral associations and antipathies" in the development of his theory of igneous rock evolution, in which he recognized that some pairs of minerals are frequently encountered in equilibrium igneous assemblages, whereas as others never occur (Hazen et al. 2023). The same principles apply to metamorphic minerals, with the caveat that overlapping prograde and retrograde reactions often result in nonequilibrium

assemblages.

546

554 In this section we amplify Bowen's approach by quantifying the extent of coexistence among

pairs and larger groupings of 94 of the commonest minerals in metamorphic rocks (Table 3;

556 Appendix 1). Of these metamorphic minerals, 66 are silicates, 14 are oxides or hydroxides, 8 are

557 carbonates or phosphates, 4 are sulfides, and 2 are polymorphs of carbon. Collectively, these 94

558 minerals incorporate 23 different essential chemical elements, including oxygen (in 88 of 94

559 minerals), Si (71), Al (44), Fe (40), Ca (38), Mg (37), H (27), Na (14), C (10), F (8), K (8), Ti (7), S

560 (5), Cl (4), Cr (3), Mn (2), Zr (2), and B, Ce, Cu, Ni, P, and Sr (all in only 1 of 94 minerals).

561 The core data of this study are found in Supplementary Table 3 (see also Supplementary 562 Read-Me File 3), which details the distribution of these 94 metamorphic minerals among 2785 563 metamorphic rock modes. We employed 29 primary sources to assemble these metamorphic 564 mineral modes: Augustithis (1985; 156 modes), Botha (1983; 95 modes), Carswell (1990; 198 565 modes), Carswell and Compagnoni (2003; 49 modes), Coleman et al. (1965; 13 modes), Grapes 566 (2006; 218 modes), Harker (1950; 285 modes); Harley (2021; 121 modes); Joplin (1968; 345 567 modes); Philpotts and Ague (2009; 885 modes); Reverdatto and Sóbolev (1973; 71 modes); and 568 Tilley et al. (1964; 50 modes), as well as 299 modes from contributions by Ferry and colleagues

(Davis and Ferry 1983; Ferry 1976, 1984, 1988, 1989, 1992, 1994, 1995, 1996, 2007; Ferry and

570 Rumble 1997; Ferry et al. 1987, 2001, 2002, 2005; Léger and Ferry 1993; Penniston-Dorland and 571 Ferry 2006).

569

Ideally, the extensive data on coexisting metamorphic minerals in Supplementary Table 3 would represent a wide range of equilibrium assemblages, while accurately documenting the relative abundances of metamorphic minerals. However, even though we have surveyed 29 diverse compilations of metamorphic modes representing several geographic areas and most distinct types of metamorphic environments, it is important to recognize at least four likely sources of bias and error in the modal mineralogy data in Supplementary Table 3. Each of these factors may distort the true distribution of mineral associations among metamorphic rocks.

(1) *Biases owing to disequilibrium*: Metamorphic mineral modes often represent nonequilibrium assemblages, with prograde and retrograde phases of differing metastability occurring together. Therefore, the coexistence data in Supplementary Table 3 cannot be employed in the same way as the corresponding table of igneous modes in Part VII of this series.

584 (2) *Biases related to optical petrography:* Most of our modal data come from studies that 585 employed optical petrography, but not electron microprobe analysis. Several inevitable biases 586 result.

Euhedral vs. poorly crystallized phases: Well-crystallized minerals such as almandine
 or staurolite are more likely to be identified than fine-grained assemblages
 containing brucite, phyllosilicates, or zeolites. For example, Ferry (1994), Ferry and
 Rumble (1997), and Ferry et al. (2002) document the widespread occurrence of

- 591 brucite and serpentine—metamorphic minerals almost never recorded in modes 592 from earlier literature.
- Exotic versus common minerals: Our compilation likely over-represents certain rare
   minerals, such as *coesite* and *diamond*, especially if they occur in optically distinctive
   crystals.
- Optically similar minerals: A number of minerals are difficult to identify via optical
   petrography. For example, distinguishing among *calcite, ankerite,* and *aragonite,* as
   well as between fine-grained *quartz/albite* and *muscovite/paragonite,* is difficult and
   may lead to errors in reported modes.
- 600 Opaque minerals: Identification of opaque minerals is another petrographic challenge ٠ 601 that leads to omissions and biases in Supplementary Table 3. For example, electron 602 microprobe analyses indicate that *pyrrhotite* is extremely common in a wide variety 603 of metamorphic rocks (e.g., Ferry 1994), though it is often unreported (or perhaps 604 misidentified as *pyrite*) in earlier descriptions of metamorphic rock modes. Similarly, 605 some opaque Fe-Ti-Cr bearing oxides, including the oxide spinels chromite and 606 hercynite, as well as ilmenite, and rutile, may be under-represented in our 607 tabulations.
- Trace minerals: Detailed studies with electron microprobe analysis suggest that
   minor phases, especially those with grain sizes < 5 micrometers, are much more</li>
   common than suggested by our survey. Examples of rarely reported trace
   metamorphic minerals include *allanite* and *monazite* (Wing et al. 2003); *baddelyite*,

- 612 *geikielite, qandilite,* and *sphalerite* (Ferry 1996); and *anhydrite, calzirtite,* and
- 613 *celestine* (Ferry et al. 2002).

614 (4) Problems in nomenclature: A number of petrographic terms lead to ambiguities when 615 listing minerals in Supplementary Table 3. General group names such as "garnet" encompass 616 several different mineral kinds in our study. Therefore, the relative abundances of almandine, 617 andradite, grossular, and pyrope may be in error. "Spinel" may refer to the mineral species 618 MgAl<sub>2</sub>O<sub>4</sub>, or to members of the oxide spinel group such as *magnetite*, *chromite*, or *hercynite*. "Ankerite" is technically CaFe(CO<sub>3</sub>)<sub>2</sub>, but is often used to designate a Fe-bearing dolomite in the 619 620 literature of metamorphic petrology. 621 (5) Missing recent research: Metamorphic studies of the past 20 years are less likely to

tabulate numerous modes; fewer than a third of the modes in Supplementary Table 3 were first
published after 2003. As a consequence, our survey under-represents more recently
documented metamorphic lithologies, such as ultrahigh temperature metamorphism (Harley
2021).

626

Metamorphic mineral coexistence: Of the 94 minerals considered in Supplementary Table 3, 21 occur commonly in several different types of metamorphic rocks and therefore are not easily grouped by community detection algorithms. Therefore, we tabulated the frequency of cooccurrence of every pairwise combination of the remaining 73 mineral kinds, all of which are more likely to be associated with only one or two metamorphic lithologies (Supplementary Table 4; see also associated Supplementary Read-Me File 4). For each mineral pair we calculated the percentage of the less common mineral that co-occurs with the more common

< n 1

634	mineral (Supplementary Table 5; see also Supplementary Read-Me File 5). For example,
635	consider matrix element W7, which relates to the coexistence of andalusite (with 146
636	occurrences, as listed in Supplementary Table 4) and cordierite (with 395 occurrences). In
637	Supplementary Table 4, matrix element W7 reveals that 84 rocks (out of 2785 tabulated)
638	contain both andalusite and cordierite. Therefore, in Supplementary Table 5, matrix element F7
639	= 84/146 x 100 = 58 %.

640 This protocol is especially important when considering the coexistence of a relatively rare 641 mineral with a common one. For example, baddeleyite is a scarce metamorphic mineral, 642 occurring in only 18 of 2785 metamorphic rocks recorded in Supplementary Table 3. However, 643 16 of those occurrences also contain the common metamorphic mineral calcite. Therefore, as 644 recorded in matrix element Q14 of Supplementary Table 5, 16/18 = 89 % of aegirine 645 occurrences also have *calcite*. Thus, *baddeleyite/calcite* is a common mineral pair whenever the 646 rare mineral baddeleyite occurs. As Bowen emphasized for igneous minerals (Bowen 1928; 647 Hazen et al. 2023), we find that a small percentage of all possible metamorphic mineral pairs 648 commonly occur almandine/biotite; diopside/grossular; chloritoid/muscovite; (e.g., 649 quartz/staurolite).

As demonstrated in our previous study of igneous minerals (Hazen et al. 2023), network analysis of mineral associations and antipathies reveals phase relationships and reaction sequences that may not be immediately recognized from tables of mineral modes. Therefore, we have applied two types of data visualization: unipartite networks that highlight Louvain community detection and bipartite networks representing relationships among minerals and their formational environments.

656

657	Unipartite networks and Louvain community detection analysis of coexisting metamorphic
658	minerals: Mineral network analysis reveals patterns among coexisting minerals (Morrison et al.
659	2017). Accordingly, Figures 1A and 1B display a unipartite network that illustrates the
660	coexistence among 73 relatively common metamorphic minerals, as listed in Table 3. This graph
661	was built on "Observable" (https://observablehq.com/), using D3js (Bostock et al. 2011). The
662	networks use the D3-force algorithm ( <u>https://d3-wiki.readthedocs.io/zh CN/master/Force-</u>
663	Layout/) for its network layout. The code and an interactive version of this network can be
664	found at: (https://observablehq.com/@anirudhprabhu/revised-evolutionary-system-of-mineralogy-
665	part-8-uni; for instructions, see Figure 1 caption).
666	Each of the 73 nodes in Figure 1 represents a metamorphic root mineral kind, with the size
667	of the node in Figure 1A indicating the abundance of that mineral in our tabulations of 2785
668	metamorphic rock modes (Supplementary Table 3). Links between pairs of nodes indicate
669	mineral coexistence. in Figure 1A, we illustrate the case where at least 6 % of occurrences of
670	the less common mineral coexist with the more common mineral, based on percentages
671	tabulated in Supplementary Table 5 (6 % is the highest percentage for which all 73 mineral
672	nodes are still interconnected). Figure 1 is a static rendering of a dynamic interactive network in
673	which the co-occurrence percentage, P, can be varied continuously from 1 to 100 %. This
674	variable feature, as well as other interactive aspects of the online version of Figure 1, facilitates
675	studies of mineral associations and antipathies.
676	

677





Figure 1. (A) A unipartite network of 73 common metamorphic minerals (colored circles), with links
 between pairs of coexisting minerals. Node sizes indicate the relative abundances of the minerals, while
 colors indicate 6 communities of metamorphic minerals that were determined using Louvain community

683 detection (see text). Each of these communities corresponds to a different temperature-pressure-684 composition regime. In this figure, links are drawn between two minerals if at least 6 % of rocks that 685 incorporate the less common mineral also incorporate the more common mineral (as tabulated in 686 Supplementary Table 3). One can vary this percentage in an interactive version of this graph at: 687 (https://observablehq.com/@anirudhprabhu/revised-evolutionary-system-of-mineralogy-part-8-uni). 688 Hover your cursor over any node to identify the corresponding mineral; click and hold your cursor to 689 move that node and identify links to other nodes; use your cursor to move the "Weight Threshold" 690 vernier to systematically eliminate links between nodes based on P values (see text). Adjust node 691 attributes by clicking on the "Size Nodes By" feature. (B) The unipartite network of 73 metamorphic 692 minerals embeds metamorphic rock types as sub-graphs. Highlighted subgraph examples include 6 693 common metamorphic lithologies.

694

Links in a mineral network are not uniformly distributed; they typically display sub-networks 695 696 of more closely connected nodes. We color mineral nodes in Figure 1 based on Louvain 697 community detection analysis (Girvan and Newman 2002; Fortunato 2010), which employs a 698 heuristic method based on modularity optimization to extract the community structure of large 699 networks (Blondel et al. 2008). This method identifies members of a group iteratively in two 700 phases: (1) starting with each node as its own distinct community, larger communities are 701 formed at a local level by maximizing modularity of certain nodes; (2) each small community is 702 aggregated into one "super node" to form a new "super node network." We repeat these steps 703 until the modularity has been optimized and there are no changes in the network. The Louvain 704 modularity approach reveals an optimal number of communities without requiring the user to 705 specify the number of clusters in a dataset. Consequently, Louvain community detection

removes some of the bias associated with some other clustering algorithms, while identifying

the most closely interconnected subsets of minerals.

708 The identification of community structures may be complicated by the inclusion of mineral 709 nodes that link to multiple communities. Such nodes typically plot near the center of a network 710 and "pull" more diagnostic nodes toward the middle while obscuring community structures 711 (Hazen et al. 2023). Therefore, we do not include 21 of the most common metamorphic 712 minerals in our networks, including such major phases as albite, almandine, augite, biotite, 713 hornblende, kspar, muscovite, plagioclase, and guartz, because they occur across a wide variety 714 of metamorphic rocks. In addition, apatite, ilmenite, magnetite, pyrite, pyrrhotite, rutile, titanite 715 and *zircon* are widespread accessory phases that we exclude from network analysis. Βv 716 contrast, each of the 73 metamorphic minerals illustrated in Figure 1 is more characteristic of a 717 restricted pressure, temperature, and/or compositional metamorphic regime. One 718 consequence of this exclusion of 21 of the most widespread metamorphic minerals is that the 719 classic Barrovian sequences of metapelite and metabasite zones are not well represented in 720 Figure 1. Rather, the subset of 73 less ubiquitous phases define six principal communities of 721 metamorphic phases, each with its own distinctive pressure-temperature-composition regime 722 (see also Table 3):

723

Community 1: Community 1 holds 15 high-temperature, low-pressure minerals, including
 such pyrometamorphic and ultra-high temperature phases as corundum, mullite,
 osumilite, sanidine, sapphirine, silicate glass, sillimanite, and tridymite, with mineral
 assemblages typical of pyroxene hornfels, sanidinite, and ultrahigh temperature

- metamorphism. Community 1 is well separated from most minerals of Communities 2
- through 6, which represent higher pressure and/or lower temperature environments.
- 730 Note that within the adjacent Communities 2 and 6, the higher-temperature minerals
- appear closest to Community 1 in Figure 1. Thus, there is a temperature gradient from
- 732 upper left to lower right in the network.
- 733 <u>Community 2:</u> Community 2 features 11 minerals, including andradite, anorthite, diopside,
- 734 grossular, hedenbergite, vesuvianite, and wollastonite, all of which are typical of mid- to
- 735 high-temperature contact and regional metamorphism of calc-silicates. Note that
- 736 Communities 2 and 5 display significant overlaps in this network, with a prominent
- temperature gradient from the top to the bottom of the network. Communities 2 and 5
- are thus relatively dispersed compared to Communities 1, 3, 4, and 6
- 739 <u>Community 3:</u> The eight minerals of Community 3—larnite, melilite, merwinite, monticellite,
- 740 *perovskite, rankinite, spurrite,* and *tilleyite*—form a tightly clustered group of idiosyncratic

phases typical of the highest temperature Ca-rich skarns. A number of petrologists have

recognized sequences of metamorphic skarn minerals as a function of temperature and

composition (Bowen 1940; Harker 1950; Tilley 1951). For example, Bowen (1940) listed

- 10 minerals in order of their increased temperature of formation: *tremolite, forsterite,*
- 745 diopside, periclase, wollastonite, monticellite, åkermanite (e.g., melilite), spurrite,
- 746 *merwinite*, and *larnite*, whereas subsequent studies slightly rearranged the order of some
- of these phases (e.g., Weeks 1956; Ferry 1976). We see suggestions of such a sequence,
- 748 with the highest temperature phases in Community 2 located in the upper right of Figure
- 749

1.

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Community 4: Thirteen minerals, including Mg-bearing brucite, cummingtonite, dolomite,
 forsterite, geikielite, humite, periclase, phlogopite, serpentine, spinel, and tremolite,
 represent contact metamorphic environments of Mg-rich skarns, as well as regional
 metamorphism of Mg-Si-silicates. Mineral assemblages of Community 4 thus have close
 links to the higher temperature, more calcic environments of Communities 2 and 3.

755 <u>Community 5:</u> Community 5 incorporates 11 metamorphic minerals, including phases from

two distinct environments. On the one hand, lower-temperature regional metamorphic

757 lithologies are represented by *chlorite, prehnite,* and *pumpellyite,* which are characteristic

of the prehnite-pumpelleyite and greenschist facies. Community 5 also includes a suite of

759 minerals typical of ophiolites and metamorphosed ultramafic rocks, including *chlorite*,

*chromite, magnesite, pentlandite,* and *talc.* Consequently, Community 5 is relatively
 dispersed and co-mingled with the higher temperature phases of Community 2.

Community 6: Diagnostic high-pressure metamorphic phases occur in Community 6. This
 well-defined cluster of 15 minerals in the lower lefthand region of Figure 1 holds typical
 high-pressure phases of blueschist (glaucophane, lawsonite), eclogite (jadeite, kyanite,
 omphacite), and ultrahigh pressure (coesite, diamond, pyrope) facies. These minerals are
 all characteristic of metamorphism under the relatively low geothermal gradients

experienced by deeply subducted crustal wedges that buoyantly rebound.

768

The degree of connectivity, or network density, of the graph in Figure 1 varies significantly with the percentage of co-occurrence, *P*. The density of a network (*D*) is defined as the fraction of all possible links that are observed; in the case of 94 minerals, there exist  $[(94^2 - 94)/2] =$
772 4371 possible links. When P = 1 %, the density of the network has a value of D = 0.400 because 773 1747 (40.0 %) of the 4371 possible links between mineral pairs are observed to occur at least 774 once. By contrast, when we restrict links to P = 25 % then 524 links remain (D = 0.120). And 775 when we consider P = 50 %, only 198 links persist – a relatively sparse network with D = 0.045. 776 Note that at P > 6 %, one or more mineral nodes is no longer connected to the network. The 777 first node to disconnect is *cummingtonite* at P = 7 %, while at P = 15 % *olivine* also becomes 778 disconnected. At P = 25 %, 66 of the original 73 mineral nodes remain interconnected, including 779 representatives of all 6 communities. However, at P = 50 %, only 39 nodes form a sparse 780 network with all 13 minerals of Community 4 (Mg skarns) forming a hub with radiating suites of 781 minerals from Community 2 (7 minerals), Community 3 (6 minerals), and Community 5 (10 782 minerals). By contrast, all but 2 minerals from Community 6 and 1 mineral from Community 1 783 remain connected at P = 50 %.

An important feature of the unipartite networks of metamorphic minerals is that every metamorphic rock, for example each of the 2785 examples in Supplementary Table 3, as well as every prograde or retrograde sequence of metamorphic rocks, is embedded as a multi-node subgraph of this network (Figure 1B). *P-T-X* series of lithologies can be represented by a sequence of subgraphs that wend their way across the larger network of Figure 1B. Thus, Figure 1 and related networks are useful visual approaches to comparing and contrasting aspects of metamorphic petrology for research and education.

