

Intraplate orogenesis as a driver of multistage karst-hosted mineralization: the Imini manganese case (Atlas, Morocco)

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1	Intraplate orogenesis as a driver of multistage karst-hosted mineralization: the Imini manganese case
2	(Atlas, Morocco)
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25 Abstract

26 The Imini mining district (southern foreland of the intraplate Atlasic belt of Morocco) hosts the largest Mn 27 resources of North Africa, consisting of two laterally extensive bodies of high-grade pyrolusite-rich manganese 28 ore and a third discontinuous medium-grade coronadite-rich Mn ore hosted in a ~10-15 meters thick Cenomanian-29 Turonian dolostone unit. Until now, the origin and timing of the Mn ore have been poorly constrained. New Pb 30 isotopic ratios show that Triassic series (basalts and red sandstone) are likely the source of the metals. ⁴⁰Ar/³⁹Ar 31 dating of K-Mn oxides shows that the Mn-rich orebodies formed during at least three periods: late Cretaceous to 32 late Paleocene (>58 Ma), late Eocene (ca. 36.3 Ma), and early Burdigalian to early Serravalian probably in two 33 pulses at ca. 19–20 Ma and ca. 13 Ma. These periods coincide with three known building phases of the Atlasic 34 relief during late Cretaceous, late Eocene and the Early(?)-Middle Miocene. We therefore propose the Atlasic 35 tectonics as the first-order control of the Mn mineralization. Periods with regionally high elevations modified the 36 climate to wetter conditions that supplied meteoric water to feed temporary aquifers. Relief building created the 37 required hydraulic head to sustain (1) fluid-rock interaction between O_2 -poor acidic fluids and the Triassic series, 38 (2) migration of the metal-rich fluid and (3) to overpressure fluid in the Imini depositional site. The decreasing 39 thickness of Triassic series in front of the Imini anticline forced these low-temperature (<100°C) fluids to mix 40 with oxygenated and alkaline ground waters in the karst system and precipitate Mn oxides. The N70°-oriented 41 Atlasic tectonic structure controls the orientation of the Mn deposits. The late Eocene - Early/Middle Miocene 42 uplifts generated additional supplies and/or in-situ remobilization of the primary late Cretaceous medium-grade 43 ore to form the high-grade pyrolusite-rich ore.

44 Keywords: ³⁹Ar/⁴⁰Ar dating, Pb isotopes, geodynamics, ore deposits, K-Mn oxides

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46 Introduction

47 Understanding the genesis and evolution of an ore requires different components of the metallogenic system: 48 fertility of the source(s), transport and concentration of metals including mineralizing fluid movement, 49 geodynamic setting and triggering events, accumulation and preservation of the metals (e.g., Laznicka 2014; 50 McCuaig et al. 2018). The geodynamic phases that affect the distribution of rocks and the building of relief may 51 modify, distance, or remove the source rocks. These changes can also determine the preferential paths followed 52 by the mineralizing fluids and consequently control the timing and duration of metal deposition (e.g. Groves and 53 Bierlein, 2007). The timing and identification of metal sources are required to describe and characterize the 54 mineral system and its connection to geological settings.

55 The karst-hosted Imini manganese district is a good example to investigate the geodynamic control of the 56 mineralization process as (i) it is located in the vicinity of the intraplate Atlas orogenic belt of Morocco (Frizon 57 de Lamotte et al. 2000) (Figs. 1 and 2), where the tectonic history is relatively well-constrained (Leprêtre et al. 58 2015), (ii) K-bearing Mn oxides in the ore can be dated and (iii) several potential source rocks are available 59 (Gutzmer et al. 2006; Dekoninck et al. 2016b, a, 2021). Several models have been proposed to explain the high-60 grade Mn ore hosted in the ~10-15 m-thick Cenomanian-Turonian (CT) dolostone involving sedimentary-61 diagenetic and hydrothermal models (Dekoninck et al. 2016b). According to Gutzmer et al. (2006) and Dekoninck 62 et al. (2016a, 2021), the Mn mineralization process involved multistage karst-related epigenesis by mixing of low-63 temperature groundwater and oxidizing meteoric water. The Imini district has the highest economic Mn grades of 64 North Africa due to the presence of two pyrolusite-rich stratabound ore (C1 and C2), and a third horizon (C3) 65 predominantly composed of coronadite group minerals (Beaudoin et al. 1976). The C1 and C2 ore occur over a 66 \sim 25–30 km strike length forming the Imini district (Fig. 2a), whereas the C3 ore occurs discontinuously over 67 ~100 km from Imini to Tasdremt (Dekoninck et al. 2021)(Fig. 1). The timing of the Imini mineralization has not 68 been investigated so far despite the presence of K-Mn oxides. In the Tasdremt area (Fig. 1b), Dekoninck et al. 69 (2021) have shown that the Mn ore is similar to the C3 ore at Imini and formed during three mineralizing periods 70 at ~91.5 Ma, ~77.5-82 Ma, and ~65-67 Ma. Likewise, source rocks of the Mn mineralization have remained speculative (Gutzmer et al. 2006; Dekoninck et al. 2016b, 2021). The possible sources include : (i) the voluminous 71 72 Neoproterozoic volcanic basement hosting numerous Mn-rich veins in the Anti-Atlas (Choubert and Faure-Muret, 73 1973) or (iii) the large Triassic basaltic rocks of the Central Atlantic Magmatic Province and red sandstone in the 74 Atlas (CAMP; Marzoli et al., 2019).

