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1 **Iron-biomineralizing organelle in magnetotactic bacteria: function, synthesis**
2 **and preservation in ancient rock samples**

3

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11

12 **Abstract**

13 Magnetotactic bacteria (MTB) are ubiquitous aquatic microorganisms that incorporate iron
14 from their environment to synthesize intracellular nanoparticles of magnetite (Fe_3O_4) or
15 greigite (Fe_3S_4) in a genetically controlled manner. Magnetite and greigite magnetic phases
16 that allow MTB to swim towards redox transitions zones where they thrive. MTB may
17 represent some of the oldest microorganisms capable of synthesizing minerals on Earth, and
18 have been proposed to significantly impact the iron biogeochemical cycle by immobilizing
19 soluble iron into crystals that subsequently fossilize in sedimentary rocks. In the present article,
20 we provide an overview of the mechanisms leading to iron mineralization in MTB. We then
21 describe their distribution in the environment, and discuss the possible function of the
22 magnetite and greigite nanoparticles. Finally, we update the methods used for the detection
23 of MTB crystals in sedimentary rocks and present their occurrences in the geological record.

24 **Introduction**

25 Biomineralization corresponds to the capacity for living organisms to produce minerals such
26 as carbonates, sulfides, phosphates and oxides. It can be an indirect process during which the
27 metabolic activity of organisms transforms the physico-chemical conditions of their
28 environment and induces the formation of mineral phases. Alternatively, biomineralization
29 can be genetically controlled and serve biological functions. Biomineralization is widespread
30 among living organisms, and can significantly impact biogeochemical cycles of elements (*e.g.*
31 C, Ca, P, S, Fe) both qualitatively and quantitatively (*e.g.* Ridgwell and Zeebe, 2005).

32 Magnetotactic Bacteria (MTB) constitute an elegant example of controlled biomineralization
33 (Fig. 1). They were first observed by Salvatore Bellini, a medical Doctor at the University of
34 Pavia in Italia in the late 1950s and early 1960s (Bellini, 2009a,b). They were then described
35 by Richard Blakemore from samples collected near Woods Hole, Massachusetts (Blakemore,
36 1975). They both noticed a group of bacteria whose swimming direction could be tuned using
37 a magnet. Such magnetic behavior was explained by R. Blakemore, who observed intracellular
38 crystals corresponding to either magnetite [Fe(II)Fe(III)₂O₄] or greigite [Fe(II)Fe(III)₂S₄] in these
39 bacteria (Blakemore, 1975; Mann *et al.*, 1990).

40 Since their discovery, MTB have been found in a large variety of aquatic ecosystems including
41 lakes, rivers, oceans, estuaries and lagoons (Lin *et al.*, 2014a). They incorporate a high amount
42 of dissolved iron from their environment for magnetite or greigite synthesis, which
43 corresponds to a mass of 10- to 100-fold higher than the one estimated in the bacterium
44 *Escherichia coli* (Amor *et al.*, 2020a). When magnetotactic cells die, their crystals may be
45 deposited in sediments and preserved during diagenesis (*i.e.* the physico-chemical
46 transformation of sediments into sedimentary rock associated with natural burial), which
47 effectively removes iron from the soluble pool (Chen *et al.*, 2014; Larrasoña *et al.*, 2014). This

48 raised the hypothesis that MTB could significantly contribute to the iron biogeochemical cycle
49 by sequestering a large amount of dissolved iron into insoluble crystals (Lin *et al.*, 2014a; Amor
50 *et al.*, 2020a). MTB could have also impacted the ancient Earth surface, since they were
51 proposed to have emerged during the Archean (Kopp and Kirschvink, 2008; Lin *et al.*, 2017a).
52 In MTB, magnetite and greigite crystals are produced in organelles called magnetosomes.
53 They correspond to prokaryotic organelles made of a bi-layered lipid membrane containing
54 proteins and surrounding a single magnetite or greigite nanoparticle. Magnetosomes are
55 aligned as chains, and provide the cell with a magnetic dipole. It is assumed that chains of iron
56 nanoparticles are used as a compass, allowing the bacteria to passively align along the
57 geomagnetic field's lines to efficiently reach the Oxic/Anoxic Transition Zone (OATZ) in water
58 columns or sediments where they thrive (Frankel *et al.*, 1981). However, observation of MTB
59 at the geomagnetic equator where the Earth's magnetic field has no vertical component
60 (Frankel *et al.*, 1981) suggests that magnetite or greigite could have an additional function for
61 the bacteria such as energy storage (Byrne *et al.*, 2015).

62 Here, we first describe the biodiversity, distribution and ecophysiology of MTB, and discuss
63 the proposed function for magnetosomes in the environment. Then, we provide an overview
64 of iron biomineralization in MTB. Mechanisms of iron incorporation and magnetite formation
65 discovered in model magnetotactic strains cultivated in the laboratory are presented. Finally,
66 we summarize occurrences of fossil MTB found in the geological record, as well as the
67 methods used for identification of biological magnetite and greigite. Based on paleontological
68 data, we speculate on the emergence of MTB and potential additional function(s) for
69 magnetosomes over geological times.

70

71 **1. Ecophysiology of magnetotactic bacteria**

72 ***Biodiversity of magnetotactic bacteria***

73 MTB diversity is reflected by a great variation of their shape, physiology and taxonomy. All
74 known cultured and uncultured MTB are motile by means of flagella and have a cell wall
75 characteristic of Gram-negative bacteria. The arrangement of flagella differs among MTB and
76 can be either polar, bipolar or arranged in bundles. Another trait showing considerable
77 diversity is the arrangement of magnetosomes within the cell (Fig. 1). In the majority of MTB,
78 magnetosomes are aligned in one or more straight chains parallel to the cell's long axis, which
79 maximizes the magnetic moment of the bacteria and reflects their evolutionary optimization
80 for magnetic navigation. However, disordered magnetosomes located at one side of the cell
81 occur in some magnetotactic cocci (Moench, 1988; Cox *et al.*, 2002; Freitas *et al.*, 2003) (Fig.
82 1C). Magnetosome mineral particles display diverse, well-defined and strain-specific
83 morphologies (Bazylinski *et al.*, 1994). Four magnetite or greigite morphologies are found in
84 MTB: cuboctahedral, elongated prismatic, tooth-shaped and bullet-shaped (Balkwill *et al.*,
85 1980; Mann *et al.*, 1984a, 1984b; Mann *et al.*, 1987a, 1987b; Heywood *et al.*, 1990; Devouard
86 *et al.*, 1998; Pósfai *et al.*, 1998a, 1998b; Lefèvre *et al.*, 2011a) (Fig. 1). In addition to
87 magnetosomes, other inclusions containing elemental sulfur, polyphosphate or poly- β -
88 hydroxybutyrate (PHB) are common in MTB (Bazylinski *et al.*, 2004; Schultheiss *et al.*, 2005;
89 Rivas-Lamelo *et al.*, 2017). A study of MTB from the Seine River (France) indicated that cells
90 of uncultured MTB contain barium-rich and calcium oxide inclusions (Isambert *et al.*, 2007).
91 Some MTB from the Nitrospirae and Omnitrophica phyla also contain numerous sulfur
92 globules (Jogler *et al.*, 2010; Lefèvre *et al.*, 2011b; Kolinko *et al.*, 2012; Qian *et al.*, 2019).
93 Another feature illustrating MTB diversity is their type of magnetotactic behavior. A study
94 carried out on 12 cultured MTB from different taxa with various morphologies, physiologies,
95 and flagellar apparatus showed six different magnetotactic behaviors described as a

96 combination of three distinct mechanisms, including dipolar, axial and unipolar mechanisms
97 (Lefèvre *et al.*, 2014).

98 A straightforward experiment to reveal the diversity of MTB consists in performing a
99 magnetic enrichment of an environmental sample containing sediment and water. In most
100 habitats the light microscope observation will allow the observation of various cell
101 morphotypes and behaviors. The main morphotypes include coccoid-to-ovoid cells (Fig. 1A-C),
102 rods (Fig. 1D), vibrios and spirilla (Fig. 1E) of various dimensions. Two unique morphotypes
103 include the multicellular magnetotactic prokaryotes (MMPs) (Keim *et al.*, 2007) (Fig. 1F), and
104 the very large rod *Candidatus Magnetobacterium bavaricum* (Jogler *et al.*, 2010) (Fig. 1G).

105 MTB do not represent a well-defined taxonomic group of prokaryotes: they are distributed
106 over several bacterial taxa containing non-magnetotactic species as well (Lefèvre and
107 Bazylinski, 2013) (Fig. 2). Most of our knowledge on MTB diversity is based on isolation,
108 cultivation, genomic, metagenomic and single-cell genomic studies (Jogler *et al.*, 2011; Kolinko
109 *et al.*, 2012; Lefèvre *et al.*, 2012; Lin *et al.*, 2018). The majority of MTB described so far belongs
110 to four subdivisions of Proteobacteria (Alpha, Beta, Gamma and Deltaproteobacteria classes),
111 and some are affiliated with the Nitrospirae and Omnitrophica phyla (Lefèvre and Bazylinski,
112 2013; Lin *et al.*, 2017b) Recent metagenomic data have suggested that even some lineages
113 within the deep-branching Latescibacteria and Planctomycetes phyla may also produce
114 magnetosomes (Lin *et al.*, 2018). With their deep-branched phylogenetic position, the latter
115 groups are particularly important to study since they would represent the earliest examples
116 of biomineralization on Earth (Parks *et al.*, 2018). Magnetosome biogenesis is thus a
117 polyphyletic, possibly ancient trait associated with a large genetic, metabolic, and
118 morphological diversity. Interestingly, there is an obvious and apparently strong correlation
119 between the composition and morphology of magnetosome mineral crystals produced by

120 MTB and their phylogenetic affiliation (Lefèvre and Wu, 2013; Lefèvre *et al.*, 2013; Pósfai *et*
121 *al.*, 2013) (Fig. 2).