In many respects, the topology of Figure 1 for common metamorphic minerals is reminiscent of the topology of the analogous network of 115 igneous minerals in Part VII of this series (Hazen et al. 2023; their Figure 1). In both instances, Louvain community detection reveals

794 several communities of mineral kinds, each from a distinct compositional and/or environmental 795 regime. Furthermore, every rock in extensive lists of mineral modes is represented by a 796 subgraph (compare Figure 1B to Hazen et al. 2023, their Figure 1B). Nevertheless, two 797 important differences exist between these two renderings.

- In the case of igneous rocks, the majority of the minerals tabulated by Hazen et al. (2023)
  are minor accessory phases (< 5 vol %), most of which incorporate relatively rare</li>
  elements. By contrast, reported modes of metamorphic rocks rarely include minor
  phases. Consequently, the network for metamorphic minerals illustrates fewer mineral
  kinds, but almost twice as many major phases spanning a much wider *P-T* range,
  compared to the network for igneous mineral kinds.
- 804 2. In our study of the evolution of igneous minerals, we attempted to include only modes 805 based on equilibrium assemblages of primary minerals. Accordingly, we have suggested 806 that phase equilibrium for multi-component chemical systems (including myriad rare 807 elements) might be extracted from the topology of the igneous mineral network. 808 However, owing to often incomplete transformations during prograde or retrograde 809 metamorphism, the assumption of equilibrum assemblages cannot be applied to 810 metamorphic rocks. On the other hand, because most metamorphic rock modes are 811 embedded in Figure 1, it should be possible to illustrate any metamorphic sequence in P-812 T-X space with an animated series that systematically moves across Figure 1, even if the 813 individual mineral assemblages are not in equilibrium.

814

815 Unipartite network graph at P = 33 %: The unipartite graph of Figure 1 represents the co-

816 occurrences of 73 different metamorphic minerals based on tabulations of 2785 modes at P = 6

- 817 %, which is the maximum value at which all minerals are still connected. However, Figure 1 may
- 818 be visually misleading in three ways.
- 819 First, Figure 1 is a two-dimensional projection of a graph in 72 dimensions [any network with

N nodes possesses (N - 1) dimensions]. Consequently, the proximity of two adjacent nodes

- does not necessarily imply that those two minerals are closely related. Thus, for example,
- 822 forsterite (Community 4) and prehnite (Community 5) lie next to each other in Figure 1, yet
- 823 those two minerals do not coexist in any rock in our survey.
- 824 Second, in some instances a mineral will be located in a position somewhat removed from its

825 community. For example, the Mg-amphibole *anthophyllite* is included in Community 1, yet it

appears near the center of Figure 1, closer to Communities 2, 4, and 5. This positioning results

827 from anthophyllite's close association with both cordierite (Community 1) and forsterite

- 828 (Community 4)—a common situation when one node is closely associated with two or more
- 829 communities.

830 Third, because Figure 1 represents all links between nodes with at least 6 % co-occurrence,

some relatively weak connections (i.e., those with only 6 % to 10 % co-occurrence) will tend to

intermingle some phases that only co-exist relatively infrequently with other minerals in two or

833 more communities.

Figure 2 is a unipartite network similar to that of Figure 1 but with P = 33 %. Therefore, a link appears only if at least one-third of occurrences of the rarer mineral also features the more common mineral. The result is a much sparser network of 66 mineral nodes and 185 links.

- 837 Seven minerals-aegirine, anorthite, cummingtonite, fayalite, hedenbergite, olivine, and
- 838 staurolite—are no longer connected. The resulting graph retains the six communities of Figure
- 839 1, but they are much more tightly clustered, revealing well-separated groups of minerals.



841 Figure 2. A unipartite network of 66 common metamorphic minerals (colored circles). Node sizes 842 indicate the relative abundances of minerals, while colors indicate six communities of metamorphic 843 minerals that were determined using Louvain community detection (see text). Each of these 844 communities corresponds to a different temperature-pressure-composition regime. In this figure, links 845 are drawn between two minerals if at least 33 % of rocks that incorporate the less common mineral also 846 incorporate the more common mineral (as tabulated in Supplementary Table 3). One can vary this 847 percentage in an interactive version of this at: graph 848 (https://observablehq.com/@anirudhprabhu/revised-evolutionary-system-of-mineralogy-part-8-uni). Hover your cursor over any node to identify the corresponding mineral; click and hold your cursor to 849 850 move that node and identify links to other nodes; use your cursor to move the "Weight Threshold"

851 vernier to systematically eliminate links between nodes based on P values (see text). Adjust node

852 attributes by clicking on the "Size Nodes By" feature.

840

853 Figure 2 displays the same six communities (with 66 of the 73 minerals) that appear in Figure 854 1. However, the more stringent criteria of P = 33 % results in a network with far greater 855 separation between communities. Community 1 now appears at the extreme right of the graph, 856 retaining 14 of the original 15 minerals (only staurolite has been lost). The only link between 857 Community 1 and the rest of the network is to Community 4 through *spinel*. 858 Community 4 has 12 of the original 13 minerals (*cummingtonite* disconnects at P = 7 %), 859 which form a cluster in the upper right of the network. Note that *tremolite* is displaced from the 860 main cluster because it forms only three links—to calcite (also in Community 4), diopside 861 (Community 2), and *pentlandite* (Community 5). Community 4 is most closely linked to 862 Communities 2 and 5, but has only 3 links to Community 3, 1 link to Community 1, and no 863 connections to Community 6.

Community 3, which represents relatively high-temperature skarn minerals, retains all of its original 8 phases in an isolated cluster that is linked only to the lower-temperature skarn minerals of Communities 2 and 4. By contrast, Community 2 is the most intermingled cluster in this network. Only 8 of the original 11 minerals remain; *anorthite, fayalite,* and *hedenbergite* are disconnected by P = 25 %.

All 11 of the original Community 5 minerals appear in Figure 2, which reveals a well-defined cluster with three subgraphs. *Chlorite-prehnite-pumpelleyite* appear as a triangle corresponding to low-temperature metamorphism, while *chromite-magnesite-pentlandite-talc* form a quartet of ophiolite phases. In addition, *allanite, Fe-dolomite, graphite,* and *tourmaline* are commonly associated accessory minerals in regional metamorphic rocks. Community 6, with 13 of the original 15 high-pressure minerals (minus *aegirine* and *olivine*), is the most isolated cluster in

Figure 2, with only a single link between *lawsonite* of Community 6 and *chlorite* of Community

876 5.

The resulting sparse network forms an arc, with the highest pressure minerals to the left and the lowest pressure minerals to the right. The central grouping of Communities 2, 3, and 4 represent metamorphosed Ca-Mg silicates and skarns, with higher temperature phases located toward the bottom of the graph. Thus, in spite of the continuous variations of temperature, pressure, and composition represented by metamorphic mineral assemblages, distinct clusters representing formational environments with idiosyncratic minerals are important characteristics of metamorphic rocks.

884 An unexpected result of this network analysis are the strong separtions among some mineral 885 communities. By their nature, metamorphic rocks represent continua of P-T-X conditions. One 886 might expect, therefore, to observe similarly continuous linkages in the network topology of 887 Figure 2. However, we observe strong clustering into six communities with signifcant gaps. 888 Most notably, Community 1 on the right and Community 6 on the left are isolated from 889 Communities 2 through 5. In addition, we observe lesser gaps between Communities 2 and 5, 890 as well as between Communities 3 and 4. Similar topologies in network graphs of igneous rocks 891 correspond to known compositional trends, such as the Daly gap (Daly 1925; Hazen et al. 2023, 892 their Figure 5). Is it possible that similar composition gaps exist among the diversity of 893 metamorphic rocks?

894

895 <u>Bipartite network of metamorphic minerals and their host lithologies</u>: Figure 3 is a bipartite 896 network (i.e., a network with links between two different kinds of nodes) that illustrates 1220

- 897 diamond-shaped nodes representing metamorphic mineral species and their 1686 links to 8 898 nodes that represent the different groups of metamorphic rocks, as first described by Hazen 899 and Morrison (2022) and modified here in Supplementary Table 1. This bipartite network was 900 made using the "visNetwork" (Almende et al. 2021) and "igraph" (Csardi & Nepusz 2006) R 901 packages. The code for construction of this network can be found 902 at: https://github.com/anirudhprabhu/StellarNet/tree/master/PartVIII. The network layout
- 903 uses the "barnesHut" approximation algorithm (Barnes and Hut 1986).



- 904
- Figure 3. This bipartite network of 1220 metamorphic minerals (colored diamond-shaped nodes)
   displays 1686 links to 8 icons representing different major groups of metamorphic rocks, as first
- 907 described by Hazen and Morrison (2022) and modified here (Table 2; Supplementary Table 1).
   908 Three-letter identifiers are XEN = pyrometamorphism of xenoliths (with 173 minerals); CON =

909contact metamorphism (424); BAM = metamorphic Ba/Mn/Pb/Zn deposits (449); OPH =910ophiolites (109); HPM = high-pressure metamorphism (113); REG = regional metamorphism911(351); MET = mantle metasomartism (37); and SHE = shear-induced minerals (30). The 6912"starburst" features around the periphery reflect 959 mineral species that are uniquely913associated with only one paragenetic mode. Most of the other nodes, representing 261 mineral914species that are connected to two or more nodes representing paragenetic modes, lie in the915dense central region of the bipartite network.

916

917 This bipartite network displays several features that are common to other mineral systems 918 (Morrison et al. 2017, 2020, 2022; Hazen et al. 2019). A minority of 261 relatively common 919 metamorphic mineral species adopt central positions, where each is linked to two or more 920 different groups of metamorphic minerals. Only 1 mineral species, magnetite, is linked to all 8 921 groups of metamorphic rocks. Six additional common minerals—calcite, diopside, ilmenite, 922 *pyrite, rutile,* and *quartz*—are linked to 7 groups. These minerals from multiple metamorphic 923 lithologies are less diagnostic in defining communities or clusters of closely-related minerals 924 than minerals known from only one or two types of metamorphic rocks.

925 By contrast, a significant majority of mineral species are rare; 959 of these minerals are 926 linked to a single metamorphic mineral group, thus creating six dramatic "starbursts" of nodes 927 decorating the periphery of the bipartite network. Metamorphosed Ba-Fe-Mn-Pb deposits 928 (node BAM) boast the greatest number of these single-node phases (375 species, or 31 % of the 929 1220 metamorphic minerals), which create the largest such display in the lower right of the 930 network. Note that this concentration of relatively rare minerals, like the BAM node with which 931 it is associated, forms a relatively isolated region of the bipartite network. Other prominent 932 starbursts are connected to contact metamorphism (CON; 241 unique mineral species), 933 regional metamorphism (REG; 151 unique species), xenoliths (XEN; 87 unique species), and

934 ophiolites (OPH; 69 unique species). Thus, as in many other mineral systems, most 935 metamorphic minerals are rare, known from 5 or fewer localities and formed by a single 936 process (Hazen and Ausubel 2016).

937 In addition, 160 minerals are associated with exactly two of the 8 metamorphic rock groups, 938 32 minerals are linked to 3 groups, and 30 minerals are linked to 4 groups. The most common 939 connection from one mineral species to two metamorphic rock groups is for contact and 940 regional metamorphic rocks (135 minerals), which explains the proximity of CON and REG 941 nodes in the upper half of Figure 3. Other strong connections of a mineral to two groups of 942 metamorphic rocks include regional and high-pressure metamorphic rocks (71 species); 943 xenolith and contact metamorphic rocks (67 species); xenolith and regional metamorphic rocks 944 (64 species); contact and high-pressure metamorphic rocks (55 species); regional and Ba-Mn 945 deposits (47 species), and contact and Ba-Mn deposits (41 species). These varied mineral 946 associations across several kinds of metamorphic lithologies are reflected in the distinctive 947 topology of Figure 3.