The aim of this study is to propose an integrated model of the karst-hosted Imini manganese deposits, from the source to ore deposition, in light of a structural study in the Imini area (Leprêtre et al. 2015) and detailed petrographic/mineralogical investigations of the manganiferous orebodies (Dekoninck et al. 2016a, 2021), with new Pb isotope constraints and ⁴⁰Ar/³⁹Ar dating of K-bearing coronadite group minerals.

79 1. Geological setting

80 1.1. Regional geology

81 The Imini Mn district is located at the junction of the High Atlas and the Anti-Atlas (Figs. 1 and 2a), along 82 the western end of the Cenozoic Ouarzazate foreland basin filled by Cenozoic syn- and post tectonic rocks (Figs. 83 1 and 2a). To the north, the area consists of thick Triassic series with basaltic rocks and Fe-rich sandstone overlain 84 by Jurassic carbonates, Cenomanian red sandstones, Cenomanian-Turonian (CT) carbonates (~10-15 m; Fig. 2b), 85 Senonian series (~160 m), of interbedded marls, evaporites and dolostones(Algouti et al., 1999; Rhalmi, 1992), 86 Eocene and Miocene continental to shallow marine rocks as well as Plio-Quaternary alluvium (El Harfi et al. 87 2001) (Figs. 2a and c). The cumulated thickness of the late Cretaceous to Pliocene sediments does not exceed 380 88 meters in the Imini area. Triassic red sandstone and basaltic rocks, together with Liassic limestones thin to the 89 south (Fig. 2c). They pinch out against Neoproterozoic or Ordovician rocks along the Imini anticline, with a N90° 90 strike becoming ENE-WSW east of the Imini anticline (Fig. 2c; Missenard et al. 2007; Leprêtre et al. 2015). The 91 CT carbonates overlie either conformably the poorly dated Lower Cretaceous red sandstones or unconformably 92 the Neoproterozoic volcanic rocks of the Anti-Atlas (Ouarzazate Group)(Thomas et al. 2004) or Ordovician shales 93 (Figs. 2a and c). The CT carbonate succession does not exceed 10-15 m in thickness and has a strong terrigeneous 94 component (Fig. 2b; Rhalmi et al., 1997). Senonian formations consist of more than 100 m of red to pink 95 argillaceous sandstones, clays and evaporites (Fig. 2a) likely of Upper Santonian age (Algouti et al., 1999). These 96 rocks are covered by >100 m-thick marine and siliciclastic Eocene carbonate, and Miocene lacustrine carbonate 97 and conglomerate (El Harfi et al. 2001).

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1.2. Geodynamic evolution

Early late Cretaceous deformation was documented in several studies (Martin 1982; Froitzheim 1984; Froitzheim
et al. 1988; Herbig 1988; Algouti et al. 1999; Fekkak et al. 2018; Cavallina et al. 2018). After an onset of
deformation during the late Cretaceous, three tectonic pulses are reported in the High Atlas (Leprêtre et al. 2015,
2018). These events are in broad agreement with tectonic events along the whole Atlas belt and elsewhere in the
other domains of Morocco (Frizon de Lamotte *et al.*, 2000, 2009). A late Eocene event resulted in large folds with

low amplitude along a ~N70° direction. An early to middle Miocene uplift induced cover deformation resulting
in gravity sheets and "décollement" nappes, recorded in the Marrakech High Atlas, only. Finally, the PlioQuaternary shortening reactivated basement faults (Figs. 2a and c). These inherited ~N70° striking faults were
possibly also active during the Miocene (Leprêtre et al. 2015; Lanari et al. 2020).

108 Three main deformation styles are recognized in the Atlas of Morocco (Frizon de Lamotte et al. 2000, 2008, 109 2009; Leprêtre et al. 2015). First, the late Cretaceous tectonic regime was responsible for low stand reliefs in 110 association with lithospheric buckling, under the compressional regime due to Africa-Europe convergence (Frizon 111 de Lamotte et al. 2009; Leprêtre et al. 2018). Second, the Late Eocene and Plio-Quaternary accounted for 112 orographic relief when compressional tectonic led to significant faulting and folding,. Third, the Miocene style 113 was, in part, due to thermal uplift, which might be controlled by a lithospheric anomaly (Missenard et al. 2006; 114 Fullea et al. 2010; Lanari et al. 2020, 2022). The late Cretaceous and Miocene phases are thought to have led to 115 minimal deformation of the foreland basins (Fig. 1) and are likely associated with limited erosion.