122 The physiology of known MTB, either determined experimentally with cultured strains or
123 inferred from genomic studies on uncultured strains, is also quite diverse. Cultured MTB
124 belong to three classes of the Proteobacteria phylum (Alpha, Gamma and
125 Deltaproteobacteria) (Fig. 2). They are obligate microaerophiles, anaerobes or both (Bazylinski
126 and Williams, 2007). Those that tolerate relatively high concentrations of oxygen do not
127 synthesize magnetite under these conditions. They are mesophilic with regard to growth
128 temperature, and none grow at temperatures much higher than 30°C. Known MTB are
129 chemoorganoheterotrophic and/or chemolithoautotrophic. In the model strains of the
130 *Magnetospirillum* genus, heterotrophic growth occurs while cells are using organic acids as a
131 source of carbon and electrons (Schleifer *et al.*, 1991). MSR-1 is also capable of autotrophic
132 and mixotrophic growth using reduced sulfur compounds as a source of electrons (Geelhoed
133 *et al.*, 2010). Species of the *Magnetococcus* (Lefèvre *et al.*, 2009; Bazylinski, Williams, Lefèvre,
134 Berg, *et al.*, 2013; Morillo *et al.*, 2014) and *Magnetospira* genera (Zhu *et al.*, 2010; Williams *et*
135 *al.*, 2012) as well as strains SS-5 and BW-2 of the Gammaproteobacteria class (Lefèvre *et al.*,
136 2012) are also growing autotrophically with reduced sulfur compounds. Species of the
137 *Magnetovibrio* (Bazylinski *et al.*, 2013) and *Terasakiella* (Monteil *et al.*, 2018) genera are
138 heterotrophic with *Magnetovibrio blakemorei* strain MV-1 that shows the greatest metabolic
139 versatility regarding the compounds that can be used as potential electron donors and carbon
140 sources for growth during microaerobic and anaerobic growth (Bazylinski *et al.*, 2013). Indeed,
141 it has a respiratory metabolism, using oxygen, nitrate, and nitrous oxide (N₂O) as terminal
142 electron acceptors (Bazylinski *et al.*, 1988). It grows chemoorganoheterotrophically and
143 chemolithoautotrophically but also chemoorganoautotrophically with formate as the electron

144 donor. The pathway of autotrophy is the Calvin-Benson-Bassham cycle since ribulose-1,5-
145 bisphosphate carboxylase/oxygenase (RubisCO) genes were found in the genome of most
146 autotrophic MTB of the Alpha and Gammaproteobacteria classes. Only *Magnetococcus*
147 species such as strain MC-1 appear to use the rTCA cycle for autotrophic carbon assimilation
148 (Bazylinski *et al.*, 2013). Cultured strains of the Deltaproteobacteria, including several species
149 of the Desulfovibrionales (Sakaguchi *et al.*, 2002; Lefèvre *et al.*, 2011c) and Desulfobacterales
150 (Abreu *et al.*, 2007; Descamps *et al.*, 2017) orders grow anaerobically while reducing sulfate.
151 Cells of some strains of the *Desulfovibrio* genus also present the potential to grow as a
152 microaerophilic band of cells at the oxic–anoxic interface in media lacking sulfate (Lefèvre *et*
153 *al.*, 2016). *Desulfovibrio magneticus* strain RS-1 is the only cultured magnetotactic bacterium
154 known to be capable of fermentation: pyruvate is fermented to acetate and hydrogen
155 (Sakaguchi *et al.*, 2002).

156 Genomic, environmental and ultrastructural evidences seem to indicate that the deep-
157 branched MTB including the Nitrospirae candidate genera *Magnetobacterium* (Jogler *et al.*,
158 2011; Lin *et al.*, 2014b; Kolinko *et al.*, 2016) and *Magnetoovum* (Lefèvre *et al.*, 2011; Kolinko
159 *et al.*, 2016) as well as some magnetotactic cocci of the Magnetococcaceae family isolated
160 from the water column of the Black Sea (Schulz-Vogt *et al.*, 2019) are growing anaerobically
161 using nitrate as respiratory electron acceptor and oxidizing reduced sulfur compounds.
162 Although some MTB have been shown to be sensitive to light (Frankel *et al.*, 1997; Chen *et al.*,
163 2011; Shapiro *et al.*, 2011) with genomic evidences showing the presence of phototrophic
164 genes in some *Magnetospirillum* strains (Wang *et al.*, 2019), no photosynthetic MTB have
165 been found. So far, no MTB were shown to be able to use reduced or oxidized forms of iron
166 for their energetic metabolism.

167

168 ***Environments hosting magnetotactic bacteria***

169 MTB have been described worldwide and found on all continents. They are ubiquitous in
170 sediments of freshwater, brackish, marine, and hypersaline habitats (Lin *et al.*, 2017b). The
171 occurrence of MTB appears to depend on the presence of an Oxic-Anoxic Transition Zone
172 (OATZ) consisting of opposing gradients of oxygen from the surface and reduced compounds
173 from the sediments (Frankel *et al.*, 1997). The largest numbers of MTB are typically found at
174 or slightly below the OATZ (Jogler *et al.*, 2010; Lefèvre *et al.*, 2011b). The different species of
175 MTB show specific distributions within the OATZ. Magnetite-producing MTB are generally
176 found at or very close to the OATZ, while greigite producers are present in reducing biotopes,
177 below the OATZ, in the sulfidic anoxic zone (Moskowitz *et al.*, 2008; Lefèvre *et al.*, 2011c).
178 Although most environmental studies on MTB were conducted on sediments of aquatic
179 environments, some freshwater and saline lakes with water column stratification, seasonally
180 or permanently, were also found to be suitable to host diverse populations of MTB (Simmons
181 *et al.*, 2004; Rivas-Lamelo *et al.*, 2017; Schulz-Vogt *et al.*, 2019).

182 Biogeochemical studies indicate that some environmental parameters such as salinity,
183 temperature, nitrate, or sulfur compounds could explain MTB abundance or community
184 differences (Martins *et al.*, 2009, 2012; Lin *et al.*, 2012, 2013). Even if the largest proportion
185 of MTB is detected within the suboxic zone, a strict correlation between the distribution of
186 MTB and specific geochemical parameters has never been demonstrated (Flies *et al.*, 2005).
187 For example, the presence of MTB is not correlated with the concentration in dissolved iron.
188 Ferrous or ferric salts in the micromolar concentrations range are sufficient for growth and
189 magnetite production (Schüler and Baeuerlein, 1996, 1998; Amor *et al.*, 2020a). Such
190 concentrations were shown to be typical of the free soluble iron found in environmental

191 sediments where MTB are most abundant (Flies *et al.*, 2005). Thus, iron does not represent
192 an element limiting growth and magnetosome formation of natural populations of MTB.
193 For years, MTB were thought to be restricted to habitats with pH values near neutral and
194 ambient temperatures. However, moderate thermophilic, psychrophilic, alkaliphilic and
195 acidophilic MTB were described including: an uncultured, moderately thermophilic bacterium
196 found in hot springs located in northern Nevada with a likely upper growth limit of about 63 °C
197 (Lefèvre *et al.*, 2010); a cocci isolated from the low-temperature (<1°C) Antarctic maritime
198 region (Abreu *et al.*, 2016); several obligatory alkaliphilic strains isolated in pure culture from
199 different aquatic habitats in California, including the hypersaline and hyperalkaline Mono Lake,
200 with an optimal growth pH of >9.0 (Lefèvre *et al.*, 2011c); and various morphotypes of MTB
201 isolated from an acidic freshwater lagoon in Brazil (pH ~ 4.4) (Abreu *et al.*, 2018). Deep-sea
202 sediments were also investigated for the presence of MTB. A study reported the presence of
203 alive MTB of different morphologies at water depths up to 3,000 m in the African continental
204 margin (Petermann and Bleil, 1993). Metagenomic analyses coupled to magnetic
205 measurements and electron microscope observations evidenced the occurrence of MTB in
206 deep-sea sediments from the eastern Pacific manganese nodule province at water depths of
207 4,970 - 5,620 m (Dong *et al.*, 2016). Given that MTB are markers of chemical and redox
208 gradients in aquatic settings, all chemically stratified environments could potentially support
209 populations of MTB.

210 Finally, magnetosome-producing cells were also found in association with other living
211 organisms. The presence of magnetotactic symbionts was observed within the marine bivalve
212 *Thyasira cf. gouldi* (Dufour *et al.*, 2014). This association is likely more related to predation
213 since the integrity of magnetosome chains and possibly flagellum is lost in the host. Predation
214 of MTB was also revealed in some protists such as bacterivorous ciliates that grazed MTB

215 (Bazylinski *et al.*, 2000; Monteil *et al.*, 2018). Interestingly, the accumulation of magnetite in
216 these microeukaryotes makes them sensitive to magnetic fields. In marine anoxic sediments,
217 another report evidenced a mutualistic symbiosis between excavate protists (Symbiontida,
218 Euglenozoa) and ectosymbiotic Deltaproteobacteria producing magnetosomes (Monteil *et al.*,
219 2019).

220

221 ***Biological function of magnetosomes***

222 The Earth's core generates a permanent magnetic dipole with field vectors having a non-zero
223 vertical component at every location on the planet but at the magnetic equator (Guyodo and
224 Valet, 1999). The magnetosome chain in MTB imparts a permanent magnetic moment to the
225 cell, and acts as a compass needle that aligns along the geomagnetic field lines (Frankel, 1984).
226 MTB can thus use the geomagnetic field to swim upward or downward along chemical
227 gradients in a process called magnetotaxis. It represents a selective advantage for
228 microorganisms living in spatially fluctuating niches such as those of chemically stratified
229 environments, and limits environmental prospection for energy sources and electron
230 acceptors to bidirectional transects instead of a volume (Frankel *et al.*, 1997). This trait
231 compensates unstable disturbances of vertical chemical gradients and help MTB to move
232 toward their optimal local niche. The geomagnetic field selects for a dominant cell polarity in
233 each hemisphere by favoring cells whose polarity caused them to swim downward along the
234 inclined field lines toward microaerobic/anaerobic sediments and away from high oxygen
235 concentrations in surface waters. This is supported by the observation of north-seeking MTB
236 that predominate in the Northern Hemisphere, and south-seeking cells predominating in the
237 Southern Hemisphere (Blakemore *et al.*, 1980). At the magnetic equator, south-seeking and
238 north-seeking MTB were found in equal concentrations (Frankel *et al.*, 1981). However,

239 magnetotaxis is not considered as a true taxis, but rather corresponds to a magnetically
240 assisted chemo-aerotaxis, directing the cells towards attractants or away from repellents
241 (Frankel *et al.*, 1997; Klumpp *et al.*, 2019). It is assumed that magnetic field passively
242 orientates the cells, which in turn actively swim using their flagellar apparatus (Frankel and
243 Bazylinski, 2006). In both hemispheres, magnetotaxis thus reduces the three-dimensional
244 random swim associated with chemo-aerotaxis to a one-dimensional search, supposedly
245 yielding an energy advantage over non-magnetotactic organisms (Klumpp *et al.*, 2019).

246 However, there are some exceptions that reported species of south-seeking MTB from the
247 Northern Hemisphere and north-seeking MTB from the Southern Hemisphere (Simmons *et al.*,
248 2006; Leão *et al.*, 2016). The magnetotactic protists whose magnetotaxis originates from the
249 interaction with ectosymbiotic magnetosome-producing bacteria were also described with an
250 atypical orientation of the cells polarity regarding the magnetic moment (Monteil *et al.*, 2019).