949

## 4. IMPLICATIONS: THE EVOLUTION OF METAMORPHIC MINERALS

950	Metamorphic	rocks display characteristics of an evolvin	g chemical syste	m, with significant
951	increases in dive	ersity and average chemical complexity ov	er 4.5 billion yea	ars of Earth history
952	(Krivovichev et a	al. 2018). Tabulations of the earliest know	n ages for diagn	ostic metamorphic
953	minerals have th	e potential to document these changes. The	e Mineral Evoluti	on Database (MED:
954	https://rruff.infc	v/evolution, accessed 11 September 202	3) records the	ages of 200,000
955	mineral/locality	pairs. While ages of metamorphic minerals	were not the pr	incipal focus of the
956	MED, most majo	or metamorphic minerals are represented by	/ multiple ages. A	Accordingly, Table 4
957	lists the oldest N	1ED ages for 47 minerals that are most com	monly found in m	netamorphic rocks.
958 959 960 961 962	Table 4. Oldes Ages are fro	t recorded ages of 47 select metamorphic n m the Mineral Evolution Database ( <u>https://r</u> September 2023) unless otherwis	ninerals arranged ruff.info/evolutic se noted.	l chronologically. on, accessed 11
/04				
963	Mineral Kind	Chemical Formula A	ge Range (Ma) <sup>1</sup>	Community <sup>2</sup>
963 964	Mineral Kind Geikielite	Chemical FormulaAMgTiO3	Age Range (Ma) <sup>1</sup> 4330—4310	Community <sup>2</sup> 4
963 964 965	<u>Mineral Kind</u> Geikielite Diopside	Chemical FormulaAMgTiO3CaMgSi2O6	Age Range (Ma) <sup>1</sup> 4330—4310 4330—4010	Community <sup>2</sup> 4 2
963 964 965 966	<u>Mineral Kind</u> Geikielite Diopside Hedenbergite	Chemical FormulaAMgTiO3 $CaMgSi_2O_6$ CaFe $^{2+}Si_2O_6$	Age Range (Ma) <sup>1</sup> 4330—4310 4330—4010 4330—4010	Community <sup>2</sup> 4 2 2
963 964 965 966 967	Mineral Kind Geikielite Diopside Hedenbergite Wollastonite	Chemical Formula $A$ MgTiO3CaMgSi2O6CaFe2+Si2O6CaSiO3	Age Range (Ma) <sup>1</sup> 4330—4310 4330—4010 4330—4010 4330—4010 4330—4010	Community <sup>2</sup> 4 2 2 2 2
963 964 965 966 967 968	Mineral Kind Geikielite Diopside Hedenbergite Wollastonite Andalusite	$\begin{tabular}{ c c c c } \hline Chemical Formula & A \\ MgTiO_3 & & \\ CaMgSi_2O_6 & & \\ CaFe^{2^+}Si_2O_6 & & \\ CaSiO_3 & & \\ Al_2SiO_5 & & \\ \hline \end{tabular}$	Age Range (Ma) <sup>1</sup> 4330—4310 4330—4010 4330—4010 4330—4010 4000—3200	Community <sup>2</sup> 4 2 2 2 2 1
963 964 965 966 967 968 969	Mineral Kind Geikielite Diopside Hedenbergite Wollastonite Andalusite Anthophyllite	Chemical FormulaAMgTiO3 $CaMgSi_2O_6$ CaFe <sup>2+</sup> Si_2O_6 $CaSiO_3$ Al_2SiO_5 $\BoxMg_2Mg_5Si_8O_{22}(OH)_2$	Age Range (Ma) <sup>1</sup> 4330—4310 4330—4010 4330—4010 4330—4010 4000—3200 4000—3200	Community <sup>2</sup> 4 2 2 2 1 1 1
963 964 965 966 967 968 969 970	Mineral Kind Geikielite Diopside Hedenbergite Wollastonite Andalusite Anthophyllite Cordierite	$\begin{tabular}{ c c c c } \hline Chemical Formula & \begin{tabular}{lllllllllllllllllllllllllllllllllll$	Age Range (Ma) <sup>1</sup> 4330-4310 4330-4010 4330-4010 4330-4010 4000-3200 4000-3200 4000-3200	Community <sup>2</sup> 4 2 2 2 1 1 1 1
963 964 965 966 967 968 969 969 970 971	Mineral Kind Geikielite Diopside Hedenbergite Wollastonite Andalusite Anthophyllite Cordierite Tremolite	$\begin{tabular}{ c c c c } \hline Chemical Formula & A \\ MgTiO_3 & & \\ CaMgSi_2O_6 & & \\ CaFe^{2+}Si_2O_6 & & \\ CaSiO_3 & & \\ Al_2SiO_5 & & \\ \Box Mg_2Mg_5Si_8O_{22}(OH)_2 & & \\ (Mg,Fe^{2+})_2Al_4Si_5O_{18} & & \\ \Box Ca_2(Mg_{5,0-4,5}Fe^{2+}_{0,0-0,5})Si_8O_{22}(OH)_2 & & \\ \hline \end{tabular}$	Age Range (Ma) <sup>1</sup> 4330-4310 4330-4010 4330-4010 4330-4010 4000-3200 4000-3200 4000-3200 4000-3200	Community <sup>2</sup> 4 2 2 2 1 1 1 1 4
963 964 965 966 967 968 969 970 971 972	Mineral Kind Geikielite Diopside Hedenbergite Wollastonite Andalusite Anthophyllite Cordierite Tremolite Chloritoid	$\begin{tabular}{ c c c c } \hline Chemical Formula & A \\ MgTiO_3 & & & \\ CaMgSi_2O_6 & & & \\ CaFe^{2^+}Si_2O_6 & & & \\ CaSiO_3 & & & \\ Al_2SiO_5 & & & \\ \Box Mg_2Mg_5Si_8O_{22}(OH)_2 & & & \\ (Mg,Fe^{2^+})_2Al_4Si_5O_{18} & & \\ \Box Ca_2(Mg_{5.0^{-4.5}}Fe^{2^+}_{0.0^{-0.5}})Si_8O_{22}(OH)_2 & & \\ Fe^{2^+}Al_2O(SiO_4)(OH)_2 & & \\ \hline \end{tabular}$	Age Range (Ma) <sup>1</sup> 4330-4310 4330-4010 4330-4010 4330-4010 4000-3200 4000-3200 4000-3200 4000-3200 4000-3200 4000-3200 4000-3040	Community <sup>2</sup> 4 2 2 2 1 1 1 1 4 6
963 964 965 966 967 968 969 970 971 972 973	Mineral Kind Geikielite Diopside Hedenbergite Wollastonite Andalusite Anthophyllite Cordierite Tremolite Chloritoid Grunerite	$\begin{tabular}{ c c c c } \hline Chemical Formula & A \\ MgTiO_3 & & & \\ CaMgSi_2O_6 & & & \\ CaFe^{2+}Si_2O_6 & & & \\ CaSiO_3 & & & \\ Al_2SiO_5 & & & \\ \Box Mg_2Mg_5Si_8O_{22}(OH)_2 & & & \\ (Mg,Fe^{2+})_2Al_4Si_5O_{18} & & \\ \Box Ca_2(Mg_{5.0-4.5}Fe^{2+}_{0.0-0.5})Si_8O_{22}(OH)_2 & & \\ Fe^{2+}Al_2O(SiO_4)(OH)_2 & & \\ \Box Fe^{2+}_2Fe^{2+}_5Si_8O_{22}(OH)_2 & & \\ \hline \end{tabular}$	Age Range (Ma) <sup>1</sup> 4330-4310 4330-4010 4330-4010 4330-4010 4000-3200 4000-3200 4000-3200 4000-3200 4000-3200 4000-3040 4000-2920	Community <sup>2</sup> 4 2 2 2 1 1 1 4 6
963 964 965 966 967 968 969 970 971 972 973 974	Mineral Kind Geikielite Diopside Hedenbergite Wollastonite Andalusite Anthophyllite Cordierite Tremolite Chloritoid Grunerite Cummingtonite	$\begin{tabular}{ c c c c } \hline Chemical Formula & A \\ MgTiO_3 & & \\ CaMgSi_2O_6 & & \\ CaFe^{2+}Si_2O_6 & & \\ CaSiO_3 & & \\ Al_2SiO_5 & & \\ \Box Mg_2Mg_5Si_8O_{22}(OH)_2 & & \\ (Mg,Fe^{2+})_2Al_4Si_5O_{18} & & \\ \Box Ca_2(Mg_{5.0-4.5}Fe^{2+}_{0.0-0.5})Si_8O_{22}(OH)_2 & & \\ Fe^{2+}Al_2O(SiO_4)(OH)_2 & & \\ \Box Fe^{2+}_2Fe^{2+}_5Si_8O_{22}(OH)_2 & & \\ e \ \Box Mg_2Mg_5Si_8O_{22}(OH)_2 & & \\ \hline \end{tabular}$	Age Range (Ma) <sup>1</sup> 4330-4310         4330-4010         4330-4010         4330-4010         4330-4010         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200	Community <sup>2</sup> 4 2 2 2 1 1 1 4 6 4
963 964 965 966 967 968 969 970 971 972 973 974 975 976	Mineral Kind Geikielite Diopside Hedenbergite Wollastonite Andalusite Anthophyllite Cordierite Tremolite Chloritoid Grunerite Cummingtonite Kyanite	$\begin{tabular}{ c c c c } \hline Chemical Formula & A \\ MgTiO_3 & & & \\ CaMgSi_2O_6 & & & \\ CaFe^{2+}Si_2O_6 & & & \\ CaSiO_3 & & & \\ Al_2SiO_5 & & & \\ \Box Mg_2Mg_5Si_8O_{22}(OH)_2 & & & \\ (Mg,Fe^{2+})_2Al_4Si_5O_{18} & & \\ \Box Ca_2(Mg_{5.0\cdot4.5}Fe^{2+}_{0.0\cdot0.5})Si_8O_{22}(OH)_2 & & \\ Fe^{2+}Al_2O(SiO_4)(OH)_2 & & \\ \Box Fe^{2+}_2Fe^{2+}_5Si_8O_{22}(OH)_2 & & \\ e \ \Box Mg_2Mg_5Si_8O_{22}(OH)_2 & & \\ Al_2SiO_5 & & \\ \hline \end{tabular}$	Age Range (Ma) <sup>1</sup> 4330-4310         4330-4010         4330-4010         4330-4010         4330-4010         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-2917         4000-2917         4000-2714	Community <sup>2</sup> 4 2 2 2 1 1 1 1 4 6 4 6 6
963 964 965 966 967 968 969 970 971 972 973 974 975 976 977	Mineral Kind Geikielite Diopside Hedenbergite Wollastonite Andalusite Anthophyllite Cordierite Tremolite Chloritoid Grunerite Cummingtonite Kyanite Sillimanite	$\begin{tabular}{ c c c c } \hline Chemical Formula & A \\ MgTiO_3 & & \\ CaMgSi_2O_6 & & \\ CaFe^{2+}Si_2O_6 & & \\ CaSiO_3 & & \\ Al_2SiO_5 & & \\ \Box Mg_2Mg_5Si_8O_{22}(OH)_2 & & \\ (Mg,Fe^{2+})_2Al_4Si_5O_{18} & & \\ \Box Ca_2(Mg_{5.0\cdot4.5}Fe^{2+}_{0.0\cdot0.5})Si_8O_{22}(OH)_2 & \\ Fe^{2+}Al_2O(SiO_4)(OH)_2 & & \\ \Box Fe^{2+}_2Fe^{2+}_5Si_8O_{22}(OH)_2 & & \\ e \ \Box Mg_2Mg_5Si_8O_{22}(OH)_2 & & \\ Al_2SiO_5 & & \\ Al_2SiO_5 & & \\ \hline \end{tabular}$	Age Range (Ma) <sup>1</sup> 4330-4310         4330-4010         4330-4010         4330-4010         4330-4010         4330-4010         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-2920         4000-2917         4000-2714         3640-3200	Community <sup>2</sup> 4 2 2 2 1 1 1 4 6 4 6 1 1
963 964 965 966 967 968 969 970 971 972 973 974 975 976 977 978	Mineral Kind Geikielite Diopside Hedenbergite Wollastonite Andalusite Anthophyllite Cordierite Tremolite Chloritoid Grunerite Cummingtonite Kyanite Sillimanite Graphite	$\begin{tabular}{ c c c c c } \hline Chemical Formula & A \\ MgTiO_3 & & \\ CaMgSi_2O_6 & & \\ CaFe^{2^+}Si_2O_6 & & \\ CaSiO_3 & & \\ Al_2SiO_5 & & \\ \Box Mg_2Mg_5Si_8O_{22}(OH)_2 & & \\ (Mg,Fe^{2^+})_2Al_4Si_5O_{18} & & \\ \Box Ca_2(Mg_{5.0^{-4.5}}Fe^{2^+}_{0.0^{-0.5}})Si_8O_{22}(OH)_2 & \\ Fe^{2^+}Al_2O(SiO_4)(OH)_2 & & \\ \Box Fe^{2^+}_2Fe^{2^+}_5Si_8O_{22}(OH)_2 & & \\ e \ \Box Mg_2Mg_5Si_8O_{22}(OH)_2 & & \\ Al_2SiO_5 & & \\ Al_2SiO_5 & & \\ C & & \\ \hline \end{tabular}$	Age Range (Ma) <sup>1</sup> 4330-4310         4330-4010         4330-4010         4330-4010         4330-4010         4330-4010         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-3200         4000-2920         4000-2917         4000-2714         3640-3200         3640-3200	Community <sup>2</sup> 4 2 2 2 1 1 1 1 4 6 4 6 1 5
963 964 965 966 967 968 969 970 971 972 973 974 975 976 977 978 979	Mineral Kind Geikielite Diopside Hedenbergite Wollastonite Andalusite Anthophyllite Cordierite Tremolite Chloritoid Grunerite Cummingtonite Kyanite Sillimanite Graphite Corundum	Chemical Formula       A         MgTiO <sub>3</sub> CaMgSi <sub>2</sub> O <sub>6</sub> CaFe <sup>2+</sup> Si <sub>2</sub> O <sub>6</sub> CaSiO <sub>3</sub> Al <sub>2</sub> SiO <sub>5</sub> $\square$ Mg <sub>2</sub> Mg <sub>5</sub> Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub> $\square$ Mg,Fe <sup>2+</sup> ) <sub>2</sub> Al <sub>4</sub> Si <sub>5</sub> O <sub>18</sub> $\square$ Ca <sub>2</sub> (Mg <sub>5.0-4.5</sub> Fe <sup>2+</sup> <sub>0.0-0.5</sub> )Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub> Fe <sup>2+</sup> Al <sub>2</sub> O(SiO <sub>4</sub> )(OH) <sub>2</sub> $\square$ Fe <sup>2+</sup> <sub>2</sub> Fe <sup>2+</sup> <sub>5</sub> Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub> e $\square$ Mg <sub>2</sub> Mg <sub>5</sub> Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub> $Al_2$ SiO <sub>5</sub> Al <sub>2</sub> SiO <sub>5</sub> $C$ Al <sub>2</sub> SiO <sub>5</sub> $Al_2$ O <sub>3</sub>	Age Range (Ma) <sup>1</sup> 4330-4310           4330-4010           4330-4010           4330-4010           4330-4010           4330-4010           4000-3200           4000-3200           4000-3200           4000-3200           4000-3200           4000-3200           4000-3200           4000-2920           4000-2917           4000-2917           4000-2917           4000-2917           4000-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3040	Community <sup>2</sup> 4 2 2 2 1 1 1 1 4 6 4 6 1 5 1
963 964 965 966 967 968 969 970 971 972 973 974 975 976 977 978 979 980	Mineral Kind Geikielite Diopside Hedenbergite Wollastonite Andalusite Anthophyllite Cordierite Tremolite Chloritoid Grunerite Cummingtonite Kyanite Sillimanite Graphite Corundum Diamond <sup>3</sup>	$\begin{tabular}{ c c c c c } \hline Chemical Formula & A \\ MgTiO_3 & & \\ CaMgSi_2O_6 & & \\ CaFe^{2+}Si_2O_6 & & \\ CaSiO_3 & & \\ Al_2SiO_5 & & \\ \Box Mg_2Mg_5Si_8O_{22}(OH)_2 & & \\ (Mg,Fe^{2+})_2Al_4Si_5O_{18} & & \\ \Box Ca_2(Mg_{5.0-4.5}Fe^{2+}_{0.0-0.5})Si_8O_{22}(OH)_2 & \\ Fe^{2+}Al_2O(SiO_4)(OH)_2 & & \\ \Box Fe^{2+}_2Fe^{2+}_5Si_8O_{22}(OH)_2 & & \\ e \ \Box Mg_2Mg_5Si_8O_{22}(OH)_2 & & \\ Al_2SiO_5 & & \\ C & & \\ Al_2O_3 & & \\ C & & \\ \hline \end{tabular}$	Age Range (Ma) <sup>1</sup> 4330-4310           4330-4010           4330-4010           4330-4010           4330-4010           4330-4010           4330-4010           4000-3200           4000-3200           4000-3200           4000-3200           4000-3200           4000-3200           4000-2917           4000-2917           4000-2917           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3040           3600-3600	Community <sup>2</sup> 4 2 2 2 1 1 1 1 4 6 4 6 4 6 1 5 1 6
963 964 965 966 967 968 969 970 971 972 973 974 975 976 977 978 979 980 981	Mineral Kind Geikielite Diopside Hedenbergite Wollastonite Andalusite Anthophyllite Cordierite Tremolite Chloritoid Grunerite Cummingtonite Kyanite Sillimanite Graphite Corundum Diamond <sup>3</sup> Spessartine	$\begin{tabular}{ c c c c } \hline Chemical Formula & A \\ MgTiO_3 & & \\ CaMgSi_2O_6 & & \\ CaFe^{2+}Si_2O_6 & & \\ CaSiO_3 & & \\ Al_2SiO_5 & & \\ \Box Mg_2Mg_5Si_8O_{22}(OH)_2 & & \\ (Mg,Fe^{2+})_2Al_4Si_5O_{18} & & \\ \Box Ca_2(Mg_{5.0-4.5}Fe^{2+}_{0.0-0.5})Si_8O_{22}(OH)_2 & & \\ Fe^{2+}Al_2O(SiO_4)(OH)_2 & & \\ \Box Fe^{2+}_2Fe^{2+}_5Si_8O_{22}(OH)_2 & & \\ e & \Box Mg_2Mg_5Si_8O_{22}(OH)_2 & & \\ Al_2SiO_5 & & \\ Al_2SiO_5 & & \\ Al_2O_3 & & \\ C & & \\ Mn^{2+}_3Al_2Si_3O_{12} & & \\ \end{tabular}$	Age Range (Ma) <sup>1</sup> 4330-4310           4330-4010           4330-4010           4330-4010           4330-4010           4330-4010           4330-4010           4000-3200           4000-3200           4000-3200           4000-3200           4000-3200           4000-3200           4000-2920           4000-2917           4000-2917           4000-2714           3640-3200           3600-3600	Community <sup>2</sup> 4 2 2 2 1 1 1 1 4 6 4 6 1 5 1 6 1 5 1 6
963 964 965 966 967 968 969 970 971 972 973 974 975 976 977 978 977 978 979 980 981 982 983	Mineral Kind Geikielite Diopside Hedenbergite Wollastonite Andalusite Anthophyllite Cordierite Tremolite Chloritoid Grunerite Cummingtonite Kyanite Sillimanite Graphite Corundum Diamond <sup>3</sup> Spessartine Phlogopite	Chemical Formula         A           MgTiO <sub>3</sub> CaMgSi <sub>2</sub> O <sub>6</sub> CaFe <sup>2+</sup> Si <sub>2</sub> O <sub>6</sub> CaSiO <sub>3</sub> Al <sub>2</sub> SiO <sub>5</sub> $\Box$ Mg <sub>2</sub> Mg <sub>5</sub> Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub> $\Box$ Mg <sub>2</sub> Mg <sub>5</sub> Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub> $(Mg, Fe^{2+})_2Al_4Si_5O_{18}$ $\Box$ Ca <sub>2</sub> (Mg <sub>5.0-4.5</sub> Fe <sup>2+</sup> 0.0-0.5)Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub> $Fe^{2+}Al_2O(SiO_4)(OH)_2$ $Fe^{2+}Al_2O(SiO_4)(OH)_2$ $\Box$ Fe <sup>2+</sup> $_2Fe^{2+}_5Si_8O_{22}(OH)_2$ $e$ $\Box$ Mg <sub>2</sub> Mg <sub>5</sub> Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub> $Al_2SiO_5$ $Al_2SiO_5$ $Al_2SiO_5$ $Al_2O_3$ $C$ $Mn^{2+}_3Al_2Si_3O_{12}$ $[[KMg_2(Mg, Fe^{2+})(Si, Al)_2Si_2O_{10}(OH, F)_2]]$	Age Range (Ma) <sup>1</sup> 4330-4310           4330-4010           4330-4010           4330-4010           4330-4010           4330-4010           4330-4010           4000-3200           4000-3200           4000-3200           4000-3200           4000-3200           4000-2920           4000-2917           4000-2917           4000-2917           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3600-3600           3293-2874           3200-3200	Community2         4         2         2         2         1         1         4         6            4         6         1         5         1         6            4         6            4
963 964 965 966 967 968 969 970 971 972 973 974 975 976 977 978 977 978 979 980 981 982 983 984	Mineral Kind Geikielite Diopside Hedenbergite Wollastonite Andalusite Anthophyllite Cordierite Tremolite Chloritoid Grunerite Cummingtonite Kyanite Sillimanite Graphite Corundum Diamond <sup>3</sup> Spessartine Phlogopite	Chemical Formula         A           MgTiO <sub>3</sub> CaMgSi <sub>2</sub> O <sub>6</sub> CaFe <sup>2+</sup> Si <sub>2</sub> O <sub>6</sub> CaSiO <sub>3</sub> Al <sub>2</sub> SiO <sub>5</sub> $\Box$ Mg <sub>2</sub> Mg <sub>5</sub> Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub> $\Box$ Mg <sub>2</sub> Mg <sub>5</sub> Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub> $\Box$ Mg <sub>2</sub> Mg <sub>5</sub> Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub> $\Box$ Mg <sub>2</sub> Mg <sub>5</sub> Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub> $\Box$ $\Box$ Ca <sub>2</sub> (Mg <sub>5.0-4.5</sub> Fe <sup>2+</sup> 0.0-0.5)Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub> $Fe^{2+}$ Al <sub>2</sub> O(SiO <sub>4</sub> )(OH) <sub>2</sub> $\Box$ Pe <sup>2+</sup> Al <sub>2</sub> O(SiO <sub>4</sub> )(OH) <sub>2</sub> $\Box$ Fe <sup>2+</sup> Al <sub>2</sub> O(SiO <sub>4</sub> )(OH) <sub>2</sub> $\Box$ Fe <sup>2+</sup> <sub>2</sub> Fe <sup>2+</sup> 5Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub> $\Box$ Mg <sub>2</sub> Mg <sub>5</sub> Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub> $\Box$ Mg <sub>2</sub> Mg <sub>5</sub> Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub> $A$ l <sub>2</sub> SiO <sub>5</sub> $\Box$ $A$ l <sub>2</sub> SiO <sub>5</sub> $\Box$	Age Range (Ma) <sup>1</sup> 4330-4310           4330-4010           4330-4010           4330-4010           4330-4010           4330-4010           4330-4010           4000-3200           4000-3200           4000-3200           4000-3200           4000-3200           4000-2920           4000-2917           4000-2917           4000-2917           4000-2917           4000-2917           4000-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3640-3200           3693-2874           3200-3200           2969-2969	Community <sup>2</sup> 4 2 2 2 1 1 1 1 4 6 4 6 1 5 1 6 4 2

Staurolite	Fe <sup>2+</sup> <sub>2</sub> Al <sub>9</sub> Si <sub>4</sub> O <sub>23</sub> (OH)	2906—2730	1
Piemontite	Ca <sub>2</sub> Al <sub>2</sub> Mn <sup>3+</sup> (Si <sub>2</sub> O <sub>7</sub> )(SiO <sub>4</sub> )O(OH)	2906—2782	
Prehnite	$Ca_2AI(Si_3AI)O_{10}(OH)_2$	2900-2810	5
Scapolite	(Na,Ca) <sub>4</sub> (Al,Si) <sub>12</sub> O <sub>24</sub> (CO <sub>3</sub> ,SO <sub>4</sub> ,Cl)	2845—1930	2
Glaucophane	$\Box$ Na <sub>2</sub> (Mg <sub>3</sub> Al <sub>2</sub> )Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub>	2845—1700	6
Andradite	$Ca_{3}Fe^{3+}{}_{2}Si_{3}O_{12}$	2742—2670	2
Baddellyite	ZrO <sub>2</sub>	2742—2670	4
Brucite	Mg(OH) <sub>2</sub>	2742-2700	4
Pumpellyite	$Ca_2AI_3(Si_2O_7)(SiO_4)(OH,O)_2 \cdot H_2O$	2680—2677	5
Pyrope	$Mg_3Al_2Si_3O_{12}$	2710-2700	6
Actinolite	$\Box$ Ca <sub>2</sub> (Mg,Fe <sup>2+</sup> ) <sub>5</sub> Si <sub>8</sub> O <sub>22</sub> (OH,F) <sub>2</sub>	2709—2701	
	-		
Sapphirine	$Mg_4(Mg_3Al_9)O_4(Si_3Al_9O_{36})$	2500-2448	1
Rhodonite	CaMn₃Mn(Si₅O15)	2500-2216	
Osumilite	(K,Na)(Fe <sup>2+</sup> ,Mg) <sub>2</sub> (Al,Fe <sup>3+</sup> ) <sub>3</sub> (Si,Al) <sub>12</sub> O <sub>30</sub>	2485—496	1
Humite	$Mg_9(SiO_4)_4(F,OH)_2$	2070—1950	4
Periclase	MgO	2058—2058	4
Coesite	SiO <sub>2</sub>	2050—2050	6
Jadeite	NaAlSi <sub>2</sub> O <sub>6</sub>	2038—1270	6
Monticellite	CaMgSiO <sub>4</sub>	2000—1109	3
Spurrite	$Ca_5(SiO_4)_2(CO_3)$	1750—248	3
Mullite	$AI_{4+2x}Si_{2-2x}O_{10-x}$ (x $\approx 0.4$ )	960—248	1
Merwinite	$Ca_3Mg(SiO_4)_2$	794—684	3
Lawsonite	(Ca,Sr)Al <sub>2</sub> (Si <sub>2</sub> O <sub>7</sub> )(OH) <sub>2</sub> ·H <sub>2</sub> O	599—497	3
Bredigite	$Ca_7Mg(SiO_4)_4$	354—248	
Larnite	Ca <sub>2</sub> SiO <sub>4</sub>	354—248	3
Rankinite	$Ca_3Si_2O_7$	354—66	3
Tilleyite	$Ca_5Si_2O_7(CO_3)_2$	300-300	3

cite both me from В different localities.

1020 2. Community as illustrated in Figure 1. Note that actinolite, bredigite, spessartine, grunerite, 1021 rhodonite, and piemontite were not included in Figure 1 minerals.

1022 3. Diamond ages were provided by Shirey et al. (2013).

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1024 Earth's first metamorphic rocks must have been thermally altered xenoliths and contact

1025 zones (hornfels and sanidinite facies) associated with early Hadean igneous activity (> 4.5 Ga).

1026 The appearance of new Hadean lithologies, including clay-rich sediments, arkosic sandstones,

1027 and carbonates, provided additional protoliths for thermal metamorphism prior to 4.0 Ga.

1028 Indeed, the 8 oldest minerals in Table 4, all Paleoarchean or older (> 3.2 Ga), are associated

1029 with higher temperature metamorphic regimes.

Also appearing in the Meso- and Neoarcheon Eras (> 2.5 Ga) are representative minerals from regional and high-pressure metamorphic rocks (Table 4). The exposure of extensive regional metamorphic terrains by orogenesis and erosion, with lithologies corresponding to the Barrovian sequence of index mineral metamorphic zones, thus appears to have occurred significantly later in Earth history, perhaps in association with plate tectonics. More recently, rapid subduction and rebound of crustal wedges, coupled with a shallowing

1036 geothermal gradient, has produced distinctive suites of blueschist, eclogite, and ultrahigh 1037 pressure metamorphic suites. Some of these phases, including *glacophane, jadeite, pyrope,* 1038 *staurolite,* are first recorded in the Neoarchean or Paleoproterozoic Eras (2.8 to 2.0 Ga), 1039 perhaps signaling the commencement of subduction-driven tectonics. However, a number of 1040 key high-pressure metamorphic minerals, including *coesite, jadeite,* and *lawsonite,* are only

1041 known since 2.05 Ga (Table 4).

1042 The evolution of the metamorphic minerals continued into the Phanerozoic Eon (< 540 Ma).

1043 Intriguingly, 5 of the most recent (< 800 Ma) metamorphic minerals in Table 4—*bredigite*, 1044 *larnite, merwinite, rankinite,* and *tilleyite*—are all Ca-(Mg)-silicates that form by low-P contact 1045 metamorphism of limestone or dolomite. The lack of older known examples of these minerals 1046 may reflect a sparsity of dated examples, but enhanced biogenic production of carbonate 1047 lithologies may also have played a role. Indeed, the Phanerozoic Eon represents a distinctive 1048 phase in metamorphic mineral evolution. New biogenic lithologies such as coal, phosphorites, 1049 and reef carbonates were subjected to both regional and thermal metamorphism, while the

intense burning of fossil hydrocarbons in the form of coal, oil, and natural gas created new
 pyrometamorphic regimes—processes to be considered in a subsequent contribution to this
 series. The evolution of metamorphic minerals thus epitomizes the kinds of changes in physical,
 chemical, and ultimately biological processes in the crust and upper mantle that exemply
 mineral evolution over more than 4.5 billion years of Earth history.
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#### Table 3. Summary of 94 frequently occurring metamorphic minerals, with their compositions, 1650 abundances, and numbers of IMA species included in each mineral kind (see Supplementary Table 3).

1651 1652

1052				• • 2	
1653	Mineral Kind	Chemical Formula	Abundance	Community	#IMA <sup>°</sup>
1654	Native Element	S			
1655	Diamond	С	10	6	1
1656	Graphite	С	105	5	1
1657	Sulfides				
1658	Chalcopyrite	CuFeS <sub>2</sub>	99	2	1
1659	Pyrite	FeS <sub>2</sub>	120		1
1660	Pyrrhotite	Fe <sub>7</sub> S <sub>8</sub>	192		1
1661	Pentlandite	(Ni,Fe)₀S <sub>8</sub>	21	5	1
1662	Oxides				
1663	Rutile	TiO <sub>2</sub>	297		1
1664	Hematite	Fe <sub>2</sub> O <sub>3</sub>	24		1
1665	Corundum	Al <sub>2</sub> O <sub>3</sub>	85	1	1
1666	Periclase	MgO	34	4	1
1667	Baddeleyite	ZrO <sub>2</sub>	18	4	1
1668	Magnetite	$Fe^{2+}Fe^{3+}{}_{2}O_{4}$	429		1
1669	Hercynite	Fe <sup>2+</sup> Al <sub>2</sub> O <sub>4</sub>	34	1	1
1670	Spinel	(Mg,Fe <sup>2+</sup> )(Al,Fe <sup>3+</sup> ,Cr <sup>3+</sup> ) <sub>2</sub> O <sub>4</sub>	263	4	1
1671	Chromite	$Fe^{2+}Cr^{3+}_{2}O_{4}$	12	5	3
1672	Ilmenite	$Fe^{2+}Ti^{4+}O_3$	181		1

1673	Perovskite	CaTi <sup>4+</sup> O <sub>3</sub>	12	3	1
1674	Geikielite	MgTiO₃	17	4	1
1675	Pseudobrooki	<i>te</i> Fe <sup>3+</sup> <sub>2</sub> TiO <sub>5</sub>	15	1	1
1676	Hydroxide				
1677	Brucite	Mg(OH) <sub>2</sub>	31	4	1
1678	Carbonates				
1679	Calcite	CaCO <sub>3</sub>	645	4	1
1680	Magnesite	MgCO <sub>3</sub>	16	5	1
1681	Dolomite	CaMg(CO <sub>3</sub> ) <sub>2</sub>	152	4	2
1682	Fe-Dolomite	Ca(Mg,Fe)(CO <sub>3</sub> ) <sub>2</sub> [0.15 < Fe/(Mg + Fe) < 0.50]	116	5	0
1683	Aragonite	CaCO <sub>3</sub>	15	2	1
1684	Spurrite	$Ca_5(SiO_4)_2(CO_3)$	51	3	1
1685	Tilleyite	$Ca_5Si_2O_7(CO_3)_2$	16	3	1
1686	Phosphates				
1687	Apatite	$Ca_5(PO_4)_3(F,OH,CI)$	155		2
1688	Nesosilicates or (	Orthosilicates			
1689	<u>Olivine Group</u>				
1690	Forsterite	Mg <sub>2</sub> SiO <sub>4</sub>	173	4	1
1691	Fayalite	Fe <sup>2+</sup> <sub>2</sub> SIO <sub>4</sub>	10	2	1
1692	Olivine	(Mg,Fe) <sub>2</sub> SiO <sub>4</sub> (0.3 < Fe/(Fe + Mg) < 0.7)	53	6	0
1693	Monticellite	CaMgSiO <sub>4</sub>	77	3	1
1694	<u>Garnet Group</u>				
1695	Almandine	$Fe^{2+}_{3}Al_{2}Si_{3}O_{12}$	367		1

1696	Andradite	$Ca_{3}Fe^{3+}{}_{2}Si_{3}O_{12}$	28	2	3
1697	Grossular	$Ca_3Al_2Si_3O_{12}$	162	2	1
1698	Pyrope	$Mg_3Al_2Si_3O_{12}$	192	6	2
1699	Aluminosilicate	Group			
1700	Andalusite	Al <sub>2</sub> SiO <sub>5</sub>	146	1	1
1701	Kyanite	Al <sub>2</sub> SiO <sub>5</sub>	102	6	1
1702	Sillimanite	Al <sub>2</sub> SiO <sub>5</sub>	235	1	1
1703	Mullite	Al <sub>4+2x</sub> Si <sub>2-2x</sub> O <sub>10-x</sub> (x ≈ 0.4)	62	1	1
1704	<u>Other Nesosilica</u>	<u>ites</u>			
1705	Larnite	Ca <sub>2</sub> SiO <sub>4</sub>	24	3	1
1706	Merwinite	Ca <sub>3</sub> Mg(SiO <sub>4</sub> ) <sub>2</sub>	38	3	1
1707	Humite	Mg <sub>9</sub> (SiO <sub>4</sub> ) <sub>4</sub> (F,OH) <sub>2</sub>	21	4	2
1708	Chloritoid	$Fe^{2+}Al_2O(SiO_4)(OH)_2$	46	6	3
1709	Staurolite	Fe <sup>2+</sup> <sub>2</sub> Al <sub>9</sub> Si <sub>4</sub> O <sub>23</sub> (OH)	52	1	1
1710	Zircon	ZrSiO <sub>4</sub>	66		1
1711	Titanite	CaTi <sup>4+</sup> SiO₅	186		1
1712	Sorosilicates or D	Disilicates			
1713	Allanite	(CaCe)(AIAIFe <sup>2+</sup> )O[Si <sub>2</sub> O <sub>7</sub> ][SiO <sub>4</sub> ](OH)	16	5	5
1714	Epidote	Ca <sub>2</sub> (Al <sub>2</sub> Fe <sup>3+</sup> )[Si <sub>2</sub> O <sub>7</sub> ][SiO <sub>4</sub> ]O(OH)	157		4
1715	Zoisite	$Ca_2AI_3[Si_2O_7][SiO_4]O(OH)$	156	6	1
1716	Melilite	$(Ca,Na)_2(Mg,Fe^{2+},Al,Si)_3O_7$	108	3	3
1717	Lawsonite	$(Ca,Sr)Al_2(Si_2O_7)(OH)_2 \cdot H_2O$	11	6	2
1718	Pumpellyite	$Ca_2AI_3(Si_2O_7)(SiO_4)(OH,O)_2 \cdot H_2O$	10	5	9

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			-		
1719	Rankinite	Ca <sub>3</sub> Si <sub>2</sub> O <sub>7</sub>	20	3	1
1720	Vesuvianite	(Ca,Na) <sub>19</sub> (Al,Mg,Fe) <sub>13</sub> (SiO <sub>4</sub> ) <sub>10</sub> (Si <sub>2</sub> O <sub>7</sub> ) <sub>4</sub> (OH,F,O) <sub>10</sub>	15	2	10
1721	Cyclosilicates				
1722	Tourmaline	$(\Box, Na, Ca)(Mg, Fe, Al)_3(Mg, Al)_6(Si_6O_{18})(BO_3)_3(OH, F)_3(OH, O)$	47	5	18
1723	Cordierite	(Mg,Fe <sup>2+</sup> ) <sub>2</sub> Al <sub>4</sub> Si <sub>5</sub> O <sub>18</sub>	395	1	2
1724	Osumilite	$(K,Na)(Fe^{2+},Mg)_2(AI,Fe^{3+})_3(Si,AI)_{12}O_{30}$	13	1	4
1725	Inosilicates				
1726	<u>Pyroxene Group</u>				
1727	Aegirine	(Ca,Na)(Fe <sup>3+</sup> ,Mg,Fe <sup>2+</sup> )Si <sub>2</sub> O <sub>6</sub>	16	6	2
1728	Augite	$(Ca,Mg,Fe^{2+},Fe^{3+})_2(Si,AI)_2O_6$ [0.15 < CaSiO <sub>3</sub> < 0.45]	150		1
1729	Diopside	$CaMgSi_2O_6$	409	2	1
1730	Hedenbergite	CaFe <sup>2+</sup> Si <sub>2</sub> O <sub>6</sub>	16	2	1
1731	Jadeite	NaAlSi <sub>2</sub> O <sub>6</sub>	34	6	1
1732	Omphacite	$(Ca,Na)(Mg,Fe,AI)Si_2O_6$	151	6	1
1733	Orthopyroxene	? (Mg,Fe <sup>2+</sup> )SiO <sub>3</sub>	281		2
1734	<u>Amphibole Group</u>	2			
1735	Actinolite	$\Box Ca_2(Mg,Fe^{2+})_5Si_8O_{22}(OH,F)_2$	74		2
1736	Anthophyllite	$\Box Mg_2 Mg_5 Si_8 O_{22} (OH)_2$	52	1	6
1737	Cummingtonite	e □Mg <sub>2</sub> Mg <sub>5</sub> Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub>	12	4	1
1738	Glaucophane	$\Box Na_2(Mg_3Al_2)Si_8O_{22}(OH)_2$	79	6	2
1739	Hornblende	(Na,K)Ca <sub>2</sub> (Mg,Fe <sup>2+</sup> ,Al,Fe <sup>3+</sup> ) <sub>5</sub> (Si,Al) <sub>8</sub> O <sub>22</sub> (OH,F,Cl) <sub>2</sub>	387		26
1740	Richterite	$Na(CaNa)(Mg,Fe^{2+})_{5}(Si,Al,Fe^{3+})_{8}O_{22}(OH)_{2}$	12	6	16
1741	Tremolite	□Ca <sub>2</sub> (Mg <sub>5.0-4.5</sub> Fe <sup>2+</sup> <sub>0.0-0.5</sub> )Si <sub>8</sub> O <sub>22</sub> (OH) <sub>2</sub>	122	4	2

1742	Other Chain Silic	<u>ates</u>			
1743	Wollastonite	CaSiO₃	158	2	1
1744	Sapphirine	$Mg_4(Mg_3Al_9)O_4(Si_3Al_9O_{36})$	28	1	2
1745	Phyllosilicates				
1746	<u>Mica Group</u>				
1747	Biotite	KFe <sup>2+</sup> <sub>2</sub> (Fe <sup>2+</sup> ,Mg,Mn <sup>2+</sup> )(Si,Al,Fe <sup>3+</sup> ) <sub>2</sub> Si <sub>2</sub> O <sub>10</sub> (OH,F,Cl) <sub>2</sub>	822		6
1748	Phlogopite	[[KMg <sub>2</sub> (Mg,Fe <sup>2+</sup> ,Mn <sup>2+</sup> ,Fe <sup>3+</sup> ,Ti <sup>4+</sup> )(Si,Al,Fe <sup>3+</sup> ) <sub>2</sub> Si <sub>2</sub> O <sub>10</sub> (OH,F) <sub>2</sub> ]]	78	4	6
1749	Muscovite	$K(AI,V,Fe^{3+},Cr)_2(Si_3AI)O_{10}(OH)_2$	515		3
1750	Phengite	$K(AI,Mg,Fe)_{2-3}(Si_3AI)O_{10}(OH)_2$	110	6	1
1751	Paragonite	$NaAl_2(Si_3Al)O_{10}(OH)_2$	67	6	1
1752	<u>Other Layer Silice</u>	ates			
1753	Chlorite	(Mg,Fe <sup>2+</sup> ) <sub>5</sub> (Al,Fe <sup>3+</sup> )(Si <sub>3</sub> AlO <sub>10</sub> )(OH) <sub>8</sub>	339	5	3
1754	Serpentine	(Mg,Fe) <sub>2</sub> Al(AlSiO <sub>5</sub> )(OH) <sub>4</sub>	68	4	5
1755	Talc	Mg <sub>3</sub> Si <sub>4</sub> O <sub>10</sub> (OH) <sub>2</sub>	73	5	1
1756	Prehnite	Ca <sub>2</sub> Al(Si <sub>3</sub> Al)O <sub>10</sub> (OH) <sub>2</sub>	16	5	1
1757	Tectosilicate				
1758	<u>Silica Group</u>				
1759	Quartz	SiO <sub>2</sub>	1353		1
1760	Tridymite	SiO <sub>2</sub>	44	1	1
1761	Coesite	SiO <sub>2</sub>	23	6	1
1762	Cristobalite	SiO <sub>2</sub>	10	1	1
1763	<u>Feldspar Group</u>				
1764	Albite	NaAlSi₃O <sub>8</sub>	177		1

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1765	Anorthite	CaAl <sub>2</sub> Si <sub>2</sub> O <sub>8</sub>	78	2	1
1766	Plagioclase	(Na,Ca)Al(Al,Si) <sub>3</sub> O <sub>8</sub>	809		0
1767	Kspar	KAISi <sub>3</sub> O <sub>8</sub>	411		1
1768	Sanidine	KAISi <sub>3</sub> O <sub>8</sub>	102	1	1
1769	1769 <u>Other Framework Silicates</u>				
1770	Scapolite	(Na,Ca) <sub>4</sub> (Al,Si) <sub>12</sub> O <sub>24</sub> (CO <sub>3</sub> ,SO <sub>4</sub> ,Cl)	33	2	3
1771	Silicate Glass	$(Si,Al,Ca,Mg,Fe,O; SiO_2 < 70 wt \%)$	68	1	0

1772<sup>1</sup> Number of occurrences in a survey of the modes of 2785 metamorphic rocks; see Supplementary Table 3.

1773<sup>2</sup> The Community Number refers to the 6 communities illustrated in Figure 1A, based on a survey of 73 minerals.

1774<sup>3</sup> The number of IMA-approved mineral species lumped into this mineral kind (see Supplementary Table 2). For example, tourmaline (which is not

1775 an IMA-approved mineral name) includes 18 IMA species. Note that *biotite, chlorite, Fe-dolomite, kspar, melilite, olivine, phengite, plagioclase,* 

1776 scapolite, serpentine, and silicate glass are not IMA-approved names and do not correspond to any single IMA species.

#### 1777 APPENDIX I. Systematic Mineralogy of Metamorphic Minerals

1778 Appendix I presents a systematic mineralogy of the 94 most frequently encountered 1779 metamorphic minerals. Supplementary Table 1 provides a list of 1220 metamorphic mineral 1780 species, the corresponding 755 metamorphic mineral kinds, and the distribution of these 1781 phases among 8 major groups of metamorphic rocks. This conversion of 1220 metamorphic 1782 minerals into 755 natural kinds requires several modifications to the IMA list, as detailed in 1783 Hazen et al (2022). In 568 instances, the IMA species name (e.g., augite) is identical to the 1784 natural kind name (augite). Note that in this contribution we italicize the names of mineral 1785 natural kinds to distinguish them from IMA-CNMNC-approved mineral species. In the case of 1786 652 IMA-approved species, we lump groups of two or more IMA species into single natural 1787 kinds. These numerous examples, resulting in a reduction from 652 species to 187 root natural 1788 kinds, are detailed in Supplementary Table 2 (see also Supplementary Read-Me File 2). For 1789 example, *pumpellyite* combines 9 IMA-approved species of the pumpellyite group. In 18 1790 instances, the name assigned to the natural kind is a group name that is not itself an approved 1791 IMA species name. Thus, hornblende lumps 26 IMA-approved species of calcic amphiboles, 1792 while *biotite* encompasses 6 species of Fe-bearing trioctahedral micas.