251 These specific behaviors, that could illustrate sensors having distinct response to chemical
252 gradients, seem to be restricted to anaerobes swimming upward to the bottom limit of the
253 OATZ. A variation in sensory response could also be at the origin of the different magneto-
254 aerotactic behaviors observed in cultured MTB that formed aerotactic band under oxygen-
255 gradient conditions (Lefèvre *et al.*, 2014). Indeed, it was suggested that the three magneto-
256 aerotactic mechanisms (*i.e.* polar, dipolar, and axial) are related to distinct oxygen sensing
257 mechanisms that regulate the direction of cells' motility in an oxygen gradient.

258 Thus, the most currently accepted hypothesis regarding the function of magnetosomes
259 corresponds to the increase of efficiency in finding preferred biotope, but alternative
260 biological functions were also proposed. Indeed, similar to other complex functions,
261 magnetosome biomineralization requires the coordination of a complex set of interacting
262 proteins with a large range of functions and domains (Uebe and Schüler, 2016; McCausland

263 and Komeili, 2020). This raises questions about the evolutionary steps and ancestral functions,
264 as well as on the environmental conditions and selective pressures that led to the emergence
265 of such a complexity. Several observations support that magnetosomes of modern lineages
266 could be used for other purposes than cell orientation. For example, some MTB produce more
267 magnetosomes than necessary for magnetotaxis (Spring *et al.*, 1993) (Figs. 1F and 1G), while
268 others thrive at the geomagnetic equator, where the Earth's magnetic field has no vertical
269 component (Frankel *et al.*, 1981). There, MTB cannot use the geomagnetic field to swim
270 upward or downward to find their optimal environment following redox gradients. When
271 grown in a metal chamber that cancels external magnetic field or without redox gradient,
272 some MTB keep producing magnetosomes (unpublished observations). Alternative
273 hypotheses for magnetosome function has thus been proposed.

274 It was originally speculated that magnetosomes could play a role in iron storage (Blakemore,
275 1982). Even though no evidence supports the role of magnetosomes as an iron-storage vesicle,
276 they could be used to prevent accumulation of free iron in the cytoplasm that would be
277 harmful to the cell (Imlay, 2003).

278 Alternatively, it has been proposed that magnetosomes could be used as energy-storing
279 systems (Kopp *et al.*, 2004; Byrne *et al.*, 2015). This could be achieved by oxidizing magnetite
280 into maghemite under oxidative conditions, thus releasing electrons that could be used as a
281 source of energy. Maghemite would then be reduced back to magnetite under reductive
282 conditions.

283 In addition, it was shown in MSR-1 and AMB-1 that proteins contained in the magnetosome
284 membrane exhibit a peroxidase-like activity (Guo *et al.*, 2012; Li *et al.*, 2015). Magnetosomes
285 were thus proposed to decrease and eliminate reactive oxygen species in the cell. However,
286 magnetite produces reactive oxygen species through Fenton reactions (He *et al.*, 2015).

287 Moreover, such hypothesis cannot explain the presence of crystal phases as only the proteins
288 located in the magnetosome membrane would be needed to eliminate reactive oxygen
289 species. Therefore, the peroxidase activity of magnetosomes may instead be used to prevent
290 accumulation of free radicals generated by magnetite crystals in the bacteria.

291

292 **2. Iron biomineralization in magnetotactic bacteria**

293 Magnetosomes are produced from the invagination of the inner cell membrane (Komeili *et al.*,
294 2006). In magnetite-forming MTB, a set of ~30 genes located in a distinct portion of the
295 genome, the Magnetosome Gene Cluster (MGC), is involved in the step-wise formation of
296 magnetosomes (Murat *et al.*, 2010). Magnetosome genes are used for (i) the formation of
297 magnetosome vesicles from invagination of the inner cell membrane, (ii) the alignment of
298 magnetosome vesicles as a chain, (iii) iron trafficking to magnetosomes for magnetite
299 precipitation, and (iv) crystal growth and maturation with controlled size and shape. Two
300 recent review articles discussed in details the genetic factors leading to magnetosome
301 formation (Uebe and Schüler, 2016; McCausland and Komeili, 2020). In the present
302 contribution, we focus on the chemical reactions leading to iron incorporation and
303 transformation in MTB during magnetosome formation. Iron biomineralization was mostly
304 studied in magnetite-forming strains, and was extensively characterized in the three
305 Alphaproteobacteria *Magnetospirillum magneticum* AMB-1, *Magnetospirillum*
306 *magnetotacticum* MS-1, and *Magnetospirillum gryphiswaldense* MSR-1, as well as in the
307 sulfate-reducing Deltaproteobacteria *Desulfovibrio magneticus* RS-1. A model compiling the
308 proposed pathways for iron biomineralization in MTB is illustrated in Figure 3.

309

310 ***Iron uptake***

311 MTB can take either dissolved Fe(II) or Fe(III) up from their environment. Incorporation of
312 reduced iron species occurs as a diffusion process (Schüler and Baeuerlein, 1996, 1998; Amor
313 *et al.*, 2018). Fe(III) assimilation requires energy-dependent mechanisms (Schüler and
314 Baeuerlein, 1996). In AMB-1 and MS-1, they involve biosynthesis of hydroxamate and catechol
315 siderophores, which are subsequently released to the external medium for Fe(III)
316 solubilization and incorporation (Paoletti and Blakemore, 1986; Calugay *et al.*, 2003, 2006).
317 Under iron-rich conditions, genes encoding for high-affinity Fe(II) transport are up-regulated,
318 while Fe(III) acquisition systems are down-regulated under the same conditions (Suzuki *et al.*,
319 2006). Contrastingly, siderophore-independent Fe(III) incorporation was observed for MSR-1
320 (Schüler and Baeuerlein, 1996). Finally, Fe(II) has been shown to be preferentially
321 incorporated by MTB when both oxidized and reduced species are available for the bacteria
322 (Amor *et al.*, 2018). This illustrates that iron assimilation favors energy-independent
323 mechanisms under sufficient iron conditions (Suzuki *et al.*, 2006).

324

325 ***Iron-storing mechanisms***

326 Once incorporated, iron is first stored into the cell as Fe(III) species (Amor *et al.*, 2018). In the
327 case of Fe(II) incorporation, iron is thus fully oxidized upon internalization (Amor *et al.*, 2018).
328 The subcellular location of these species remains debated. They could be contained in the
329 cytoplasm and/or periplasm. Several proteins are known to store iron, including ferritins,
330 bacterioferritins and DNA-binding proteins (Dps) (Arosio *et al.*, 2017; Uebe *et al.*, 2019).
331 Ferritin-like structures have been identified in AMB-1 (Baumgartner *et al.*, 2013), MSR-1
332 (Faivre *et al.*, 2007; Fdez-Gubieda *et al.*, 2013; Uebe *et al.*, 2019) and MS-1 (Frankel *et al.*, 1983).
333 From Mössbauer characterization of iron in MS-1 and MSR-1, it was proposed that oxidized
334 iron present as ferritin-like structures is then partially reduced into ferrous iron for trafficking

335 to magnetosomes and precipitation as magnetite (Frankel *et al.*, 1983; Faivre *et al.*, 2007).
336 These results were confirmed by measurement of iron isotopes in AMB-1 (Amor *et al.*, 2016,
337 2018). RS-1 can also accumulate phosphate- and sulfur-associated Fe(II) species upon iron
338 assimilation (Baumgartner *et al.*, 2016).

339

340 ***Iron delivery to magnetosomes***

341 In contrast to ferric iron, ferrous iron is soluble at circumneutral pH, which can help iron
342 trafficking in the cell (Millero *et al.*, 1995). Fe(II) could be addressed to magnetosomes *via* the
343 periplasm when the magnetosome membrane is still invaginating from the cell's inner
344 membrane (Faivre *et al.*, 2007; Jones *et al.*, 2015). Alternatively, ferrous iron could be
345 transported to mature magnetosome vesicles across the magnetosome membrane. The latter
346 hypothesis was recently supported by two high-resolution electron microscopy and
347 fluorescence studies, which evidenced a layer of unmineralized ferrous ions surrounding
348 magnetite in the magnetosome membrane in both cultured and environmental bacteria
349 (Werckmann *et al.*, 2017; Amor *et al.*, 2020b). In that regard, genes encoding Fe(II)
350 transporters (*feoAB* operon) have been found in the vicinity of all MCGs of MTB with their
351 genome sequenced (Suzuki *et al.*, 2006; Rong *et al.*, 2008, 2012; Uebe and Schüler, 2016).
352 Deletion of these genes causes a strong impairment of magnetite biomineralization.

353

354 ***Magnetite formation and possible precursors***

355 Fe(II) / Fe(III) ratio at the magnetosome location needs to be tightly controlled for magnetite
356 or greigite formation. This ratio is 0.5 in both mineral phases, although slightly oxidized
357 magnetite in MTB [*i.e.* Fe(II) / Fe(III) ratios lower than 0.5] has been consistently reported
358 (Weiss *et al.*, 2004; Kopp *et al.*, 2006; Moskowitz *et al.*, 2008; Li *et al.*, 2009). Several groups

359 proposed that magnetite precursors are first precipitated. In AMB-1, MS-1, and MSR-1, these
360 precursors include oxidized phases such as ferrihydrite [Fe(III)₂O₃·5H₂O] (Frankel *et al.*, 1983;
361 Faivre *et al.*, 2007; Baumgartner *et al.*, 2013; Fdez-Gubieda *et al.*, 2013). One study also
362 suggested that hematite [α -Fe(III)₂O₃] could represent a magnetite precursor (Le Nagard *et*
363 *al.*, 2019). Fe(II) transported to magnetosomes would thus have to be oxidized for
364 precipitation of these precursors (Amor *et al.*, 2018). The candidate proteins for oxidation
365 reactions include MamP, which contains a c-type cytochrome domain (Siponen *et al.*, 2012,
366 2013; Jones *et al.*, 2015). It has been detected in the magnetosome membrane (Raschdorf *et*
367 *al.*, 2018). Fe(II) oxidation could thus happen during iron diffusion across the magnetosome
368 membrane or in the magnetosome lumen. Magnetite would then form upon further Fe(II)
369 addition in magnetosomes (Tronc *et al.*, 1992; Ahn *et al.*, 2012). This proposed
370 biomineralization pathway was recently challenged by Uebe and co-workers, who showed
371 that ferritins and ferritin-like structures (*i.e.* ferrihydrite) are not necessary for magnetite
372 precipitation in MSR-1 (Uebe *et al.*, 2019). They produced a mutant strain lacking iron-storing
373 proteins, and characterized iron species in these bacteria as well as in wild-type cells. They
374 found that the presence of ferrihydrite in MSR-1 is tied to the presence of iron-storing proteins.
375 However, magnetite biomineralization was not altered in the mutant strain. They rather
376 proposed a distinct function for these proteins, which protected the bacteria from oxidative
377 stress. From these results, the authors concluded that iron-storing proteins are not involved
378 in biomineralization and that ferrihydrite does not represent a magnetite precursor. However,
379 the possibility of alternative biomineralization pathways triggered by the absence of iron-
380 storing proteins cannot be completely ruled out. Moreover, the mutant strains lacking iron-
381 storing proteins were cultivated under a limited number of growth conditions. It is thus
382 difficult to give a definitive conclusion about the role of ferritin-like structures. Finally, green

383 rust [a mixed Fe(II)/Fe(III) hydroxide] was proposed as a potential magnetite precursor in RS-
384 1, a strain genetically distant from species of the *Magnetospirillum* genus (Baumgartner *et al.*,
385 2016). Although the genetic content appears to be quite conserved among MTB, it is possible
386 that they developed different strategies for magnetite biomineralization depending on their
387 physiology and environment.

388

389 ***Iron distribution in magnetotactic bacteria***

390 Physical and chemical characterizations of model magnetotactic strains demonstrated that
391 different iron species can be present in MTB. Time-course experiments conducted with AMB-
392 1 and MSR-1 observed that virtually all iron was contained in magnetite when
393 biomineralization was complete (Baumgartner *et al.*, 2013; Fdez-Gubieda *et al.*, 2013). In
394 contradiction with these findings, direct measurements of iron in wild-type AMB-1 and MSR-
395 1, as well as an AMB-1 mutant strain unable to form magnetosomes, evidenced a large pool
396 of iron distinct from magnetite (Amor *et al.*, 2016, 2018, 2020a; Berny *et al.*, 2020). Such
397 additional pool of iron can represent half of the total intracellular iron in wild-type AMB-1 and
398 MSR-1 (Amor *et al.*, 2020b; Berny *et al.*, 2020). The discrepancy between these findings and
399 the time-course experiments was proposed to raise from experimental conditions. Indeed,
400 time-course experiments require to first grow bacteria under iron-starving conditions (*i.e.* no
401 iron provided to the growth medium), which trigger overexpression of iron transporters
402 (Suzuki *et al.*, 2006; Wang *et al.*, 2017). This could optimize the transfer of iron to
403 magnetosomes for magnetite precipitation, and prevent accumulation of iron in a reservoir
404 distinct from magnetite. Interestingly, mutant AMB-1 strains unable to form magnetosomes
405 also accumulated iron (Amor *et al.*, 2020a). However, the total mass of iron incorporated by
406 these mutant bacteria was significantly lower than the one contained in wild-type bacteria.

407

408 ***Fe(II) dynamics in magnetotactic bacteria***

409 Finally, iron isotope modeling during magnetite biomineralization in AMB-1 allowed the
410 identification of Fe(II) diffusion from the cell's internal medium to the external growth
411 medium (Amor *et al.*, 2016, 2018). A release of intracellular Fe(II) to the external medium
412 could have resulted from cell lysis. However, monitoring of cell concentration during bacterial
413 cultures ruled out such possibility. Bacterial Fe(II) outward diffusion thus rather reflects
414 passive Fe(II) exchange with the external medium.

415

416

417 **3. Tracking the footprint of MTB in the fossil record**

418 ***Criteria for detection of MTB fossils***

419 The specific mineralization pathway provides MTB magnetite with unique features that are
420 used for the detection of MTB fossils (hereafter referred to as magnetofossils) in ancient rock
421 samples. For several decades, magnetofossil identification relied on morphological,
422 crystallographic and magnetic criteria, which are now completed by chemical and isotopic
423 fingerprints, as well as additional magnetic methodologies. Thomas-Keprta and co-workers
424 (2000) defined six robust criteria for magnetofossil identification (see also review and
425 discussion in Jimenez-Lopez *et al.*, 2010; Li *et al.*, 2013). Here we summarize these criteria and
426 update them using the most recent discoveries (Fig. 4)

427 Unlike abiotic magnetite, MTB crystals show blunt edges associated with typical shapes (see
428 below) (Kopp and Kirschvink, 2008; Witt *et al.*, 2005). Their size ranges from less than 35 nm
429 up to 120 nm (Konhauser *et al.*, 1998). Because crystal growth in MTB is tightly controlled by
430 proteins, the particle sizes and shape factors (*i.e.* width/length ratios) in a given population

431 show a narrow skewed gaussian distribution (Devouard *et al.*, 1998) compared to their abiotic
432 counterparts as well as magnetite produced through induced biomineralization (Loveley *et al.*,
433 1987; Sparks *et al.*, 1990; Arató *et al.*, 2005). However, this criterion is not sufficient to
434 distinguish magnetofossil from abiotic magnetite in natural sedimentary rocks because
435 abundance peaks of biotic and abiotic magnetite can overlap and create more complex
436 distributions (Arató *et al.*, 2005).

437 The size and shape of MTB nanoparticles fall into the stable single magnetic domain, which
438 maximizes the magnetic moment for each particle (Witt *et al.*, 2005; Muxworthy and Williams,
439 2008). The alignment of Stable Single-Domain (SSD) magnetite particles in the magnetosome
440 chain imparts a strong magnetic moment to the bacteria. The unique chain arrangement of
441 SSD nanoparticles provides MTB with distinct magnetic features characterized by a large
442 remanent magnetization as well as a limited interaction field between particles (Li *et al.*, 2012).
443 First Order Reversal Curves (FORC), a magnetic method based on the measurement of
444 hysteresis curves, as well as Ferromagnetic Resonance (FMR) spectroscopy have successfully
445 identified intact chains of fossilized magnetosomes (Roberts *et al.*, 2013; Heslop *et al.*, 2014;
446 Roberts *et al.*, 2019). However, such magnetite alignment may be lost during sediment burial
447 and fossilization processes owing to the degradation of proteins assembling magnetosome
448 chains (Li *et al.*, 2012). In that case, the typical FORC signatures of magnetosome chains are
449 lost. The Verwey temperature (*i.e.* the temperature below which magnetite undergoes a
450 phase transition associated with a modification of surface charge configuration and a sharp
451 change of susceptibility and remanent magnetization) has also been used to test the
452 magnetotactic origin of fossil magnetite samples (Chang *et al.*, 2016). Finally, Isothermal
453 Remanent Magnetization (IRM) and Anhysteretic Isothermal Remanent (ARM) analyses
454 allowed to cluster the origins of magnetite and to estimate the proportion of biogenic

455 magnetite in a bulk sample using a non-negative matrix factorization (Egli, 2004; Heslop and
456 Dillon, 2007; Roberts *et al.*, 2013). This method has been used efficiently on magnetite
457 samples from Eocene sedimentary rocks that survived diagenetic modifications (Day *et al.*,
458 1977; Roberts *et al.*, 2013; Roberts *et al.*, 2019).

459 Transmission Electron Microscopy (TEM) analyses also showed that magnetite produced by
460 MTB usually grows along the [111] crystalline axis without lattice defects (Mann *et al.*, 1987a;
461 Vali *et al.*, 1987; Devouard *et al.*, 1998). However, evidences of elongation anomalies along
462 the [110] or [100] axes were reported (Isambert *et al.*, 2007; Benzerara and Menguy, 2009; Li
463 *et al.*, 2015). So far, the crystal growth along the [111] axis still seems to be congruent for
464 prismatic magnetite.

465 Laboratory experiments demonstrated a low incorporation of trace elements contained in the
466 bacterial growth medium into AMB-1 magnetite (Amor *et al.*, 2015). For most tested elements,
467 such incorporation was at least 100 times lower compared to abiotic magnetite samples
468 synthesized from co-precipitation of Fe(II) and Fe(III) ions in aqueous solutions (Amor *et al.*,
469 2015). Chemical purity can thus be used as a chemical fingerprint of MTB magnetite. However,
470 a few elements such as molybdenum and tin showed preferential incorporation into AMB-1
471 magnetite. They could reflect metabolic reactions in which these two elements are involved,
472 and thus represent biological signatures of MTB activity recorded in magnetite. We note that
473 several publications reported improved incorporation of elements from the iron series such
474 as cobalt and manganese into MTB magnetite for biotechnology purposes (*e.g.* Staniland *et al.*,
475 2008; Keim *et al.*, 2009). Trace elements incorporation was not compared with abiotic
476 conditions under such high concentrations. Therefore, they have limited paleontological
477 applications.

478 Finally, isotopic tools were used to search for magnetofossils in ancient rock samples. Oxygen
479 isotope composition of magnetite produced by the strains MS-1 and MV-1 has been
480 investigated, and showed a temperature dependency (Mandernack *et al.*, 1999). However, no
481 difference was found between MTB magnetite and magnetite formed extracellularly by
482 Dissimilatory-Iron Reducing Bacteria (DIRB), a group of iron-metabolizing microorganisms that
483 perform magnetite induced biomineralization. In the same study, no specific iron isotope
484 signatures were evidenced in the magnetite produced by the magnetotactic strains MS-1 and
485 MV-1 (Mandernack *et al.*, 1999). Contrastingly, magnetite produced by AMB-1 demonstrated
486 a significant depletion in heavy iron isotopes relative to the growth medium and the organic
487 fraction of the cell (Amor *et al.*, 2016). Additionally, the odd iron isotope (^{57}Fe) showed a
488 specific incorporation into AMB-1 magnetite. Such isotopic anomaly with preferential
489 incorporation of ^{57}Fe compared to the three other isotopes (^{54}Fe , ^{56}Fe and ^{58}Fe) has never
490 been observed before and may thus constitute a unique biosignature of magnetite produced
491 by MTB. However, the origin of this isotopic signature remains unclear and needs to be
492 investigated further on other MTB strains and natural samples.

493 So far, it can be highlighted that neither a single nor couples of criteria are sufficient to attest
494 for magnetite biogenicity in the past sedimentary record. A whole body of evidences based
495 on mineralogical, magnetic, chemical and isotopic tools is required for a firm identification of
496 magnetofossils (Kopp and Kirschvink, 2008). Such difficulty raises from the limited number of
497 magnetotactic strains tested for their morphologic, magnetic, chemical, and isotopic
498 specificities. Finally, additional work should be carried out to determine whether these criteria
499 can survive sediment burial and fossilization processes (see below).

500

501 ***Magnetofossil occurrences in the rock record***

502 Magnetofossils were identified in sedimentary rocks from various ages, most of which were
503 observed in quaternary sediments from lakes, carbonate platforms, pelagic or hemi-pelagic
504 environments (*e.g.* Mc Neill *et al.*, 1988; Yamazaki and Ioka, 1998). The oldest observation
505 during the current geologic eon Phanerozoic (0-540 Ma) was reported in Late Cretaceous
506 chalks from Southern England that were deposited about 85 Ma ago (Montgomery *et al.*,
507 1998). This study reported detailed paleomagnetic data based on SSD magnetite-bearing
508 samples. Their observation using TEM demonstrated the presence of well-preserved chains of
509 magnetosomes, constituted mainly of prismatic magnetite although cuboctahedral and
510 bullet-shaped particles were also present.