We include five phases that do not correspond to an IMA-CNMNC-approved species in Supplementary Table 1. In the instance of *plagioclase*, we recognize intermediate compositions of the albite (NaAlSi<sub>3</sub>O<sub>8</sub>)—anorthite (CaAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>) solid solution with 0.15 < Ca/(Ca + Na) < 0.85 as a separate natural kind. Similarly, *olivine* refers to intermediate compositions of the forsterite (Mg<sub>2</sub>SiO<sub>4</sub>)—fayalite (Fe<sub>2</sub>SiO<sub>4</sub>) solid solution for which 0.3 < Fe/(Fe + Mg) < 0.7. *Fe-Dolomite* refers to intermediate compositions of the dolomite-ankerite solid solution, where 0.15 <

1799 Fe/(Fe + Mg) < 0.50, many examples of which are incorrectly described as ankerite (Ferry et al. 1800 2015). Phengite is a fine-grained variety of muscovite  $[K(AI,Mg,Fe)_{2-3}(AISi_3)O_{10}(OH)_2]$ , typically 1801 with excess Si, that is common in high-pressure metamorphic deposits. We also introduce 1802 silicate alass, an important amorphous phase in some pyrometamorphic lithologies, as a 1803 natural kind. 1804 Of the 755 mineral kinds recorded in Supplementary Tables 1 and 2, 94 phases are relatively 1805 common based on their occurrence in at least 10 rocks in 2785 metamorphic rocks (Table 3; 1806 Supplementary Table 3). Supplementary Table 3 also records a literature reference for each

1807 rock mode, the rock's locality, and information on the type of metamorphism, the metamorphic

1808 facies, and its protolith. Here we present brief descriptions of these 94 mineral kinds, arranged

1809 according to the New Dana Classification (Gaines et al. 1997).

### 1810 Native Elements

1811 Two polymorphs of carbon (C), graphite and diamond, with 105 and 10 occurrences in 1812 Supplementary Table 3, respectively, are the only native element minerals that occur in any 1813 significant abundance in metamorphic rocks, though more than two dozen rare native elements 1814 and metal alloys are listed in Supplementary Table 1. Graphite most frequently occurs in 1815 carbon-rich metapelites (Landis 1971; Diessel et al. 1978; Buseck and Huang 1985). However, 1816 graphite is also reported to occur as a product of retrograde metamorphism of diamond, at 1817 times as euhedral pseudomorphs (Pearson et al. 1989; Ferry 1992; Davies et al. 1993; Leech and 1818 Ernst 1998). In addition, Ross et al. (1991) reported an occurrence of *graphite* formed by the 1819 shearing of coal—an example of Phanerozoic metamorphism of a biotic protolith.

Most *diamond* formation occurs in the mantle by precipitation from carbon-rich fluids (Jacob and Mikhail 2022; Kjarsgaard et al. 2022). However, in some instances, micrometer-scale *diamond* forms during subduction, ultra-deep metamorphism, and subsequent rebound of carbon-bearing crustal wedges (Dobrzhinetskya et al. 1995, 2022).

1824

1825 Sulfides

1826 More than 50 sulfide and other chalcogenide minerals have been reported from 1827 metamorphic rocks (Supplementary Table 1). However, only four iron-bearing sulfides, pyrite 1828 (FeS<sub>2</sub>), pyrrhotite (Fe<sub>7</sub>S<sub>8</sub>), chalcopyrite (CuFeS<sub>2</sub>), and pentlandite [(Ni,Fe)<sub>9</sub>S<sub>8</sub>], are relatively 1829 common. *Pyrrhotite* is the most commonly observed sulfide in our survey, occurring in 192 1830 metamorphic rocks. It frequently occurs with *graphite* in higher-grade regional metamorphic 1831 rocks (Hoscheck 1984; Ferry 1992), and it may form by solid-state transformation of pyrite 1832 coupled with sulfur loss (Bowles et al. 2011). It is also a common sulfide mineral associated with 1833 skarn deposits (Einaudi and Burt 1982).

1834 *Pyrite* (FeS<sub>2</sub>) is the next most frequently reported metamorphic sulfide, with 120 occurrences

1835 in Supplementary Table 3. *Pyrite* is found in all metamorphic grades, including high-pressure

regimes, and may form through solid-state transformation of *pyrrhotite* (Hall et al. 1987). *Pyrite* 

also occurs via contact metamorphism associated with skarn formation (Einaudi et al. 1981),

1838 and in shear zones (e.g., Harker 1950).

1839 *Chalcopyrite*, the only copper-bearing mineral among the most common metamorphic 1840 phases, is reported from 99 rocks in our survey. Most occurrences are as minor grains in 1841 regionally metamorphosed igneous and sedimentary lithologies (Ferry 1984, 1992, 1994; Ferry

1842 et al. 2001). It is likely that chalcopyrite, as an opaque and often micrometer-scale phase, is

- 1843 underrepresented in our study.
- 1844 The 21 occurrences of *pentlandite*, the only common nickel-bearing metamorphic minerals,
- 1845 are recorded exclusively from ultramafic lithologies (Ferry 1995; Ferry et al. 2005).
- 1846 In addition, it should be noted that *sphalerite* (ZnS) occurs in 9 of the rocks surveyed—a
- 1847 number that likely significantly underestimates the frequency of this opaque and typically
- 1848 minute zinc-bearing phase.
- 1849
- 1850 **Oxides**
- 1851 Oxide minerals occur in the entire range of metamorphic rocks. More than 50 species, most

1852 of them relatively rare, have been reported (Supplementary Table 1). We list 13 mineral kinds

1853 among the most frequently encountered metamorphic minerals.

1854 The simple oxide *rutile* (TiO<sub>2</sub>) is reported in 297 of the metamorphic rocks from our study

1855 (Supplementary Table 3), most often in regional metamorphic rocks by transformation of prior

1856 Ti-bearing phases, including *titanite*, *ilmenite*, and titaniferous micas and *magnetite* (Bowles et

al. 2011), but also in a range of pyrometamorphic (Grapes 2006), contact metamorphic

1858 (Reverdatto and Sóbolev 1973), and shear zone (Harker 1950) rocks.

1859 Additionally, among the more common simple oxides is *corundum* (Al<sub>2</sub>O<sub>3</sub>; 85 occurrences),

- 1860 observed most frequently in silica-poor lithologies subjected to high temperature, notably in
- 1861 contact metamorphic zones. Al-rich lithologies with prominent *corundum*, often in association
- 1862 with *spinel, mullite, cordierite*, and/or *sanidine*, are known as "emery." *Corundum* also occurs in

1863 pyrometamorphosed limestone xenoliths (Joplin 1968; Grapes 2006), as well as in high-grade

1864 regional metamorphic rocks (Harker 1950; Augustithus 1985).

Periclase (MgO) is a common mineral in *calcite/dolomite* marbles, often in association with *brucite* (Carpenter 1967; Bowles et al. 2011). As early as 1940, Bowen described *periclase* as part of an evolutionary thermal metamorphic sequence (Bowen 1940). We record 36 occurences of *periclase*, primarily in limestone xenoliths and contact metamorphic environments (Augustithus 1985; Grapes 2006; Ferry and Rumble 2007).

1870 Hematite (Fe<sub>2</sub>O<sub>3</sub>), represented by 24 occurrences in our tabulation, forms from Fe-rich

1871 protoliths in oxidized environments. Contexts of metamorphic hematite include xenoliths,

1872 contact environments, iron formations, and regional metamorphism (Bowles et al. 2011).

1873 We record 18 occurrences of the zirconium oxide baddeleyite (ZrO<sub>2</sub>) in both contact and 1874 regional metamorphic contexts (Ferry and Rumble 2007). That number that may 1875 underrepresent the frequency of baddeleyite because of its typical low modal abundance and 1876 small grain size.

The most frequently encountered metamorphic oxides are members of the spinel group, which occur in 672 (24 %) of the 2785 metamorphic rocks we tabulated. With the general formula  $[(Mg,Fe^{2+})(Al,Fe^{3+},Cr^{3+},Ti)_2O_4]$ , the spinel group encompasses a complex range of solid solutions (Bowles et al. 2011), of which four end-members are among the most commonly reported metamorphic minerals: *magnetite* (Fe<sup>2+</sup>Fe<sup>3+</sup><sub>2</sub>O<sub>4</sub>), *spinel* (MgAl<sub>2</sub>O<sub>4</sub>; although "spinel" may also refer to the mineral group rather than the species in some modes), *chromite* (Fe<sup>2+</sup>Cr<sup>3+</sup><sub>2</sub>O<sub>4</sub>), and *hercynite* (Fe<sup>2+</sup>Al<sub>2</sub>O<sub>4</sub>). In addition, several reports cite "pleonaste," which

1884 refers to Fe<sup>2+</sup>-bearing intermediate compositions of the spinel-hercynite solid solution—

1885 examples that we include with *spinel*.

1886 *Magnetite* is by far the most frequently reported metamorphic oxide, occurring in 429 (15 %)

1887 of the 2785 metamorphic rocks we compiled, and in a wide range of metamorphic contexts

1888 (Joplin 1968; Reverdatto and Sóbolev 1973; Grapes 2006). *Magnetite* often forms by thermal

1889 alteration of ferric iron oxide/hydroxides, as well as through the introduction of Fe-rich fluids—

1890 a situation in which the distinction between metamorphism and metasomatism may be

1891 blurred.

1892 The Mg-Al oxide *spinel*, with 263 occurrences in our list, is found in numerous contact and

1893 regional metamorphic contexts, principally as a high-temperature mineral in metacarbonates

and Al-rich protoliths (Harker 1950; Botha 1983; Carswell 1990; Grapes 2006).

1895 *Hercynite*, which forms in high-grade metamorphic environments, often in association with

1896 *corundum, mullite,* and/or *sillimanite,* is represented by 34 examples in Supplementary Table 3.

1897 Metamorphic contexts include xenolith, contact, and regional environments (Harker 1950;

1898 Grapes 2006; Bowles et al. 2011).

1899 We list 12 occurrences of metamorphic *chromite*—a number that likely underestimates this

1900 most common chromium mineral because it is opaque and easily mistaken for other Fe-bearing

1901 oxides. *Chromite* is most often associated with ultramafic lithologies, particularly ophiolites.

1902 Four titanium-bearing double oxides are listed among the 94 most common metamorphic

1903 phases. We record *Ilmenite* (FeTiO<sub>3</sub>) from 181 xenoliths, contact metamorphic rocks, or regional

1904 metamorphic formations, notably forming via alteration of mafic and Fe-bearing lithologies

1905 (Reverdatto and Sóbolev 1973; Carswell 1990; Grapes 2006). Geikielite (MgTiO<sub>3</sub>), the

magnesium analog of ilmenite, occurs in 17 contact and regional metasedimentary rocks in Supplementary Table 3. Geikeilite protoliths include carbonates, calc silicates, pelites, and sandstones. *Perovskite* (CaTiO<sub>3</sub>) is reported from 12 rocks, primarily in xenoliths and contact metamorphic environments with impure limestone (Murdoch 1951; Fulignati et al. 2000; Grapes 2006). *Pseudobrookite* ( $Fe^{3+}_2TiO_5$ ), which we record in 15 primarily xenolith and contact metamorphic rocks, forms most often by the high-temperature oxidation of *ilmenite* (Agrell and Langley 1958; Smith 1969; Basta and Shaalan 1974).

1914 occurred in 31 rocks of our survey. It is typically the consequence of hydration of *periclase* in

*Brucite* [Mg(OH)<sub>2</sub>], the only relatively common hydroxide mineral in metamorphic rocks,

altered Ca-Mg carbonates (Nakajima et al. 1992; Ferry et al. 2002; Ferry and Rumble 2007;

1916 Bowles et al. 2011).

1917

1913

## 1918 Carbonates

1919 At least 70 carbonate minerals have been reported from metamorphic rocks (Supplementary 1920 Table 1), though only seven species occur with any significant frequency. By far the most 1921 abundant carbonates are *calcite* (CaCO<sub>3</sub>), *dolomite* [CaMg(CO<sub>3</sub>)<sub>2</sub>], and the intermediate 1922 composition *Fe-dolomite* [*dolomite* with a significant *ankerite* CaFe( $CO_3$ )<sub>2</sub> component], with 1923 645, 152, and 116 occurrences in our compilation, respectively. *Calcite* and *dolomite* most often 1924 occur in metamorphosed limestones and other carbonate-bearing protoliths (Harker 1950; 1925 Reverdatto and Sóbolev 1973; Chang et al. 1996; Grapes 2006). In many instances, these phases 1926 form through recrystallization of prior carbonates to form a marble (Chang et al. 1996; Philpotts 1927 and Ague 2009). Fe-dolomite (often reported as "ankerite" in the petrologic literature) is found

1928 primarily in regionally metamorphosed sediments (Ferry 1992, 1994, 2007; Ferry et al. 2015)

1929 where it formed through carbonation reactions during metamorphism/metasomatism (Spooner

1930 and Fyfe 1973).

1931 Metamorphic *magnesite* (MgCO<sub>3</sub>) with 16 occurrences is characteristic of altered ophiolites,

1932 where it occurs in association with talc and serpentine. It typically forms via the carbonation of

- 1933 Mg-bearing oxides and silicates (Chang et al. 1996).
- 1934 *Aragonite*, a high-pressure form of CaCO<sub>3</sub>, was recorded in 15 rocks of eclogite, blueschist,
- and ultrahigh-pressure facies carbonate rocks (Carswell 1990; Carswell and Compagnoni 2003;

1936 Philpotts and Ague 2009).

1937 Two silicate carbonates, *spurrite*  $[Ca_5(SiO_4)_2(CO_3)]$  with 51 examples and *tilleyite* 1938  $[Ca_5Si_2O_7(CO_3)_2]$  with 16 examples, arise when *calcite* and *wollastonite* react at high 1939 temperature (Tuttle and Harker 1957; Zharikov and Shmulovich 1969).

1940

### 1941 **Phosphates**

Of the more than 50 metamorphic phosphates recorded in Supplementary Table 1, only the calcium phosphate *apatite* [Ca<sub>5</sub>(PO<sub>4</sub>)<sub>3</sub>(F,OH)] is widely reported, with 155 occurrences in Supplementary Table 3. We lump two common species, fluorapatite and hydroxylapatite, which are rarely differentiated in reports of metamorphic mineral modes. The majority of these *apatite* occurrences are in high-grade metamorphosed mafic igneous rocks, including granulites and eclogites (Harker 1950; Joplin 1968; Carswell 1990). In addition, *apatite* has been reported from contact metamorphic and shear environments (Harker 1950; Joplin 1968).

1949

#### 1950 Silicates

- 1951 Silicates constitute the majority of metamorphic minerals, both volumetrically and in terms 1952 of diversity. In Supplementary Tables 1 and 2 we record 746 silicate mineral species, 1953 corresponding to 418 root natural kinds, of which 66 are frequently encountered metamorphic 1954 phases. In the following sections we review these more common silicates.
- 1955

## 1956 Nesosilicates or Orthosilicates

Orthosilicates, with silicon exclusively in insular  $SiO_4^{4-}$  structural groups, are characteristic minerals in environments with relatively low Si, notably those associated with carbonate, calcsilicate, or aluminous protoliths (Deer et al. 1982). We detail 21 orthosilicates in addition to the orthosilicate-carbonate mineral *spurrite*, described above. Of these 22 phases, 16 contain essential Ca and/or Al.

1962 *Olivine Group:* We recognize four members of the olivine group  $[(Mg,Fe,Ca)_2SiO_4]$  as 1963 important metamorphic minerals (Deer et al. 1982). The Mg olivine forsterite (ideally Mg<sub>2</sub>SiO<sub>4</sub>) 1964 is reported in 173 of the rocks we surveyed, notably via contact, regional, or high-pressure 1965 metamorphism of silica-poor ultramafic (Springer 1974; Pinsent and Hirst 1977) or carbonate-1966 bearing (Weeks 1956; Schreyer et al. 1972; Suzuki 1977) protoliths. We also distinguish olivine 1967 as intermediate Mg-Fe compositions with 0.3 < Fe/(Fe + Mg) < 0.7, which are common in 1968 metamorphosed mafic and ultramafic rocks (53 occurrences; Reverdatto and Sóbolev 1973; 1969 Ferry et al. 1987).

1970 *Fayalite* (ideally Fe<sub>2</sub>SiO<sub>4</sub>) is much less common, occurring in 10 rocks with Fe-rich protoliths, 1971 including mafic rocks and iron formations (Joplin 1968; Simmons et al. 1974; Floran and Papike

1972 1978; Carswell 1990). Note that metamorphic Fe-dominant olivines with intermediate

1973 compositions are less common than examples close to either end-member (Deer et al. 1982).

*Monticellite* (CaMgSiO<sub>4</sub>), with 77 occurrences, is commonly found in contact metamorphic environments with siliceous carbonate lithologies, often forming with increasing temperature at the expense of *diopside*, *forsterite*, and/or *wollastonite* (Bowen 1940; Turner 1967; Deer et al. 1982). *Monticellite* frequently co-occurs with *forsterite*, as the solid solution between these

1978 two olivine group minerals is limited (Warner and Luth 1973).

1979 Garnet Group: The garnet group is represented by four relatively common metamorphic 1980 phases, occurring in 745 (27 %) of 2785 rocks in our survey. Garnets collectively display a 1981 significant compositional range, typically with solid solutions among two or three end-members 1982 (Deer et al. 1982; Chiama et al. 2020, 2022). Ideal end-members of these minerals are almandine  $[Fe^{2+}_{3}Al_{2}(SiO_{4})_{3}]$ , and radite  $[Ca_{3}Fe^{3+}_{2}(SiO_{4})_{3}]$ , grossular  $[Ca_{3}Al_{2}(SiO_{4})_{3}]$ , and pyrope 1983  $[Mg_3Al_2(SiO_4)_3]$ , often with a significant spessartine  $[Mn^{2+}_3Al_2(SiO_4)_3]$  component, as well, 1984 1985 though true Mn-dominant spessartine is recorded in only 9 occurrences in our compilation 1986 (Woodland 1938; Roy 1965; Jan and Symmes 1977).

In some instances, such as *pyrope-almandine-spessartine* ("pyralspite") from ecologites and other high-grade metamorphic rocks, *grossular-andradite* ("grandite") from the contact metamorphism of carbonate-bearing sediments, and contact metamorphic garnets in the *grossular-spessartine-almandine* field (Shimazaki 1977), the compositional ranges among endmembers may be continuous, thus warranting lumping of species into a single metamorphic mineral kind. However, until cluster analysis (Gregory et al. 2019; Boujibar et al. 2021; Hystad et

al. 2021) can be performed on a wide range of garnet compositions from known paragenetic

1994 environments, we will treat these five types of metamorphic garnet separately.

1995 Almandine, with 367 occurrences in Supplementary Table 3, is the commonest garnet in 1996 metamorphic rocks (Harker 1950; Joplin 1968; Reverdatto and Sóbolev 1973; Botha 1983; 1997 Augustithus 1985). Most *almandine* forms in a regional metamorphic context, derived from 1998 mafic or pelitic protoliths (Atherton 1964; Deer et al. 1982), including high-pressure examples 1999 from blueschist (Coleman and Lee 1963; Banno and Matsui 1965), eclogite (Coleman et al. 2000 1965), and granulite (Buddington 1952; Eskola 1952) facies. In addition, almandine from 2001 contact metamorphism of pelites is not uncommon (Tilley 1926; Stewart 1942), while it also 2002 occurs in some metamorphosed iron formations (Klein 1966). 2003 *Pyrope*'s 192 entries are overwhelmingly from high-pressure metamorphic environments, in

2004 many instances from ecologite-grade rocks with mafic precursors, most commonly in 2005 association with *omphacite* (Carswell 1990). Metamorphic *pyrope* typically has a significant 2006 *almandine* component (Deer et al. 1982).

2007 The great majority of 162 *grossular* occurrences in Supplementary Table 3 arise from contact 2008 metamorphism of calcareous rocks, often in association with *diopside* and/or *wollastonite* 2009 (Watters 1958; Reverdatto and Sóbolev 1973), or in regional metamorphic formations, also 2010 with carbonate-bearing protoliths (Tilley 1927; Sylvester and Anderson 1976). In addition, 28 occurrences of the Ca-Fe<sup>3+</sup> garnet *andradite* arise predominantly from contact and regional 2011 2012 metamorphism of calc-silicate rocks (Harker 1950; White 1959; Shedlock and Essene 1979). In 2013 several instances, contact metamorphic garnets have so-called "grandite" compositions 2014 intermediate between grossular and andradite (Coombs et al. 1977; Tulloch 1979).

2015	Three additional Ca-Mg orthosilicates, bredigite $[Ca_7Mg(SiO_4)_4]$ with 6 occurrences (Tilley
2016	and Vincent 1948; Grapes 2006), <i>larnite</i> (Ca <sub>2</sub> SiO <sub>4</sub> ) with 24 occurrences (Deer et al. 1986), and
2017	merwinite $[Ca_3Mg(SiO_4)_2]$ with 38 occurrences (Larsen and Foshag 1921; Reverdatto and
2018	Sóbolev 1973), are frequently found in contact metamorphosed calc-silicate protoliths, often in
2019	association with the calc-silicates melilite, rankinite, and spurrite (Joplin 1968; Deer et al. 1986;
2020	Grapes 2006). Merwinite is also reported as an ultrahigh pressure mantle phase (Zedgenizov et
2021	al. 2014).

2022 We lump three compositionally similar IMA species—humite, clinohumite, and 2023 hydroxylclinohumite—into humite  $[Mg_{7-9}(SiO_4)_4(F,OH)_2]$ . Members of the humite group differ in 2024 the ratios of two structural modules, one of forsterite composition  $[Mg_2(SiO_4)]$  and the other of 2025 brucite composition [Mg(OH,F)<sub>2</sub>]. Reports of metamorphic mineral modes seldom distinguish 2026 between humite  $[Mg_9(SiO_4)_4(OH,F)_2]$  and clinohumite  $[Mg_7(SiO_4)_4(OH,F)_2]$ , nor between the OH-2027 and F-dominant species. We record 21 occurrences of humite, all of which are characteristic of 2028 the contact metamorphism of dolomite-bearing sediments (Tilley 1951; Joplin 1968; Deer et al. 2029 1982). Note that two other members of the humite group (Van Valkenburg 1961), norbergite 2030  $[Mg_3(SiO_4)_4(OH,F)_2]$  and *chondrodite*  $[Mg_5(SiO_4)_4(OH,F)_2]$ , are also contact metamorphic 2031 minerals, but did not appear as common phases in our tabulations of metamorphic rock modes. 2032 Three aluminosilicate  $(Al_2SiO_5)$  polymorphs, and alusite, kyanite, and sillimanite, are 2033 abundant constituents of many metapelites, with 146, 102, and 235 occurrences in 2034 Supplementary Table 3, respectively. These phases, which can coexist at their invariant triple 2035 point (approximately 500 °C and 0.4 GPa; Hodges and Spear 1982; Bohlen et al. 1991; Pattison 2036 2001), have received special attention for their ability to document the pressure-temperature

regimes of their host rocks (Barrow 1893; Zen 1969; Deer et al. 1982; Whitney 2002; Philpotts and Ague 2009). *Sillimanite*, the high-temperature, low-pressure polymorph, is a common phase in a variety of metapelites subjected to hornblende hornfels, granulite, and pyrometamorphic (sanidinite) conditions (Reverdatto and Sóbolev 1973; Botha 1983; Grapes 2040 *Descriptional Conditions* (Reverdatto and Sóbolev 1973; Botha 1983; Grapes 2041 2006). *Mullite* [Al<sub>4+2x</sub>Si<sub>2-2x</sub>O<sub>10-x</sub> (x  $\approx$  0.4)] is also a high-temperature, low-pressure orthosilicate that we record from 62 pyrometamorphic rocks, often in association with *sillimanite* (Grapes

2043 2006).

2044 *Andalusite* forms in pelitic protoliths at low pressure and moderate temperature (< 770 °C), 2045 notably from albite-epidote hornfels and hornblende hornfels facies (Read 1923; Guitard 1965;

2046 Reverdatto and Sóbolev 1973). *Kyanite*, the highest-pressure crustal polymorph of Al<sub>2</sub>SiO<sub>5</sub>, is 2047 frequently encountered in regional and high-pressure metamorphic rocks with aluminous 2048 precursors (Carswell 1990; Carswell and Compagnoni 2003). The aluminosilicates may record 2049 either prograde or retrograde metamorphism. For example, Lal (1969) described *andalusite* and 2050 *kyanite* formed via retrograde metamorphism from cordierite-bearing rocks, and Gates and

2051 Speer (2022) record retrograde *kyanite* after *sillimanite* in metapelite shear zones.

2052 *Chloritoid*  $[(Fe^{2+},Mg,Mn^{2+})Al_2O(SiO_4)(OH)_2]$ , with 46 occurrences in our tabulation, is most 2053 commonly formed by regional or high-pressure metamorphism of pelitic rocks (Joplin 1968; 2054 Carswell 1990). We lump three IMA-CNMNC-approved species, chloritoid  $[Fe^{2+}Al_2O(SiO_4)(OH)_2]$ , 2055 magnesiochloritoid  $[MgAl_2O(SiO_4)(OH)_2]$ , and ottrelite  $[Mn^{2+}Al_2O(SiO_4)(OH)_2]$ , because they 2056 form a continuous solid solution and they are rarely differentiated in reports of metamorphic 2057 rock modes. The broad pressure-temperature stability field of *chloritoid* leads to a wide range

2058 of assemblages, from low-grade, clay-mineral- and *phengite*-bearing facies to high-grade rocks

with *kyanite*, *pyrope-almandine*, and/or *staurolite* (Halferdahl 1961).

Staurolite [(Fe<sup>2+</sup>,Mg)<sub>2</sub>Al<sub>9</sub>Si<sub>4</sub>O<sub>23</sub>(OH)] is another common phase derived by regional or highpressure metamorphism of pelitic sediments. Its 52 occurrences in Supplementary Table 3 reflect a range of *P-T* conditions of formation, from low-grade assemblages with *chloritoid* and *quartz*, medium-grade assemblages with *almandine* and *kyanite*, and high-grade assemblages with *sillimanite* and *plagioclase* (Deer et al. 1982; Augustithus 1985; Carswell 1990). *Staurolite* is also observed in the contact metamorphism of pelites (Reverdatto and Sóbolev 1973; Grapes 2006).

2067 *Titanite* (CaTiSiO<sub>5</sub>; commonly reported as "sphene") occurs as a minor phase in 186 2068 metamorphic rocks in our survey in a variety of contexts (Harker 1950; Joplin 1968; Reverdatto 2069 and Sóbolev 1973; Carswell 1990). Zircon (ZrSiO<sub>4</sub>), another volumetrically minor phase, is listed 2070 in 66 of 2785 rocks in our tabulation, including a wide range of contact and regional 2071 metamorphic lithologies (Joplin 1968; Carswell 1990; Grapes 2006). Titanite and zircon are 2072 particularly durable accessory minerals that are widespread in igneous and sedimentary 2073 formations; therefore, their occurence in metamorphic rocks sometimes derives from protolith 2074 minerals that have been little altered.

Though not among the more common metamorphic orthosilicates, *willemite* (Zn<sub>2</sub>SiO<sub>4</sub>) is an important mineral in some metamorphosed Pb-Zn deposits, such as the sillimanite-grade deposits at Franklin, New Jersey (Pinger 1950; Frondel 1990). In this case, which may be relevant to minerals in many metamorphic environments, *willemite* occurs both as a major phase in the ore and as a secondary phase in thin hydrothermal veins. These two generations of

willemite, furthermore, have distinct properties: both forms are fluorescent, but only the
 secondary willemite has persistent luminescence as a consequence of its greater arsenic
 content (Rakovan and Waychunas 1996). With their distinct modes of formation and attributes,
 these coexisting forms of willemite represent two different mineral kinds in our evolutionary
 system.

2085

### 2086 Sorosilicates or Disilicates

Sorosilicates incorporate the double-tetrahedron pyrosilicate group ( $Si_2O_7^{6^-}$ ). We find eight sorosilicate root natural kinds, corresponding to at least 35 IMA-CNMNC-approved species, among the 94 most frequently encountered metamorphic mineral kinds. All of these phases, in addition to the disilicate-carbonate mineral *tilleyite* described earlier, are calcium-bearing minerals that occur most frequently in the contact metamorphic zones of limestone and dolomite.

2093 The most common metamorphic sorosilicates are from the diverse epidote group (Deer et al. 1986; Armbruster et al. 2006). We lump 4 monoclinic species (including Fe<sup>3+</sup>-bearing 2094 clinozoisite) into *epidote* [Ca<sub>2</sub>(Al<sub>2</sub>Fe<sup>3+</sup>)[Si<sub>2</sub>O<sub>7</sub>][SiO<sub>4</sub>]O(OH)] with 149 occurrences; 5 rare-earth 2095 2096 element-bearing epidote group minerals into *allanite* [(CaCe)(AIAIFe<sup>2+</sup>)O[Si<sub>2</sub>O<sub>7</sub>][SiO<sub>4</sub>](OH)] with 2097 16 (though certainly under-reported); occurrences and orthorhombic zoisite 2098 [Ca<sub>2</sub>Al<sub>3</sub>[Si<sub>2</sub>O<sub>7</sub>][SiO<sub>4</sub>]O(OH)] with 156 occurrences. In addition, Mn-bearing *piemontite* 2099  $[Ca_2Al_2Mn^{3+}(Si_2O_7)(SiO_4)O(OH)]$  is an important phase in metamorphosed manganese deposits. 2100 though we record only 7 occurrences. Metamorphic epidote is found most commonly via 2101 contact metamorphism of carbonate-bearing sediments and mafic igneous rocks (Joplin 1968;

2102 Reverdatto and Sóbolev 1973), but also in regional (Harker 1950), high-pressure (Carswell 2103 1990), and xenolith (Grapes 2006) contexts. Note, however, that it may be difficult to 2104 distinguish metamorphic *epidote* and *zoisite* (see below) from examples formed by 2105 metasomatism (Joplin 1968).

2106 *Zoisite* generally forms at lower metamorphic grades than *epidote*, though it can coexist with

2107 epidote in medium-grade regional metamorphic rocks derived from calcareous sediments or

2108 mafic igneous rocks (Myer 1966; Ackermand and Rasse 1973; Raith 1976). Zoisite is also

2109 common in *kyanite*-bearing ecologite (Carswell and Compagnoni 2003), where it may form by a

2110 prograde reaction from *lawsonite* (Deer et al. 1986).

2111 We lump two species, lawsonite and itoigawaite, into the root kind *lawsonite* 2112  $[(Ca,Sr)Al_2(Si_2O_7)(OH)_2 H_2O]$ , with 11 occurrences in Supplementary Table 3. *Lawsonite* forms 2113 exclusively in high-pressure blueschist or eclogite facies rocks (Philpotts and Ague 2009), 2114 commonly in association with *glaucophane* and an epidote group mineral, either *zoisite* or 2115 *epidote* (Coleman et al. 1965; Carswell 1990).

2116 *Rankinite* (Ca<sub>3</sub>Si<sub>2</sub>O<sub>7</sub>), with 20 occurrences, is found exclusively in contact metamorphic rocks,

2117 commonly in association with *larnite, melilite, spurrite,* and/or *wollastonite* (Reverdatto and
2118 Sóbolev 1973; Grapes 2006).

The melilite group includes the solid solution between åkermanite  $[Ca_2(Al_2SiO_7)]$  and gehlenite  $[Ca_2(MgSi_2O_7)]$ , as well as alumoåkermanite  $[(Ca,Na)_2(Al,Mg,Fe^{2+})(Si_2O_7)]$ —minerals that we lump into the root mineral kind *melilite*. With 108 occurrences in Supplementary Table 3, *melilite* is a common mineral in pyrometamorphosed siliceous limestone and dolomite,

2123 particularly at pyroxene hornfels and sanidinite facies (Reverdatto and Sóbolev 1973; Grapes

2124 2006), often forming at the expense of *diopside* or *anorthite* (Bowen 1940; Reverdatto 1970).

2125 *Pumpellyite* [Ca<sub>2</sub>(Al,Fe<sup>2+</sup>,Fe<sup>3+</sup>)<sub>3</sub>(Si<sub>2</sub>O<sub>7</sub>)(SiO<sub>4</sub>)(OH,O)<sub>2</sub>·H<sub>2</sub>O] lumps 9 closely-related species of 2126 Ca-Al-Fe sorosilicates that are found most frequently in the low-grade zeolite and pumpellyite-2127 prehnite facies of regional metamorphism. We list 10 occurrences, all of which occur in low-2128 grade metapelites (Joplin 1968; Botha 1983; Augustithus 1985). However, Deer et al. (1986) 2129 note that Al-rich *pumpellyite* also occurs in blueschist facies, and Fe- and Mn-rich pumpellyite

2130 may occur in mineralized skarn zones.

2131 We lump 10 IMA-CNMNC-approved species, most of which are rare compositional variants,

into vesuvianite [(Ca,Na)<sub>19</sub>(Al,Mg,Fe)<sub>13</sub>(SiO<sub>4</sub>)<sub>10</sub>(Si<sub>2</sub>O<sub>7</sub>)<sub>4</sub>(OH,F,O)<sub>10</sub>]. We record 15 occurrences in

2133 contact metamorphosed limestone, in which it is a characteristic skarn mineral, commonly in

association with *diopside*, *grossular*, and/or *wollastonite* (Harker 1950). *Vesuvianite* occurs less

commonly in regional metamorphosed limestones (Tilley 1927; Deer et al. 1982). Note that, as with the example of *epidote*, it may be difficult to distinguish *vesuvianite* formed by

2137 metamorphism versus metasomatism (Joplin 1968).

2138

# 2139 Cyclosilicates

2140 Members of the cordierite, tourmaline, and osulmilite groups are relatively common 2141 metamorphic cyclosilicates. We lump the species cordierite (with Mg) and sekaninaite (with 2142 Fe<sup>2+</sup>) into the root mineral kind *cordierite* [(Mg,Fe<sup>2+</sup>)<sub>2</sub>Al<sub>4</sub>Si<sub>5</sub>O<sub>18</sub>], which, with 395 occurrences in 2143 Supplementary Table 3, is among the most common minerals in contact and regionally 2144 metamorphosed pelites (Joplin 1968; Reverdatto and Sóbolev 1973; Botha 1983; Grapes 2006).

2145 Deer et al. (1986) detail a wide range of *cordierite* parageneses, including pyrometamorphosed 2146 xenoliths, contact metamorphosed argillaceous sediments, and a range of regional 2147 metamorphic facies, including low-pressure, high-temperature assemblages with *andalusite*; 2148 moderate-pressure assemblages with *sillimanite* and garnet; and high-pressure assemblages 2149 with *kyanite*.

2150 *Tourmaline* [(Na,Ca, $\Box$ )(Mg,Al,Fe<sup>2+</sup>,Fe<sup>3+</sup>,Ti<sup>4+</sup>)<sub>3</sub>(Al,Fe<sup>3+</sup>,Mg)<sub>6</sub>(Si<sub>6</sub>O<sub>18</sub>)(BO<sub>3</sub>)<sub>3</sub>(OH)<sub>3</sub>(O,F)] is the only

2152 Henry and Dutrow 2012). We lump 18 IMA-CNMNC-approved species of the tourmaline group

common boron-bearing mineral in metamorphic rocks (Deer et al. 1986; Henry et al. 2011;

2153 (Supplementary Table 1), all of which have been reported from regional metamorphic

2154 environments. The 47 *tourmaline* occurrences listed in Supplementary Table 3, including

2155 several examples of *tourmaline*-dominant tourmalinites, are from metapelites (Harker 1950;

2156 Joplin 1968). Joplin (1968) suggests that metamorphic tourmaline occurs in 3 distinct ways: as a

2157 remnant mineral of the protolith, through metamorphism of a borate-containing lithology, or as

2158 the result of boron metasomatism.

2159 Osumilite  $[(K,Na)(Fe^{2+},Mg)_2(AI,Fe^{3+})_3(Si,AI)_{12}O_{30}]$  was reported in 13 of our mineral modes,

2160 typoically from ultrahigh temperature metamorphosed pelites, in which it commonly occurs

2161 with cordierite, orthopyroxene, sanidine, and/or sillimanite (Harley 2021).

2162

2151

# 2163 Inosilicates

Among the 94 relatively common metamorphic minerals listed in Table 2, 16 are chain silicates, including several members of the pyroxene (7 kinds) and amphibole (7 kinds) groups

2166 (Deer et al. 1997a, 1997b). Inosilicates are one of the most common classes of metamorphic

2167 minerals, occurring in 1387 (50 %) of the 2785 metamorphic rocks in Supplementary Table 3.