511 In contrast, scarce traces of magnetofossils were reported in rock records of the Precambrian
512 representing the earliest and longest period of Earth's history, from 3.8 Ga to 540 Ma (Fig. 5).

513 In the 1980s, Chang and co-workers examined a number of Precambrian stromatolitic
514 limestones and cherts using rock magnetic methods and high-resolution electron microscopy
515 on magnetic extracts (Chang *et al.*, 1989). They described several potential magnetofossils,
516 among which the most robust and ancient traces including magnetosome chains were
517 recorded in the 2.0 Ga-old Gunflint Formation, Western Australia. Several years later, Akai and
518 coworkers (1997) provided evidence for the oldest putative magnetofossils ever reported in
519 terrestrial samples in stromatolites from the Tumbiana Formation, Western Australia, formed
520 in a marine basin 2.7 Ga ago. Although very promising, these results rely only on the size (in
521 the SSD) and shape of magnetite crystals determined by TEM analyses (Akai *et al.*, 1997); they
522 thus require to be validated by further investigations.

523 The presence of magnetofossils in ancient extraterrestrial material was also suggested, as
524 illustrated by the controversial studies of the Martian meteorite ALH84001 (McKay *et al.*,
525 1996; Thomas-Keprta *et al.*, 2000, 2009; Brearley, 2003; Treiman, 2003; Jimenez-Lopez *et al.*,

2012). This sample might represent the oldest evidence for magnetofossils in the solar system. The meteorite ALH84001 is an orthopyroxenite that crystallized 4.1 Ga ago (Lapen *et al.*, 2010). It contains carbonate globules formed in a late stage about 3.9 Ga ago, likely from low-temperature hydrothermal fluids. Nano-sized magnetite crystals with cuboctahedral, teardrop, prismatic and irregular shapes were identified in the carbonate globules (McKay *et al.*, 1996; Thomas-Keprta *et al.*, 2009). Large statistical TEM analyses indicate that 30% of the nanomagnetites were elongated prisms showing a striking similarity with those formed on Earth by the magnetotactic strain MV-1 (Thomas-Keprta *et al.*, 2000). In addition, extraterrestrial SSD nanomagnetites exhibited restricted anisotropy, crystallographic perfection (*i.e.* free of internal defects), elongation along the [111] axis and chemical purity, but no chain arrangement (Thomas-Keprta *et al.*, 2000, 2009). It should be noted that chemical purity was assessed from energy dispersive X-ray spectroscopy, a method that is not suitable to strictly evaluate the biogenicity of magnetite based on its trace element content (Amor *et al.*, 2015). The nanomagnetite biogenicity in the ALH84001 meteorite is still a matter of debate because alternative hypotheses have been proposed to explain their formation. These crystals could also have been formed from shock-induced thermal decomposition and metamorphism of Fe carbonates (Brearley, 2003; Treiman, 2003), even though chemically-pure magnetite can hardly be produced from impure carbonates (Jimenez-Lopez *et al.*, 2012). These last ten years, new magnetofossil criteria were identified such as trace element and iron isotope compositions, which could help to further decipher the origin of nanomagnetites in the ALH84001 meteorite.

547

548 ***Alteration vs preservation of magnetofossils in sediments***

549 As stated in the introduction, magnetite can be degraded during diagenesis. Three main
550 diagenetic processes can alter magnetite and prevent the identification of primary
551 magnetosomes: (i) oxidation, (ii) chain breakup, and/or (iii) reductive dissolution (see review
552 of Roberts, 2015). The first process happens when magnetite experiences oxidizing conditions,
553 due to diffusion of molecular oxygen in sediment or late exposition of sedimentary rock to the
554 atmosphere. This leads to partial or total transformation of magnetite to maghemite [γ -
555 $\text{Fe(III)}_2\text{O}_3$] (Chang *et al.*, 2013). In the second and third processes, one must consider that all
556 sediments contain a fraction of organic matter used as electron donor for aerobic or anaerobic
557 respiration processes. Various oxidants are utilized as electron acceptors following a well-
558 established sequence depending on the free energy of each respiration pathway: O_2 , NO_3^- ,
559 manganese oxides, iron oxides, and SO_4^{2-} (Froelich *et al.*, 1979). The magnetosome
560 membranes of MTB are mostly made of lipids, which can be degraded by microbial oxidative
561 process. The chain of magnetosomes is held by a filament mainly constituted by the actin-like
562 MamK protein (Komeili, 2012; Uebe and Schuler, 2016), and can thus also be degraded by
563 diagenetic microbial processes. The last stage of anaerobic respiration during sediment burial
564 is based on microbial SO_4^{2-} reduction that releases H_2S in porewater. Many studies of marine
565 sediments demonstrated that H_2S reduces and dissolves magnetite (*e.g.* Canfield and Berner,
566 1987; Mohamed *et al.*, 2011), with magnetite half-life ranging from 5 to 1000 years (Canfield
567 and Berner, 1987; Emiroglu *et al.*, 2004). If SO_4^{2-} and organic matter are abundant in sediments,
568 one might expect magnetite to be mostly consumed and not preserved in rocks (Roberts,
569 2015). What might thus be a factor of magnetofossil preservation in sediments? SO_4^{2-} -poor
570 environments such as modern lake and river systems provide potentially favorable conditions
571 for preservation. Indeed, only a small amount of H_2S can be produced in SO_4^{2-} -poor sediments
572 and reductive dissolution of magnetite may be limited. Significant amount of magnetofossils

573 could also be present in Archean sedimentary rocks (3.8 to 2.5 Ga old) since SO_4^{2-}
574 concentrations dissolved in the ocean at that time were extremely low ($< 2.5 \mu\text{M}$; Crowe *et*
575 *al.*, 2014), and organic matter supply to the sediments was likely weak due to low biological
576 productivity (Canfield, 2005). It can be noted that the biggest natural reservoir of magnetite
577 in the Earth's crust is from the Archean banded iron formation, termed BIF (Konhauser *et al.*,
578 2017). This indirectly confirms the high capacity of magnetite preservation in Archean
579 sediments even though BIFs have never been supposed to be formed from MTB remaining.
580 Magnetofossil preservation in sediments can also be inferred from data available for
581 Phanerozoic sedimentary rocks, in which a striking relationship was observed between
582 magnetofossil features and glacial-interglacial cycles (Paasche *et al.*, 2004, 2011). Analysis of
583 sediments covering the last million years showed that glacial stages are associated with an
584 enrichment of organic matter but a decrease in magnetofossil abundance (Lean and McCave,
585 1998; Hesse, 1994; Yamazaki and Kawahata, 1998). These variations are interpreted as an
586 increase of biological productivity leading to higher organic matter flux and stronger reductive
587 dissolution of magnetite during sediment diagenesis. In contrast, interglacial periods
588 correspond to lower biological productivity, thus lowering organic carbon flux. The O_2
589 diffusion from bottom water to sediment extended deeper in the sediment and increased the
590 size of the OATZ, where MTB thrive, up to tens of meters. This resulted in (i) a decrease of
591 organic matter content (inducing a lower H_2S production and reductive dissolution) and (ii) an
592 increase of MTB in the larger OATZ (Roberts, 2015). Both effects globally increase the capacity
593 of magnetofossil preservation in sediments. Another scenario has also been suggested to
594 explain abundant magnetofossils in Phanerozoic sediments. Several authors proposed that
595 the amount of iron-rich dust could control the production and preservation of MTB (Roberts
596 *et al.*, 2011; Yamazaki and Ikehara, 2012; Savian *et al.*, 2014). Indeed, iron-dust particles

597 increase the primary productivity in the water column but can also be delivered to the
598 sediments and be used for anaerobic microbial oxidation of organic matter. This latter process
599 would increase the availability of dissolved Fe(II) in sediment porewater (used by MTB for
600 magnetite production), and decrease organic matter content, which in turn would decrease
601 the relative production of H₂S (and dissolution of magnetite) by sulfate-reducing bacteria.
602 Finally, magnetofossil spikes were also evidenced during a global warming event ~55 Ma ago
603 known as the Paleocene-Eocene Thermal Maximum (PETM) (Kopp *et al.*, 2007, 2009).
604 Unusually large magnetite crystals sharing morphological and crystallographic features typical
605 of MTB were observed during the PETM (Schumann *et al.*, 2008). As suggested by the
606 glacial/interglacial cycles, warmer climate conditions could thus favor MTB magnetite
607 formation and/or preservation.

608

609 **Tracking the age of the last common MTB ancestor**

610 Resolving the evolution of controlled iron biomineralization in MTB is a major challenge in
611 evolutionary biology because this trait is polyphyletically distributed in taxa that are separated
612 by billion years of evolution from their most recent common ancestor. This intriguing
613 phylogenetic distribution of MTB raised three main questions tackled by the scientific
614 community over the last three decades: *(i)* has magnetosome formation a single genetic
615 origin?, *(ii)* was it an ancestral character shared by the ancestors of all phyla, genera and
616 species including MTB lineages?, and *(iii)* when this function emerged during the evolution of
617 life on Earth? To figure this out, several research groups performed whole genome sequencing
618 and inferred molecular phylogenies of MTB species and magnetosome genes (Abreu *et al.*,
619 2011; Lefèvre *et al.*, 2013; Lin *et al.*, 2017a, 2018; Monteil *et al.*, 2018). Over the years,
620 datasets got progressively stronger and stronger by including a higher taxonomic sampling

621 and a higher degree of phylogenetic signal thanks to whole genome datasets. Together with
622 molecular genetics, they provided evidence of a single genetic origin followed by a duplication
623 event that led to the emergence of the modern greigite and magnetite Magnetosome Gene
624 Clusters (MGCs) at some point in the MTB diversification. Two scenarios are currently
625 proposed: (i) a duplication of an unknown form of the MGCs that occurred well before the
626 emergence of the deep-branched PVC group (*i.e.* the superphylum gathering the
627 Planctomycetes, Verrucomicrobia and Chlamydiae phyla) that contain MTB, and (ii) a much
628 more recent duplication of the magnetite MGC in the Deltaproteobacteria that was
629 neofunctionalized to biomineralize greigite (Lin *et al.*, 2018). Whenever this duplication
630 occurred, the distribution of magnetotaxis in the different bacterial phyla was mainly
631 explained by the vertical inheritance of the MGCs followed by numerous independent gene
632 losses in the non-magnetotactic lineages over phyla diversification (Lefèvre *et al.*, 2013; Lin *et*
633 *al.*, 2017a). Accordingly, controlled iron biomineralization emerged just before the emergence
634 of Nitrospirae and Proteobacteria phyla, which was estimated to have occurred during the
635 Archean over a timescale between 3.21 to 3.38 Ga ago with 95% confidence intervals ranging
636 over approximately 500 million years (Lin *et al.*, 2017a). As a consequence, the ancestor of a
637 tremendous amount of modern bacterial lineages, but not all, could have performed
638 controlled iron biomineralization.