2168

2169 <u>Pyroxene Group:</u> We consider 7 root kinds of pyroxene group single-chain silicates, most of

2170 which lie in or near the  $[(Ca,Mg,Fe)_2Si_2O_6]$  quadrilateral (Deer et al. 1997a).

2171 *Orthopyroxene* lumps 281 occurrences of orthorhombic pyroxenes, most often described as

2172 enstatite (the Mg end-member) or "hypersthene" (with Mg ~ Fe<sup>2+</sup>) but sometimes "bronzite"

2173 (with Mg > Fe<sup>2+</sup>) or ferrosilite (the Fe<sup>2+</sup> end-member), always lying close to the MgSiO<sub>3</sub>—

2174 Fe<sup>2+</sup>SiO<sub>3</sub> binary. *Orthopyroxene* most often occurs in granulite, ecologite, and UHT facies of

2175 metamorphosed ultramafic and mafic igneous rocks, in which it is often the most abundant

2176 mafic phase (Joplin 1968; Augustithus 1985; Carswell and Compagnoni 2003). It also occurs in

2177 lower-grade regional metamorphic rocks (Joplin 1968; Botha 1983), with iron-rich examples in

2178 metamorphosed iron formations (Kranck 1961; Simmons et al. 1974). *Orthopyroxene* is not 2179 uncommon in contact metamorphic environments, including pyrometamorphosed xenoliths

2180 (Reverdatto and Sóbolev 1973; Grapes 2006). In some instances, Mg-rich *orthopyroxene* is

associated with carbonate minerals (Schreyer et al. 1972; Ohnmacht 1974).

Three Ca-bearing clinopyroxenes, *diopside*  $[Ca(Mg,Fe^{2+})Si_2O_6;$  409 occurrences in Supplementary Table 3), *hedenbergite* (CaFe<sup>2+</sup>Si<sub>2</sub>O<sub>6</sub>; 16 occurrences), and the ternary solid solution *augite*  $[(Ca,Mg,Fe^{2+})_2Si_2O_6,$  typically with 0.5 < Ca/(Mg+Fe) < 0.9; 150 occurrences], are common in a wide variety of metamorphic rocks (Deer et al. 1997a). Pyroxenes close to the continuous CaMg—CaFe<sup>2+</sup> solid solution between *diopside* and *hedenbergite* are most typical of thermally metamorphosed carbonate and calc-silicate rocks, occurring in xenolithes and

contact metamorphic contexts (Grapes 2006). We define *diopside* broadly to include most
intermediate Mg-Fe compositions (e.g., "salite" and "ferrosalite"), as well as Al-bearing
"fassaite." More than 80% of occurrences of *diopside*, the most abundant pyroxene in our
survey, arise from contact metamorphism. Diopside also occurs in regional and high-pressure
metamorphic rocks, with several examples from amphibolite facies (Harker 1950; Augustithus
1985) and eclogite facies (Carswell 1990; Carswell and Compagnoni 2003). *Hedenbergite* displays much the same parageneses as *diopside*, but with Fe-rich protoliths

(Joplin 1968; Augustithus 1985). We debated whether to lump these two end-members, but *hedenbergite* appears to form a discrete cluster of metamorphic clinopyroxenes with low Mg.
Cluster analysis of igneous and metamorphic clinopyroxenes represents an important future
research goal.

2199 The IMA-CNMNC-approved species augite, including clinopyroxenes in the  $[(Ca,Mg,Fe^{2+})_2Si_2O_6]$  system with Ca occupying between ~25 and ~45 atom percent of the Ca-2200 2201 Mg-Fe sites, is equivalent to our root kind *augite*. The 150 occurrences in Supplementary Table 2202 3, while mostly from contact metamorphism or pyrometamorphism of ultramafic/mafic 2203 lithologies (Reverdatto and Sóbolev 1973; Grapes 2006), also include representatives derived 2204 by regional metamorphism of mafic and intermediate igneous protoliths (Harker 1950; Joplin 2205 1968; Ferry 1987).

Three Na-bearing clinopyroxenes, *Aegirine* [(Ca,Na)(Fe<sup>3+</sup>,Mg,Fe<sup>2+</sup>)Si<sub>2</sub>O<sub>6</sub>; 16 occurrences], *jadeite* (NaAlSi<sub>2</sub>O<sub>6</sub>; 34 occurrences), and *omphacite* [(Ca,Na)(Mg,Fe,Al)Si<sub>2</sub>O<sub>6</sub>; 151 occurrences] are especially characteristic of high-pressure metamorphic environments. *Aegirine*, in which we lump two IMA-CNMNC-approved species aegirine (NaFe<sup>3+</sup>Si<sub>2</sub>O<sub>6</sub>) and aegirine-augite

[(Ca,Na)(Fe<sup>3+</sup>,Mg,Fe<sup>2+</sup>)Si<sub>2</sub>O<sub>6</sub>], is the least common of these phases in metamorphic rocks, being found primarily in the context of mafic and intermediate igneous rocks subjected to ultrahigh pressure and ecologite facies (Carswell 1990), though *aegirine* also occurs via contact metamorphism of alkaline rocks (Reverdatto and Sóbolev 1973). A complication is the formation of *aegirine* through sodium metasomatism of prior pyroxenes (Moore 1973; Deer et al. 1997a). *Jadeite* is a diagnostic phase found exclusively in high-pressure metamorphic environments,

2217 including blueschist facies, eclogite facies, and ultrahigh pressure metamorphic rocks (Carswell

1990). It often forms through the iconic reaction *albite*  $\rightarrow$  *jadeite* + *quartz* (Deer et al. 1997a,

2219 and references therein). Jadeite commonly incorporates up to 15 mol % of an

2220 aegirine/omphacite component; however, a significant compositional gap separates jadeite

from these phases, which often coexist in high-pressure assemblages (Coleman and Clark 1968).

2222 Omphacite, which represents a solid solution among aegirine, diopside, and jadeite, is a

2223 relatively common phase in high-pressure metamorphic rocks. All 151 occurrences in our

2224 tabulation were reported from blueschist, eclogite, or ultrahigh-pressure environments, most

2225 often with ultramafic or mafic protoliths and often in association with glaucophane,

2226 pyrope/almandine, and quartz/coesite (Carswell 1990; Carswell and Compagnoni 2003).

2227

2228 <u>Pyroxenoid Group:</u> Two members of the inosilicate pyroxenoid group, *wollastonite* (CaSiO<sub>3</sub>; 2229 158 occurrences) and the Mn-bearing *rhodonite* [CaMn<sub>3</sub>Mn(Si<sub>5</sub>O<sub>15</sub>); 8 occurrences, not listed 2230 among the top 94 phases], are most commonly associated with skarn zones. Almost all

2231 *wollastonite* reports are from carbonate or calc-silicate protoliths subjected to pyroxene

hornfels or sanidinite facies metamorphism (Reverdatto and Sóbolev 1973; Grapes 2006).

2233 *Rhodonite*, which lumps 4 closely-related species of Mn pyroxenoids, is most frequently 2234 encountered in the high-pressure metamorphic environments of Mn-rich protoliths (Carswell 2235 1990), notably by reaction of *rhodochrosite* (MnCO<sub>3</sub>; Hori 1962), though it can also form 2236 through Mn metasomatism (Bilgrami 1956).

2237

2238 Amphibole Group: The amphibole group of double-chain silicates, which boasts more than 2239 110 IMA-CNMNC-approved species (https://rruff.info/ima; accessed January 13, 2023), is likely 2240 the most diverse of all mineral structure types (Deer et al 1997b; Hawthorne et al. 2011). Here, 2241 we provisionally lump 55 amphibole species known to occur in metamorphic rocks into 7 root 2242 mineral kinds. It should be noted, however, that the variety of amphibole parageneses, coupled 2243 with the extensive and complex solid solutions and miscibility gaps among many species, render 2244 any suggestion of amphibole mineral kinds tentative, at best. In the context of metamorphism, 2245 we have yet to determine if different facies, different protoliths, effects of metasomatism, 2246 prograde versus retrograde formation, and other factors may yield numerous distinct 2247 combinations of paragenesis and attributes. We require data resources with analyses of tens of 2248 thousands of well-characterized amphibole specimens, coupled with advanced methods of 2249 cluster analysis (Boujibar et al. 2021; Hystad et al. 2021). Such an epic endeavor could represent 2250 a lifetime of fruitful study for an ambitious young mineralogist.

2251 Anthophyllite  $[\Box(Mg,Fe^{2+})_2(Mg,Fe^{2+},Fe^{3+},AI)_5(Si,AI)_8O_{22}(OH)_2;$  with 52 occurrences in 2252 Supplementary Table 3], lumps 6 species of the complex anthophyllite/ferro-

2253 anthophyllite/gedrite/ferro-gedrite solid solution of orthorhombic amphiboles (Ferré 1989). An 2254 unresolved question regards the possible presence of a miscibility gap in this system between 2255 Al-rich (at times with Na) and Al-poor orthoamphiboles (Hawthorne et al. 1980; Spear 1982). If 2256 so, then at least two root mineral kinds would be warranted. Most of the examples in our 2257 compilation arise from hornblende-hornfels or pyroxene-hornfels facies contact metamorphism 2258 of ultramafic/mafic igneous rocks or pelitic sediments, often in association with biotite, 2259 cordierite, and quartz (Reverdatto and Sóbolev 1973). We also record a number of instances of 2260 anthophyllite in regional metamorphic settings, including amphibolite and granulite facies 2261 metamorphism of pelites and ultramafic rocks (Joplin 1968).

2262 The closely-related clinopyroxenes *cummingtonite*  $[\Box Mg_2Mg_5Si_8O_{22}(OH)_2;$  with 12 occurrences] and *grunerite* [ $\Box$ Fe<sup>2+</sup><sub>2</sub>Fe<sup>2+</sup><sub>5</sub>Si<sub>8</sub>O<sub>22</sub>(OH)<sub>2</sub>; with 8 occurrences, hence not listed among 2263 2264 the top 94] are the monoclinic polymorphs of anthophyllite and ferro-anthophyllite. All but one 2265 of the *cummingtonite* examples in our tabulation, with Mg/(Mg+Fe) generally > 0.4 (i.e., in 2266 some instances with Fe > Mg), are from metapelites subjected to hornblende-hornfels facies 2267 contact metamorphism (Reverdatto and Sóbolev 1973). The more iron-rich grunerite examples, 2268 by contrast, are primarily from amphibolite or granulite facies regionally metamorphosed iron 2269 formations (Joplin 1968; Kimball and Spear 1984). Therefore, we provisionally distinguish these 2270 two closely-related mineral kinds based on paragenetic mode, even though they may display continuous solid solution between the Mg and  $Fe^{2+}$  end-members. It should be noted that as a 2271 2272 result of miscibility gaps, cummingtonite and grunerite often occur in assemblages with multiple amphiboles, including calcic hornblende, Al-bearing anthophyllite, and/or a sodic 2273 2274 amphibole (Deer et al. 1997b).

2275 Several calcic clinoamphiboles are common constituents of metamorphic rocks. Tremolite  $[\Box Ca_2(Mg_{5.0-4.5}Fe^{2+}_{0.0-0.5})Si_8O_{22}(OH,F)_2;$  with 122 occurrences] is typically an almost pure Ca-Mg 2276 phase (i.e., low Fe<sup>2+</sup>) formed from ultramafic/mafic igneous or calc-silicate sediments in 2277 2278 contact, regional, and high-pressure metamorphic environments. Most of the examples in 2279 Supplementary Table 3 are from muscovite-hornfels, hornblende-hornfels, or pyroxene-2280 hornfels contact metamorphic zones, in which tremolite is associated with diopside, dolomite, 2281 grossular, talc, and/or other Ca-Mg phases (Reverdatto and Sóbolev 1973). We lump the OH-2282 and F-bearing species, which display a complete solid solution and share the same paragenesis. Actinolite  $[\Box Ca_2(Mg, Fe^{2+})_5Si_8O_{22}(OH, F)_2;$  with 74 occurrences] lumps the species actinolite 2283 and ferro-actinolite, spanning a range  $0.9 > Mg/(Mg + Fe^{2+}) > 0$  (Deer et al. 1997b). Though 2284 2285 chemically and structurally similar to tremolite, actinolite is distinguished both by its greater 2286 Fe<sup>2+</sup> content and by its common association in metapelites or metabasites with *biotite*, *epidote* 2287 or zoisite, and/or quartz in high-pressure, regional, or contact metamorphic settings (Harker 2288 1950; Joplin 1968; Botha 1983; Carswell 1990).

2289 We lump 26 IMA-CNMNC-approved metamorphic species of Ca-(+/-Na,K)-clinoamphiboles into *hornblende* [(Na,K)Ca<sub>2</sub>(Mg,Fe<sup>2+</sup>,Al,Fe<sup>3+</sup>)<sub>5</sub>(Si,Al)<sub>8</sub>O<sub>22</sub>(OH,F,Cl)<sub>2</sub>; with 387 occurrences]. This 2290 2291 complex group displays significant compositional plasticity, with solid solutions among Na, K, and vacancies in alkali sites; Mg-Fe<sup>2+</sup>-Fe<sup>3+</sup>-Al in octahedral sites; and Al-Fe<sup>3+</sup>-Si in tetrahedral 2292 2293 sites, as well as among OH, F, and Cl (Deer et al. 1997b; Hawthorne et al. 2011; see 2294 Supplementary Tables 1 and 2). Hornblende, a defining phase in hornblende-hornfels and 2295 amphibolite facies rocks, appears in numerous metamorphic environments, including contact 2296 metamorphism (albite-epidote to sanidinite facies; Reverdatto and Sóbolev 1973; Grapes

2297 2006), regional metamorphism (greenschist to granulite facies; Harker 1950; Joplin 1968), and 2298 high-pressure metamorphism (blueschist to eclogite facies; Reverdatto and Sóbolev 1973; 2299 Carswell 1990). Hornblende protoliths, similarly, span a wide range of igneous, sedimentary, 2300 and metamorphic rocks. Hornblende in metamorphic rocks commonly coexists with other 2301 amphiboles, including anthophyllite, cummingtonite, and grunerite (Deer et al. 1997b). Given 2302 the complexity of this mineral group, cluster analyses of numerous hornblende samples based 2303 on composition, paragenesis, and mineral associations, would doubtless reveal many distinct 2304 kinds of *hornblende*.

2305 We lump a wide range of Na-Ca clinoamphiboles into richterite, defined here as  $[(\Box, Na)(NaCa)(Mg, Fe^{2+}, Al, Fe^{3+})_{5}(Si, Al, Fe^{3+})_{8}O_{22}(OH)_{2}]$ . Although we record only 12 occurrences 2306 2307 in Supplementary Table 3, most of which were originally described as barroisite [nominally 2308  $(\Box NaCa)(Mg_3Al_2)(Si_7Al)O_{22}(OH)_2]$  or winchite  $[(\Box NaCa)(Mg_4Al)Si_8O_{22}(OH)_2]$ , we lump 16 IMA-2309 CNMNC-approved species into the root natural kind *richterite*, while acknowledging that much 2310 more work is needed to fully characterize these minerals and their associated parageneses. 2311 Most of the *richterite* occurrences that we record are metamorphosed mafic rocks from 2312 eclogite facies, almost always in association with *omphacite*, pyrope, and rutile (Binns 1967; 2313 Carswell 1990), though it is reported from regionally metamorphosed basalt, as well (Iwasaki 2314 1960).

The sodium amphibole *glaucophane*  $[\Box Na_2(Mg,Fe^{2+})_3Al_2Si_8O_{22}(OH)_2;$  with 79 occurrences] lumps 2 species, glaucophane and ferro-glaucophane, which form a Mg-Fe<sup>2+</sup> solid solution. All of the examples in Supplementary Table 3 are from high-pressure metamorphism (most

2318 blueschist or eclogite facies, but also ultrahigh-pressure facies) of commonly 2319 mafic/intermediate igneous rocks or Mg-bearing sediments (Augustithus 1985; Carswell 1990). One additional inosilicate group, sapphirine  $[(Mg, Fe^{2+}, Al, Fe^{3+})_8O_2(Al, Si)_6O_{18}, 28 \text{ occurrences}],$ 2320 2321 is important as a key indicator of the temperature (> 900 °C) of ultrahigh temperature 2322 metamorphic rocks formed from ultramafic protoliths (Monchoux 1972; Deer et al. 1997a; 2323 Carswell and Compagnoni 2003; Harley 2021). It commonly occurs in association with 2324 orthopyroxene and sillimanite.

2325

# 2326 **Phyllosilicates**

2327 *Mica Group:* With 1244 occurrences in Supplementary Table 3 (45% of the 2795 rocks 2328 surveyed), the mica minerals are prominent constituents of many metamorphic lithologies 2329 (Guidotti 1984; Fleet 2003). We lump 15 IMA-CNMNC-approved species into five root mineral 2330 kinds of micas: biotite, phlogopite, phengite, muscovite, and paragonite. However, there 2331 undoubtedly exist many more kinds of metamorphic micas, the identification of which will 2332 require the construction of extensive mica databases and application of cluster analysis (Hazen 2333 2019). In paticular, widely occurring metamorphic *biotite* and *muscovite* will likely be split into 2334 multiple natural kinds.

We define the familiar group of dark-colored, Fe<sup>2+</sup>-bearing trioctahedral mica species as biotite [KFe<sup>2+</sup><sub>2</sub>(Fe<sup>2+</sup>,Mg,Mn<sup>2+</sup>)(Si,Al,Fe<sup>3+</sup>)<sub>2</sub>Si<sub>2</sub>O<sub>10</sub>(OH,F,Cl)<sub>2</sub>; 822 occurrences]. We lump 6 IMA-CNMNC-approved species, which are themselves rarely identified in the petrographic literature. Few minerals occur in as a diverse array of metamorphic environments as *biotite*, which we record from low-pressure pyrometamorphic, contact, regional, and high-pressure
2340 metamorphism, typically of pelites, but also of ultramafic, mafic, intermediate, acidic, and 2341 (rarely) agpaitic igneous rocks, as well as Fe-bearing impure carbonate and calc-silicate 2342 protoliths (Harker 1950; Joplin 1968; Reverdatto and Sóbolev 1973; Botha 1983; Carswell 1990; 2343 Fleet 2003; Grapes 2006).

2344 We also lump 6 Mg-dominant species of trioctahedral micas, which are generally lighter in color than *biotite*, as *phlogopite*  $[KMg_2(Mg,Fe^{2+},Mn^{2+},Fe^{3+},Ti^{4+})(Si,Al,Fe^{3+})_2Si_2O_{10}(OH,F)_2;$  with 78 2345 2346 occurrences]. Though often combined with *biotite* in some descriptions of mica (e.g., Fleet 2347 2003), and consequently sometimes reported as *biotite* in the petrologic literature, *phlogopite* 2348 displays distinct mineral associations in metamorphosed high-Mg, low-Fe environments, 2349 including ultramafic and dolomitic carbonate protoliths (Joplin 1968; Reverdatto and Sóbolev 2350 1973). Owing to their diverse parageneses and compositional range, trioctahedral micas 2351 represent yet another mineral group that is ripe for investigation by cluster analysis.

The dioctahedral aluminous mica *muscovite*  $[K(AI, Fe^{3+}, Cr)_2(Si_3AI)O_{10}(OH)_2; 515$  occurrences], 2352 2353 commonly reported with the varietal names illite, phengite, or sericite, is most characteristic of 2354 regionally and contact metamorphosed pelites (Reverdatto and Sóbolev 1973; Philpotts and 2355 Ague 2009), which represent most of the occurrences recorded in Supplementary Table 3. 2356 Muscovite, among the first phases to form during diagenesis of clay minerals (Fleet 2003), also 2357 occurs in a wide range of other contexts, including contact and regional metamorphosed 2358 arkosic, calc-silicate, and impure carbonate sediments (Philpotts and Ague 2009), as well as a 2359 variety of igneous protoliths (Harker 1950; Carswell 1990).

2360 We also include the fine-grained, Si-rich white mica *phengite* (110 occurrences) as a separate 2361 kind, even though it falls under IMA's definition of muscovite. Phengite is most commonly

associated with high-pressure metamorphism (Carswell 1990).

2363 The sodium trioctahedral mica *paragonite* [NaAl<sub>2</sub>(Si<sub>3</sub>Al)O<sub>10</sub>(OH)<sub>2</sub>; 67 occurrences] is 2364 characteristic of high-pressure eclogite facies metamorphism of mafic igneous rocks, often in 2365 association with glaucophane, kyanite, omphacite, and/or pyrope (Carswell 1990). Paragonite, 2366 often intimately intermixed with phengite (with which it has limited solid solution; e.g., 2367 Thompson and Thompson 1976; Guidotti et al. 1994), also occurs in low- and medium-grade 2368 metapelites, in which it can form by both prograde and retrograde reactions (Chatterjee 1970; 2369 Guidotti 1984; Guidotti and Sassi 1998; Fleet 2003). Paragonite also frequently co-occurs with 2370 the so-called "brittle mica" margarite  $[CaAl_2(Si_2Al_2)O_{10}(OH)_2]$  in a wide range of metamorphic 2371 grades of metasediments (Guidotti 1984; Fleet 2003); however, margarite is relatively rare in 2372 comparison to the micas described above, being reported from only one of the metamorphic 2373 rocks in our survey (Carswell and Compagnoni 2003, their Table 2).

2374

2375 <u>Other phyllosilicates:</u> More than 30 other IMA-CNMNC-approved layer silicates occur in 2376 metamorphic rocks (Supplementary Table 1). Most of these minerals (e.g., apophyllite group, 2377 *gillespite, pyrophyllte, stilpnomelane, zussmanite*) occur only rarely in metamorphic rocks. Note 2378 that we do not list diagenetically-formed clay minerals as metamorphic phases (Wilson 2013); 2379 they will be considered further in Part IX of this series. However, *chlorite, prehnite, serpentine,* 2380 and *talc* are included in our list of 94 relatively common metamorphic phases (Deer et al. 2009).

*Chlorite* [(Mg,Fe<sup>2+</sup>)<sub>5</sub>(Al,Fe<sup>3+</sup>)(Si<sub>3</sub>AlO<sub>10</sub>)(OH)<sub>8</sub>; 339 occurrences] encompasses 3 IMA-CNMNC-2381 approved species of Mg-Fe<sup>2+</sup>-Al-(Fe<sup>3+</sup>) layer silicates: chamosite, clinochlore, and sudoite. 2382 2383 *Chlorite* is common in pelites subjected to greenschist and amphibolite facies metamorphism 2384 (Harker 1950: Joplin 1968: Botha 1983), as well as from muscovite-, hornblende-, and pyroxene-2385 hornfels facies contact metamorphism of pelites and basic igneous rocks (Joplin 1968; 2386 Reverdatto and Sóbolev 1973), often in association with albite, biotite, muscovite, and/or 2387 auartz. In addition, Coleman et al. (1965) and Carswell (1990) record more than a dozen 2388 examples of *chlorite* in eclogite facies high-pressure metamorphism. *Chlorite* forms through a 2389 variety of pathways, including prograde and retrograde metamorphism, both with and without 2390 external aqueous fluids. As such, chlorite represents a mineral whose often uncertain 2391 parageneses grade continuously from regional or "burial" metamorphism to metasomatism to 2392 hydrothermal alteration (Deer et al. 2009).

2393 We lump 5 IMA-CNMNC-approved species, including three structural variants of Mg<sub>3</sub>Si<sub>2</sub>O<sub>5</sub>(OH)<sub>4</sub> (antigorite, chrysotile, and lizardite), aluminous amesite, and Fe-bearing 2394 greenalite, into *serpentine* [(Mg,Fe<sup>2+</sup>,Al,Fe<sup>3+</sup>)<sub>3</sub>(Al,Si)Si(OH)<sub>4</sub>; 68 occurrences]. Most examples in 2395 2396 Supplementary Table 3 are from low- to moderate-grade regional metamorphism of 2397 ultramafic/mafic igneous or pelitic protoliths (Joplin 1968; Philpotts and Ague 2009), in which 2398 they form primarily by retrograde/hydrothermal reactions from olivine and Mg-rich pyroxene 2399 or by prograde metamorphism of serpentinite (Deer et al. 2009, and references therein). Given 2400 its varied modes of formation, coupled with multiple polymorphs, cluster analysis of 2401 metamorphic serpentine is warranted.

2402  $Talc [Mg_3Si_4O_{10}(OH)_2; 73 \text{ occurrences}]$  is characteristic of Mg-rich protoliths over a wide 2403 range of pressure-temperature condition. Examples include thermal metamorphism of 2404 dolomite-bearing sediments (Tilley 1948; Reverdatto and Sóbolev 1973; Augustithus 1985), 2405 high-pressure (blueschist and eclogite facies) metamorphism of basic igneous rocks (Chopin 2406 1981; Carswell 1990), and greenschist to amphibolite grade regional metamorphism of 2407 ultramafic rocks (Harker 1950; Joplin 1968). Of special note are high-pressure to ultrahigh 2408 pressure (> 0.6 GPa) talc-kyanite-(quartz/coesite) assemblages known as "whiteschists" 2409 (Schreyer 1977), which form in the Mg-Al-Si-H ("MASH") system, at times with  $P_{H_{2O}}$ 2410 approximately equal to the total pressure.

*Prehnite* [Ca<sub>2</sub>Al(Si<sub>3</sub>Al)O<sub>10</sub>(OH)<sub>2</sub>; 16 occurrences], though most familiar as a hydrothermal phase associated with zeolites in amygdaloidal basalt, is also common in the eponymous prehnite-pumpellyite facies of low-grade regional metamorphism (Coombs 1960; Philpotts and Ague 2009). *Prehnite*, often in association with *chlorite* and *quartz*, is also present occasionally in metamorphosed basic igneous and pelitic sedimentary rocks from zeolite to lower amphibolite grades (Harker 1950; Joplin 1968; Botha 1983), at times the result of retrograde reactions (Coombs 1993).

2418

## 2419 **Tectosilicates**

A wide range of framework silicates, including the silica group, feldspars, feldspathoids, and zeolites (Deer et al. 2001, 2004), occur in metamorphic rocks, with one or more examples reported in 1595 (67 %) of the 2785 rocks surveyed in Supplementary Table 3. We focus attention on 10 mineral kinds that occur most frequently.

2424

2425 <u>Silica Group</u> : Four silica group minerals—quartz, high-pressure coesite, and high
2426 temperature <i>cristobalite</i> and <i>tridymite</i> —span the entire range of metamorphic environments
occurring in all but the most Si-deficient rocks (Deer et al. 2004).
2428 <i>Quartz</i> (SiO <sub>2</sub> ), with 1353 occurrences in our study (49 % of rocks in Supplementary Table 3)
2429 is the most common metamorphic mineral. It occurs in pyrometamorphosed xenoliths, contac
2430 metamorphic rocks, metamorphosed iron-manganese formations, high-pressure and regiona
2431 metamorphic settings, metasomatized rocks, and shear zones (Supplementary Table 3 and
references therein), often by recrystallization of protolith <i>quartz</i> (Deer et al. 2004). <i>Quartz</i> i
2433 stable in all crust and upper mantle pressure-temperature regimes except above ~2.7 GPa
2434 where it transforms to <i>coesite</i> , or at low pressure above ~850 °C, where <i>cristobalite</i> an
2435 <i>tridymite</i> are the stable silica phases.
2436 <i>Coesite</i> (SiO <sub>2</sub> ; 23 occurrences) is restricted to ultrahigh pressure (> 2.7 GPa) metamorphi
2437 environments, where it is typically associated with <i>kyanite</i> , <i>omphacite</i> , and/or <i>pyrope</i> (Carswe
and Compagnoni 2003). Several reports describe <i>coesite</i> as inclusions in upper mantle phases
including <i>pyrope</i> (Chopin 1984; Schertl et al. 1991) and <i>diamond</i> (Stachel et al. 2022, and
2440 references therein).
2441 <i>Cristobalite</i> (10 occurrences) and <i>tridymite</i> (44 occurrences) are high-temperature, low
pressure polymorphs of SiO <sub>2</sub> that occur almost exclusively in sedimentary rocks that have bee
2443 thermally metamorphosed (pyroxene hornfels or sandinite facies) by basic igneous rocks (Agre

and Langley 1958; Reverdatto and Sóbolev 1973; Black 1989; Grapes 2006). These two minerals

2445 co-occur in 7 of the 10 reported rocks with *cristobalite*. *Tridymite* and *cristobalite* also are

associated in some burning coal deposits with temperatures that may exceed 1100 °C (Bustin
and Matthews 1982; Grapes 2006), and therefore are the consequence of Phanerozoic
biological precursors (to be considered in Part XII).

2449

2450 <u>*Feldspar Group:*</u> Metamorphic feldspar group minerals display compositions close to two 2451 binary solid solutions (Deer et al. 2001): the Na-Ca plagioclase feldspars and the Na-K alkali 2452 feldspars. In both instances we suggest modifications of the nomenclature approved by the

IMA-CNMNC.

In the case of the plagioclase series [(CaAl,NaSi)AlSi<sub>2</sub>O<sub>8</sub>], we identify *albite* as compositions

close to NaAlSi<sub>3</sub>O<sub>8</sub> [Na/(Na+Ca) > 0.85 and often with > 10 mol % KAlSi<sub>3</sub>O<sub>8</sub>], anorthite as close to

CaAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub> [Ca/(Ca+Na) > 0.85], and *plagioclase* as having intermediate compositions between  $^{An_{20}}$  and  $^{An_{70}}$  as valid root mineral kinds. We justify this division based on the existence of the so-called peristerite and Huttenlocker miscibility gaps between  $^{An_{2-17}}$  and  $^{An_{65-88}}$ , respectively. As a consequence, several authors have recorded coexiting *albite-plagioclase* and *plagioclase-anorthite* pairs (Evans 1964; Botha 1983).

Albite, with 177 occurrences in Supplementary Table 3, is observed in a wide range of thermal, regional, and high-presssure metamorphic environments, with both igneous and sedimentary protoliths (Harker 1950; Joplin 1968; Reverdatto and Sóbolev 1973; Philpotts and Ague 2009). *Anorthite* (78 occurrences) is more restricted in its occurrences, being found primarily as a contact metamorphic mineral derived from calc-silicate and carbonate-bearing sediments (Grapes 2006), though it is also found in regionally metamorphosed mafic and calcsilicate rocks from amphibolite to granulite facies (Joplin 1968). *Plagioclase* (809 occurrences),

like *quartz*, *albite*, and *kspar* (see below) is not a particularly diagnostic phase in metamorphic rocks because it occurs across the full spectrum of thermal, regional, and high-pressure metamorphic environments, with an equally broad range of igneous, sedimentary, and metamorphic protoliths. For a given protolith, the anorthite content of plagioclase tends to increase with metamorphic grade (Deer et al. 2001). Note that most reports of "plagioclase" in the older metamorphic literature lack compositional information; thus, some of these occurrences may fit our definitions of *albite* or *anorthite*.

2475 The alkali feldspars are complicated by the existence of three K-rich (KAlSi<sub>3</sub>O<sub>8</sub>) variants—the

2476 higher-temperature (> 500 °C) monoclinic sanidine and two lower-temperature triclinic phases,

2477 microcline and orthoclase, which are often reported as "kspar" in the literature of metamorphic

2478 petrology. An additional consideration is that alkali feldspars of intermediate compositions

often exsolve Na- and K-rich phases, typically reported as "perthite." In our study, we adopt

the name *kspar* for microcline and orthoclase, and record both *albite* and *kspar* for perthite.

2481 Sanidine (102 occurrences) is most commonly found in thermally metamorphosed rocks of

2482 pyroxene hornfels or sanidinite grade (Grapes 2006), though it also has been reported from

2483 ultrahigh pressure metamorphism of pelites (Carswell 1990; Carswell and Compagnoni 2003)

and ultrahigh temperature regimes (Harley 2021).

*Kspar* (411 occurrences) has been reported from thermal (zeolite to pyroxene hornfels facies), regional (amphibolite to granulite facies), and high-pressure (eclogite to ultrahigh pressure facies) metamorphic environments. Protoliths for *kspar* include mafic, acidic, and agpaitic igneous rocks and arkosic, pelitic, and carbonate-bearing sedimentary rocks (Harker 1950; Joplin 1968; Reverdatto and Sóbolev 1973; Carswell 1990; Deer et al. 2001).

At the temperatures of UHT metamorphism (> 900 °C), an additional complication is the occurrence of Ca-Na-K ternary feldspars, which typically exsolve to a perthite with coexisting plagioclase and alkali feldspar lamellae (Harley 2008; Harley 2021, their Figure 20).

2493

2494 <u>Scapolite Group:</u> Three species of the scapolite group are lumped as *scapolite* 2495 [(Na,Ca)<sub>4</sub>Al<sub>3</sub>(Al,Si)<sub>3</sub>Si<sub>6</sub>O<sub>24</sub>(CO<sub>3</sub>,SO<sub>4</sub>,Cl), with 33 occurrences]. *Scapolite* is not infrequently 2496 observed in medium- to high-grade contact (hornblende-hornfels and pyroxene hornfels facies) 2497 and regional (amphibolite and granulite facies) metamorphosed pelites and calc-silicate 2498 sediments (Joplin 1968; Reverdatto and Sóbolev 1973), and has also been reported from the 2499 albite-epidote hornfels facies contact metamorphism of amygdaloidal basalt (Joplin 1968).

2500

2501 Zeolite Group: The zeolite facies is the lowest pressure-temperature regime of 2502 metamorphism, with temperatures < 200 °C at pressures < 0.3 GPa (Philpotts and Ague 2009). 2503 Most zeolite minerals form via low-temperature aqueous processes, including fluid interactions 2504 with cooling basalt and authigenesis (Deer et al. 2004). Nevertheless, some zeolite occurrences 2505 are attributed to metamorphism, sensu stricto. Though not sufficiently abundant to include 2506 among the most common metamorphic phases, analcime (2 occurrences), laumontite (1), and 2507 wairakite (2), as well as 6 undifferentiated reports of "zeolite," were listed in modes of low-2508 grade metamorphosed mafic igneous rocks and pelites (Joplin 1968).

2509

2510 <u>*Feldspathoid Group:*</u> None of the members of the sub-silicic feldspathoid framework silicate 2511 group is common in metamorphic rocks. Nevertheless, *kalsilite, leucite,* and *nepheline* (with 5,

2512 2, and 4 occurrences, respectively) are representative of undersaturated pyrometamorphosed

2513 calc-silicates (Grapes 2006).

2514

2515 To these framework silicates, we add *silicate glass* [(Si,Al,Ca,Mg,Fe)O; SiO<sub>2</sub> > 70 wt %; with 2516 68 occurrences] as an important yet often poorly characterized metamorphic phase in thermal 2517 metamorphic environments, particularly of arkosic sandstones and pelites (Reverdatto and 2518 Sóboley 1973: Grapes 2006). Two varietal names of metamorphic glass are "buchite." which 2519 forms when a silica-rich pelitic rock is altered by igneous contact, and "porcellanite," a glass 2520 derived from pyrometamorphosed clay, marl, shale, or bauxite (Grapes 2006). Melting and 2521 glass formation may occur as low as ~650 °C at 0.5 GPa in a granite protolith, or > 1000 °C at 2522 low pressure and dry conditions. Grapes (2006) details how "Si-rich glass" in many 2523 pyrometamorphic zones typically contains significant  $Al_2O_3$  and alkalis, a consequence of 2524 quartz-feldspar melting. Note that pure SiO<sub>2</sub> melts at 1700 °C—a temperature only attainable 2525 by lightning strikes or bolide impacts.

2526

2527 <u>Rarer Metamorphic Minerals:</u> In addition to the 94 mineral kinds outlined above, 661 other 2528 mineral kinds occur rarely as trace phases in metamorphic rocks, as documented by reports in 2529 numerous primary sources and compilations, notably Anthony et al. (1990-2003) and 2530 references cited in <u>https://mindat.org</u> and <u>https://rruff.info/ima</u> (both accessed 20 January 2531 2023). Most of these scarce minerals, which are listed in Supplementary Table 1, were not 2532 recorded from any of the 2785 metamorphic rock modes in Supplementary Table 3.

- 2533 However, a few of the less common minerals in Supplementary Table 1, were also noted in 2534 one or more metamorphic rock modes. Among these minor minerals, listed alphabetically, are 2535 alleghanvite (1 occurrence), analcime (2), anhydrite (2), ankerite (6), ardennite (1), arsenopyrite 2536 (1), axinite (4), bornite (1), braunite (7), bredigite (6), brownmillerite (5), bustamite (4), calzirtite 2537 (3), celestine (1), chondrodite (4), cuspidine (2), deerite (2), diaspore (1), fluorite (1), 2538 fluormayenite (5), friedelite (1), galaxite (1), giuseppeite (1), grunerite (8), hausmannite (1), 2539 hillbrandite (1), högbomite (2), ilvaite (2), jacobsite (1), kalsilite (5), kornerupine (1), kutnohorite 2540 (1), laumontite (1), leucite (2), margarite (1), monazite (3), nepheline (4), norbergite (1), 2541 piemontite (7), pigeonite (2), pyrophanite (1), pyroxmangite (2), qandilite (3), rhodocrosite (2), 2542 rhodonite (8), riebeckite (4), scawtite (1), siderite (9), sonolite (1), sphalerite (9), spessartine (9),
- stilpnomelane (3), suenoite (5), tephroite (1), thompsonite (3), uvarovite (2), wairakite (2), and
- 2544 "zeolite" (6).