639 However, the above-mentioned phylogenies and the use of molecular clocks for prokaryotic
640 organisms were discussed by other research groups. Tackling MTB evolution is equivalent to
641 inferring the timing of the explosive radiation that led to the emergence of all major
642 Eubacteria phyla billions of years ago (Battistuzzi *et al.*, 2004; Betts *et al.*, 2018). At such
643 evolution timescale, methods in molecular phylogeny faces numerous limitations that
644 introduces uncertainties in reconstruction of tree topologies and estimation of event timing

645 [e.g. long branch attraction artifacts (Bergsten, 2005), impracticable and miscalibrated
646 molecular clocks (Graur *et al.*, 2004), effect of gene flow blurred over timescales and site
647 saturation (Duchêne *et al.*, 2016)]. According to some authors (Wang and Chen, 2017), the
648 current proposed scenario may not be parsimonious as long as it involves too many loss events
649 and rely on an uncertain tree topology. Additional investigations will help to refine these
650 inferences, and in the case of controlled iron biomineralization, magnetofossils could
651 represent a major asset to calibrate evolutionary history of life.

652

653 ***Predicting MTB evolution in relation to the Precambrian ocean chemistry***

654 Geochemical traces of biogenic reduced carbon in turbiditic and pelagic sedimentary rocks
655 from the Isua supracrustal belt in west Greenland suggested that early life emerged long
656 before 3.7 Ga (Rosing, 1999). Molecular clock analyses later confirmed that the last universal
657 common ancestor of cellular life emerged ~4 Ga ago followed by an evolutionary burst of all
658 Eubacteria phyla that likely occurred all over the Archean eon (Battistuzzi *et al.*, 2004; Betts
659 *et al.*, 2018). Do current knowledge of MTB evolution and magnetofossils allows to estimate
660 the time of emergence of controlled iron biomineralization among prokaryotes and its role in
661 Earth's evolution?

662 Although the presence of magnetofossils in the geological record before the end of the
663 Paleoproterozoic needs to be further investigated, controlled iron biomineralization could
664 have evolved before -2.7 Ga. From the angle of molecular phylogeny, this event could have
665 occurred a lot earlier. Indeed, the age of the last common MTB ancestor was estimated
666 between the Paleo- and Mesoarchean era, which corresponds to the timescale of the radiative
667 evolution at the origin of all the major Eubacteria phyla. On such evolutionary timescale,
668 deepest branching relationships of the tree of life are extremely difficult to reconstruct and

669 interpret because of the paucity of fossils and the tremendous uncertainty of phylogenetic
670 reconstructions. However, given the currently known MTB phylogenetic distribution, it seems
671 very unlikely that controlled iron biomineralization emerged before the radiation of the
672 Terrabacteria, Actinobacteria, Cyanobacteria, Firmicutes, and all candidate phyla radiations
673 revealed in the dark microbial matter (Hug *et al.*, 2016). Although no geological data have ever
674 supported these inferences yet, paleontology may represent a major ally by reconstructing
675 the global chemical environment over time scales and help to determine if both biology and
676 environmental conditions were suitable for magnetosome biogenesis.

677 In the Archean eon (-4 to -2.5 Ga), early life undoubtedly thrived in the oceans that were
678 dominantly anoxic and ferruginous [rich in dissolved Fe(II)], with limited zones of surface
679 waters being oxygenated by photosynthetic activity (Lyons *et al.*, 2014). During this period,
680 other strategies than photosynthesis emerged to exploit energy sources in anoxic conditions
681 including methanogenesis and methanotrophy (Rosing, 1999). Anoxic and ferruginous waters
682 of the Archean ocean, with local redox gradients such as those in which modern MTB are
683 observed, could have represented ideal conditions for the development and evolution of MTB.
684 Unlike modern oceans, redox boundary of the Archean ocean was likely located in the water
685 column (not in the sediment), and possibly extended in the photic zone (exposed to sunlight)
686 since O₂ was produced in shallow water by photosynthesis. As mentioned above, MTB can
687 respire oxygen but can also get energy anaerobically by respiring nitrate, nitrous oxide or
688 sulfate. However, it is still unclear whether they can oxidize hydrogen, sulfur or iron, or even
689 reduce Fe(III), metabolism that would be more suitable in Archean environments. This gap is
690 mainly due to the lack of representative deep-branching MTB for which full genomes and
691 cultures are available, especially those from ferruginous environments (Rivas-Lamelo *et al.*,
692 2017).

693 After 2.4 Ga, a progressive surface ocean oxygenation occurred in the early Paleoproterozoic
694 and diffused into the atmosphere, which is a stage known as the great oxidation event (GOE)
695 (Holland, 2002). As a consequence of atmospheric oxygenation, continental sulfides were
696 oxidized into highly soluble sulfates that were massively transferred to the ocean by river
697 systems (Canfield, 1998). From a paleontological perspective, it is widely accepted that
698 environmental conditions strongly influenced the evolution of life on Earth, and life in turn
699 impacted environmental conditions (Anbar and Knoll, 2002). It is likely that dissimilatory
700 sulfate-reducing prokaryotes have progressively reduced sulfates into sulfides in deep anoxic
701 ocean waters that precipitated as Fe sulfides. Such geochemical processes changed ocean
702 chemistry by decreasing the dissolved Fe concentrations. While dominated by iron in the early
703 Precambrian, deep ocean became S-dominated for a period that persisted at least from -1.8
704 Ga to -700 Ma (Canfield, 1998). Complete oxygenation of the oceans similarly to current state
705 occurred only between -700 and -600 Ma. The evolution of ocean chemistry towards more
706 sulfidic conditions after -2.4 to -1.8 Ga (*i.e.* Paleoproterozoic) (Canfield, 1998) could thus have
707 conducted to the development of other magnetotactic Deltaproteobacteria forming greigite
708 rather than magnetite. Although more speculative, this assertion could be tested by looking
709 in detail the magnetofossil record and crossing these data with phylogenetic reconstructions.
710 Overall, available data indicate that redox gradients generated by oxygen diffusion and
711 sustaining modern MTB could have existed since the Archean. This is critical to discuss the
712 evolution of MTB, since the magnetosome chains is thought to be used for navigation toward
713 optimal redox conditions in the water column or sediments. In the case of distinct function(s),
714 one cannot rule out the possibility that MTB magnetosomes evolved for purposes other than
715 magnetotaxis. Thus, first magnetosomes and MTB could have appeared even if no redox
716 gradients were established. Future studies will have to elucidate these points.

717

718 **Concluding remarks**

719 More than 40 years after the discovery of the first iron-biomineralizing magnetotactic
720 bacterium, technological advances in cell-sorting, genome sequencing, microscopy and
721 chemical analyses keep revealing the magnetosome complexity at the genetic, biological and
722 chemical levels. Magnetosome properties and its preservation in the geological record make
723 this organelle as one of the most promising tool to calibrate early Earth's chemistry and life
724 evolution. We believe that interdisciplinary collaborations between microbiologists,
725 paleobiologists, bioinformaticians and geochemists will foster innovative researches that will
726 answer to major questions in their fields and determine the origin of biomineralization and its
727 contribution to Earth.

728

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734

735 **Conflicts of interest**

736 The authors declare no conflict of interest.

737

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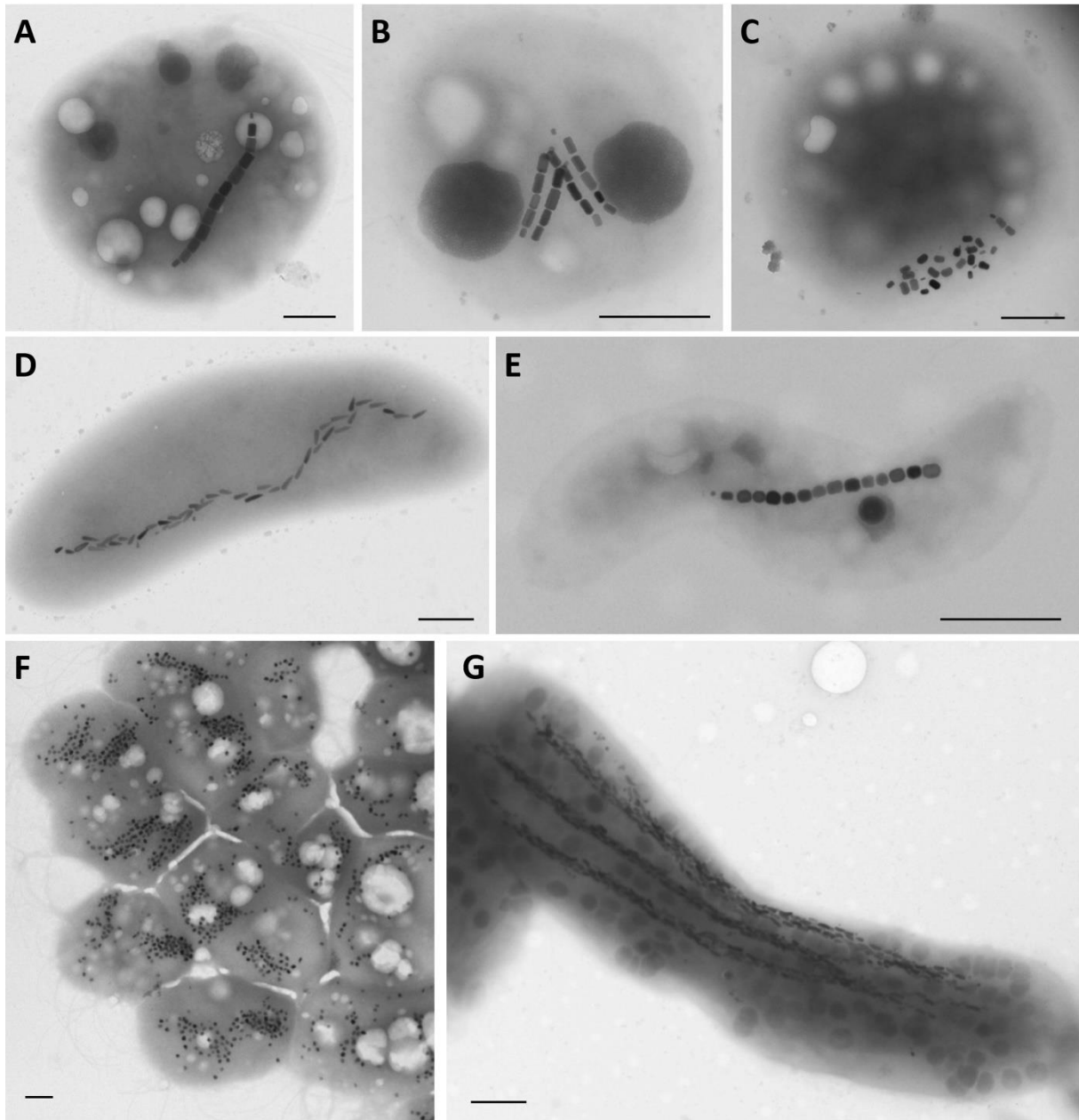
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- 1296

1297 **Figure legends**



1298

1299 **Fig. 1** Transmission electron microscope images of magnetotactic cocci (A-C), curved rod (D)
1300 spirillum (E), disaggregated multicellular magnetotactic prokaryote (MMP) (F) and *Ca.*
1301 *Magnetobacterium bavaricum*-like (G) isolated from various freshwater (B, C, E and G) and
1302 marine (A, D and F) environments. Magnetotactic cells in (A-E and G) produce elongated
1303 prismatic (A-C and E) or bullet-shaped (D and G) magnetite magnetosomes while the MMP (F)
1304 produce octahedral greigite magnetosomes. Scale bars represent 0.5 μm.

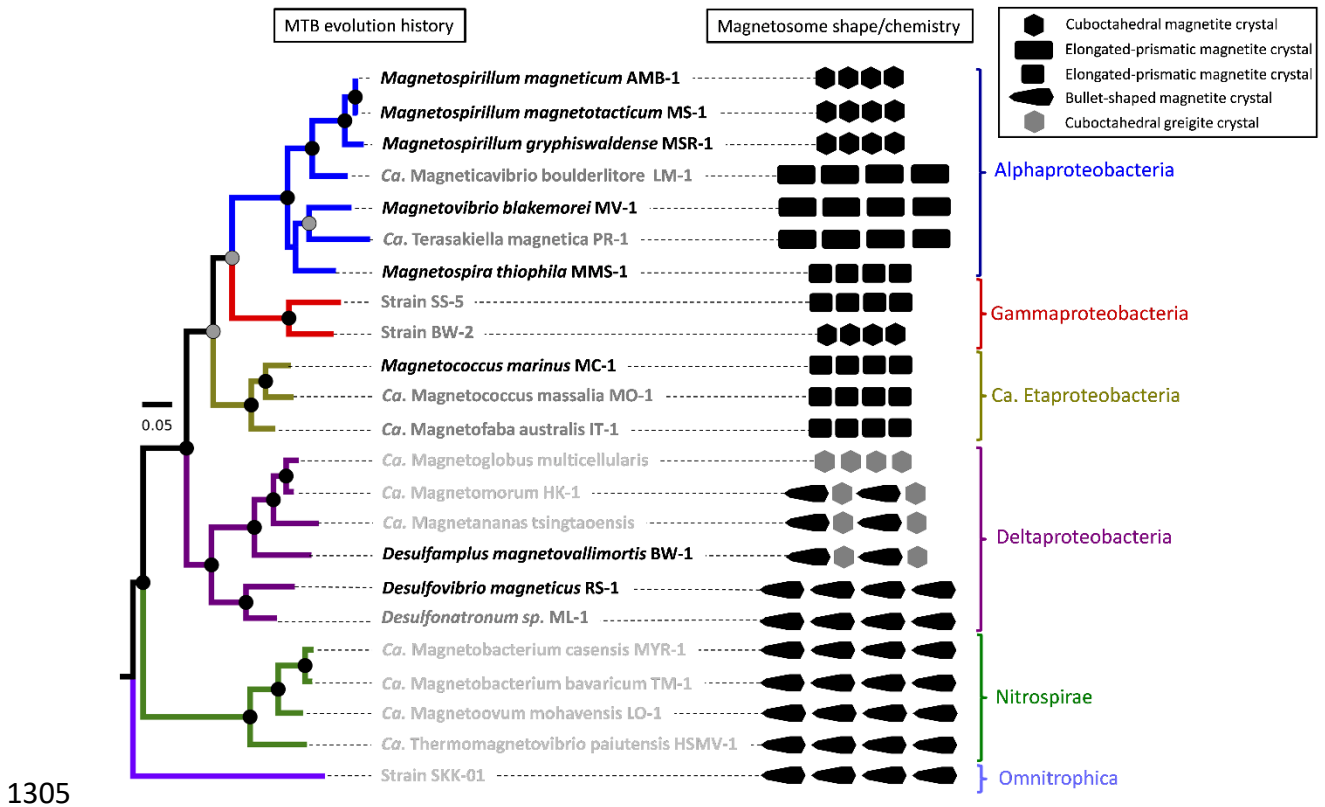
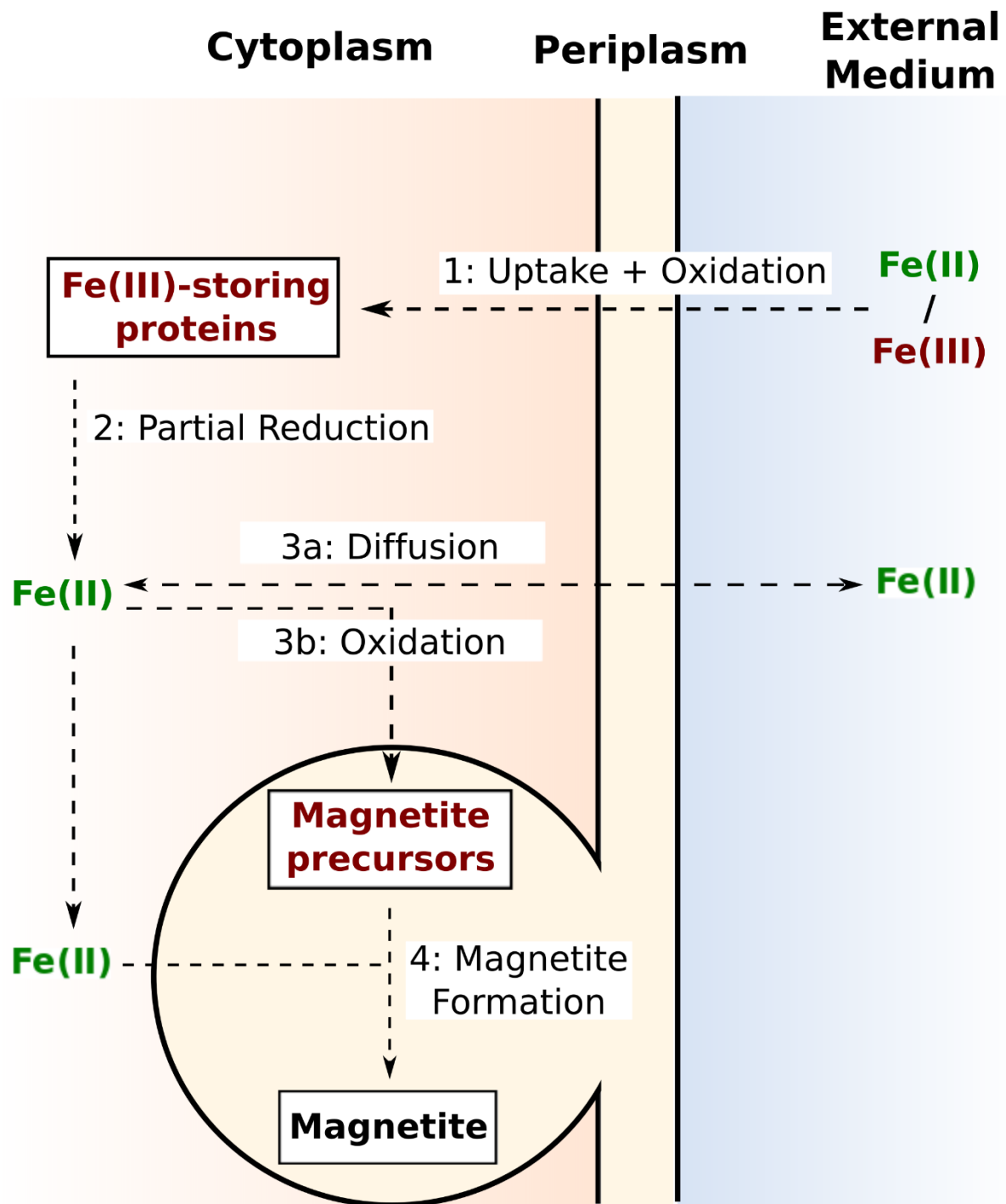


Fig. 2 Correlation between MTB evolution and magnetosome shape and composition. The maximum likelihood tree was built with 16S rRNA gene sequences of selected MTB for which magnetosomes were already characterized. The tree was rooted with the Omnitrophica sp. strain SKK-01. Cultured and taxonomically characterized MTB are written in dark, cultured but not yet taxonomically characterized MTB are written in dark gray while uncultured MTB are written in light gray.



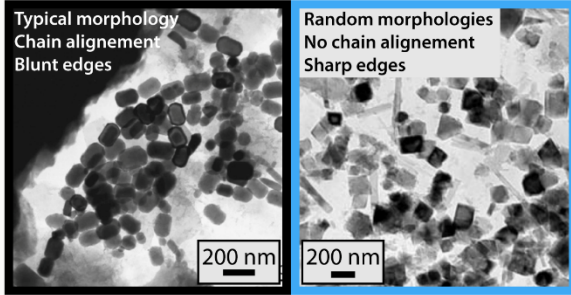
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1314 **Fig. 3** Schematic model of iron biomineralization in MTB. Reduced or oxidized species are
 1315 incorporated into bacteria and stored as Fe(III) species. Partial reduction of this pool leads to
 1316 production of Fe(II) for trafficking to magnetosomes. Magnetite and its potential precursors
 1317 are precipitated from oxidation of Fe(II) which could happen either in the cytoplasm or in the
 1318 magnetosome membrane.

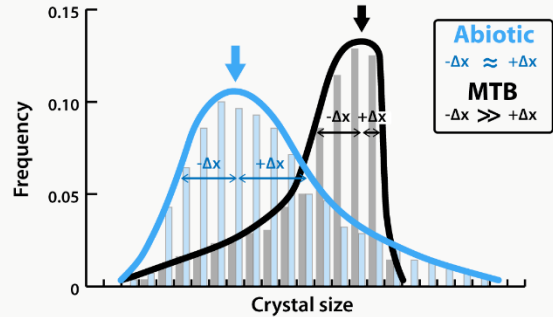
MORPHOLOGICAL CRITERIA

Abiotic/DIRB vs. MTB

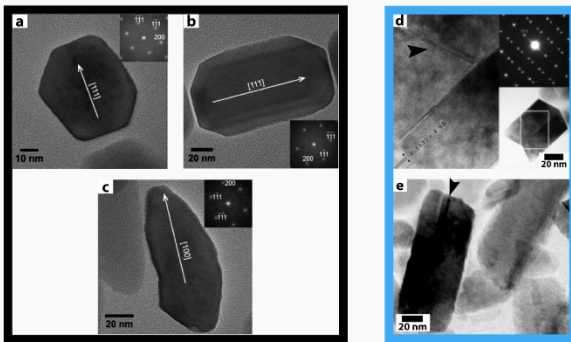
A TEM description



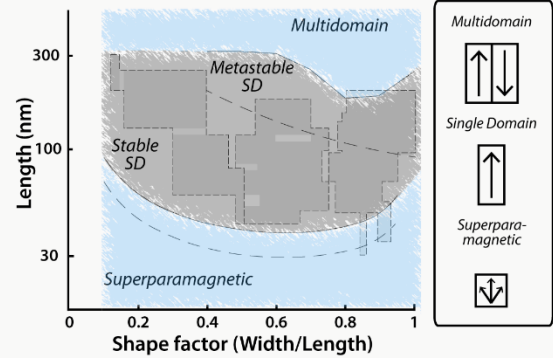
B Theoretical crystal size distribution



C Crystallographic perfection

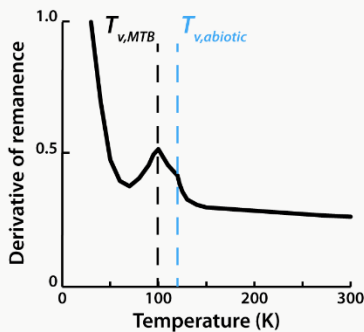


D SSD magnetite

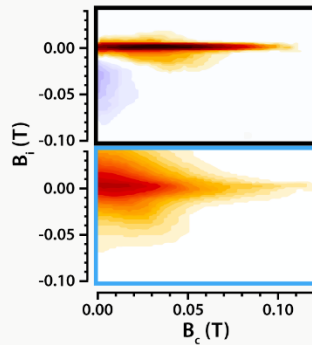


MAGNETIC CRITERIA

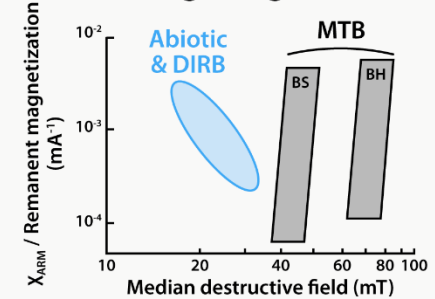
E Verwey temperature



F FORC diagrams

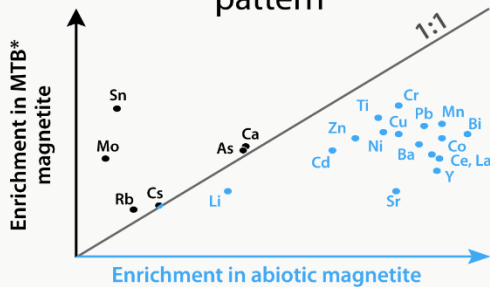


G IRM and ARM analyses (Egli diagram)

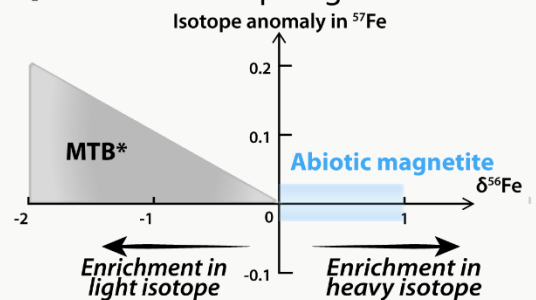


CHEMICAL AND ISOTOPIC CRITERIA

H Trace element incorporation pattern

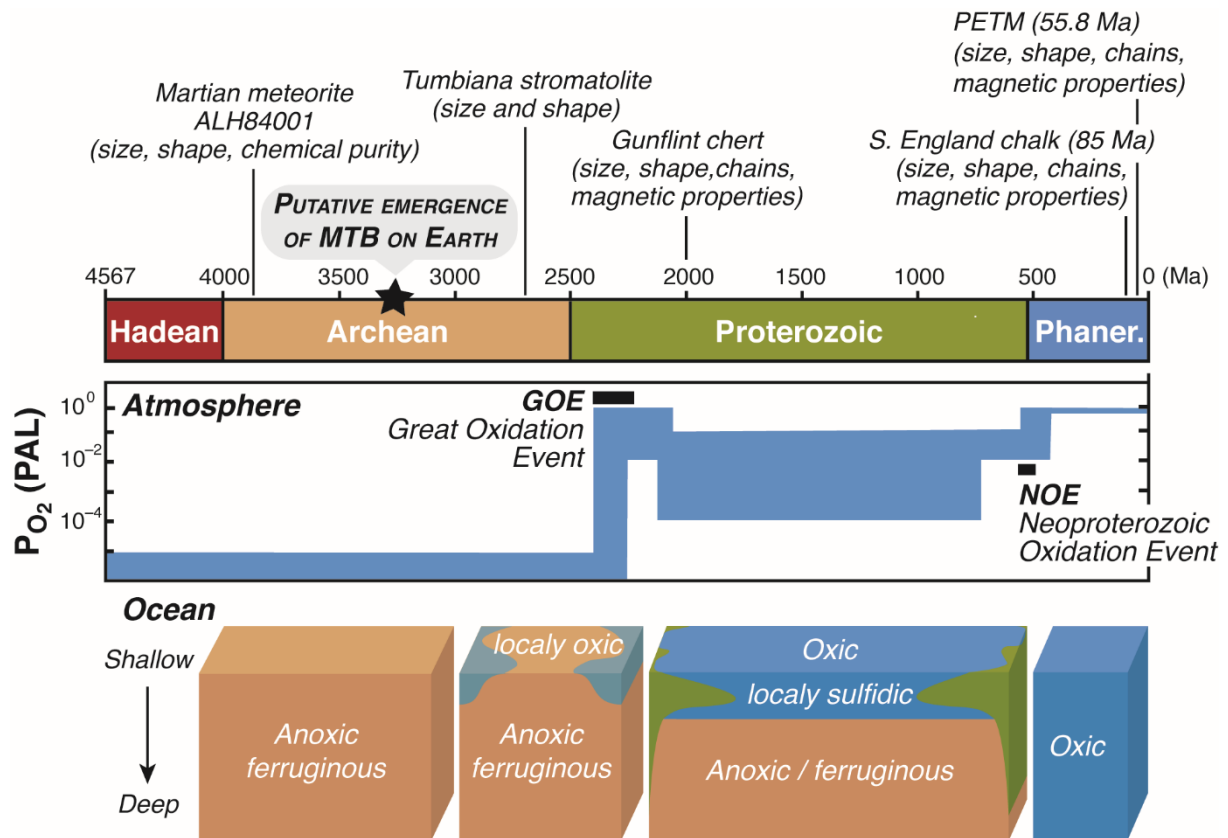


I Iron isotope signature



1320 **Fig. 4** Morphological, magnetic, chemical and isotopic criteria used to distinguish MTB
1321 magnetite (in black) from abiotic/induced magnetite (in blue). **Morphological criteria** (A) TEM
1322 images of Miocene magnetofossils from South Atlantic Ocean sediments (on the left; Kopp
1323 and Kirschvink, 2008) and abiotic magnetite precipitated from FeSO₄ solution (on the right;
1324 Arató *et al.*, 2005). (B) Theoretical size distribution histograms showing specific patterns for
1325 MTB magnetite and abiotic precipitation of magnetite. (C) Crystal lattices evidenced in (d)
1326 abiotically-precipitated twinned nanocrystal of magnetite (Devouard *et al.*, 1998) and (e),
1327 DIRB magnetite (Vali *et al.*, 2004); while (a-c) MTB nanocrystals of magnetite from different
1328 species exhibits no internal defects (Li *et al.*, 2013). (D) Length versus shape factor
1329 (width/length ratio) for magnetite nanocrystals. MTB magnetites mostly fall within the stable
1330 single-domain (SSD, grey zone). Superparamagnetic domain (SP) and multidomain (MD) can
1331 also contain some bacterial nanocrystals as shown by the grey squared areas obtained by
1332 measuring magnetosomes. **Magnetic criteria** (E) Verwey transition of MTB magnetite (above
1333 100°K) and abiotic magnetite (above 120°K) occurs at two different temperatures (Chang *et*
1334 *al.*, 2016). (F) FORC diagrams illustrating the specific pattern expected from MTB magnetite
1335 (upper diagram) and an example containing significant amount of detrital magnetite (lower
1336 diagram) (from Lascu *et al.*, 2015). The axes represent the grain microcoercivity (BC) and the
1337 interaction field illustrating magnetostatic interactions between particles (Bi). (G) Isothermal
1338 remanent magnetization (IRM) and anhysteretic remanent magnetization (AMR) measured
1339 on samples allow to construct Egli Diagram. This representation separates clusters of MTB
1340 magnetite (biogenic soft, BS, with low coercivity, and biogenic hard, BH, with a higher
1341 coercivity mostly linked with their elongation) from other forms, but this technique is time-
1342 consuming and complex (Roberts *et al.*, 2019). **Chemical and isotopic criteria** (H) Trace
1343 element partition coefficients normalized to iron content for MTB and abiotic magnetite. The

1344 grey line indicates similar trace element enrichment in both biotic and abiotic magnetites
1345 relative to the growth medium (Amor *et al.*, 2015). MTB* stands for chemical signatures
1346 tested on AMB-1 only. (I) Iron isotope composition measured on magnetite produced by the
1347 AMB-1 strain (Amor *et al.*, 2018) and abiotic magnetite (Friedrich *et al.*, 2014). Abiotic
1348 magnetite is enriched in heavy isotopes with no anomaly on ^{57}Fe while AMB-1 magnetite is
1349 enriched in light isotopes and exhibits a ^{57}Fe anomaly. Microscope images were reproduced
1350 with permission.
1351



1352

1353 **Fig. 5** Main geological eons on Earth's history, evolution of atmospheric oxygen (Kump, 2008 ;
 1354 Lyons et al., 2014), ocean chemistry (Poulton and Canfield, 2011; Lyons et al., 2014) and
 1355 occurrences of magnetofossils in the deep time (see main text for details). Oxygen partial
 1356 pressure expressed in percent of PAL (Present Atmospheric Level).