116 The main regional tectonic structure of the Imini area is an E-W anticline – the Imini fold (Figs. 2a and c). 117 The rocks and Mn orebodies around this structure are cut by faults and flexures (Missenard et al. 2007). The 118 southern limb of the Imini anticline shows higher deformation withlocal vertical bedding including the CT 119 dolostone and associated Mn orebodies (Figs. 3a-b; Timkit site; Leprêtre et al. 2015). Manganese deposits are 120 folded on the northern limb of the anticline (Figs. 2a, 3b). Post-Senonian deformation is marked by steep drag 121 folding of the manganese-bearing carbonate unit and the overlying Senonian red beds. Cenozoic shortening led to tectonic inversion of these structures and numerous N70° small-scale flexures of metric to decametric amplitude 122 123 (Fig. 3c). In the Lantenois mining site (Fig. 2a), a flexure of the CT dolostone marks the boundary between 124 mineralized and barren zones (Fig. 3c). These structures fold the CT, Senonian and Eocene cover and display 125 symmetric cylinder shape (Figs. 2a).

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1.3. Manganese ore style and paragenesis

The manganese ores are hosted in a 10–15 m thick Cenomanian-Turonian (CT) dolostone unit (Thein 1990; Dekoninck et al. 2021, Fig. 1b). In the Imini area, the manganese karst-hosted orebodies are aligned along an ENE-WSW strike forming a narrow horizon (25–30 km in length with a thickness of 400-1000 m; Figs. 2a and 3). Mining activity is mostly concentrated on the C1 and C2 levels (Figs 3a-d), which have the highest Mn grades (74-92% MnO₂) due to the massive late stage pyrolusite (Fig. 4). The uppermost C3 level (Figs. 3a, d and e) is predominantly composed of coronadite group minerals [(K,Ba,Pb)Mn₈O₁₆: cryptomelane, hollandite and 133 coronadite] that provide a metallurgic ore (40-48% Mn; Gutzmer et al. 2006; Rhalmi et al. 2011; Dekoninck et 134 al. 2016b). Extensive stockwork mineralization occurs between the Mn orebodies (Gutzmer et al. 2006). Both 135 pyrolusite and coronadite group minerals occur in the three orebodies and in fractures (Gutzmer et al. 2006; 136 Dekoninck et al. 2016a). In the early stage, coronadite group minerals replace two generations of dolomite while 137 preserving their original texture (Dekoninck et al., 2016a; Fig. 5a). This stage is followed by the neoformation of 138 K-Mn oxides in open spaces (stage 2), forming colloform concretions of hollandite alongside pyrolusite beds 139 (Dekoninck et al. 2016a) (Figs. 5b-c). Intense brecciation is observed and is related to the late stage crystallization 140 of Mn oxides and calcite (Dekoninck et al., 2016b; Gutzmer et al., 2006; Figs. 4 and 5b-c). Romanechite 141 [(Ba,H₂O)₂Mn₅O₁₀], lithiophorite [(Al,Li)MnO₂(OH)₂] and goethite are accessory minerals (Dekoninck et al. 142 2016a), the latter being present in the northwestern part of the Imini area (Bouladon and Jouravsky 1952; Pouit 143 1964). Goethite occurs before the Mn oxides (Fig. 4) and has been dated between ca. 80 to ca. 50 Ma (Verhaert 144 et al. 2022). The association of these oxides with dissolution features confirms the karst-hosted model for the 145 Imini deposits (Gutzmer et al. 2006). Gutzmer et al. (2006) reported that Senonian sediments filled karst cavities 146 and were not affected by Mn mineralization.

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2. Sampling and analytical procedure

148 2.1. ⁴⁰Ar/³⁹Ar geochronology

Potassium-bearing Mn oxides have been collected from six sampling sites, from east to west (Fig. 2a): Bou
Aguioun (6), Timkit (4), Lantenois (3), Tiffersine (5), Far West (1) and Plateau (1). At Tiffersine , the analyzed
K-Mn oxides coat colloform goethite (Fig. 5c)(Table 2).

152 Coronadite group minerals have been collected in the three stratabound orebodies (C1, C2 and C3) or in connected 153 stockwork mineralization (Fig. 2b; Table 1). All dated minerals belong to the second stage of Mn mineralization 154 (Figs. 4 and 5; Dekoninck et al., 2016b, 2016a). None of these samples comes from internal karstic sediments. 155 The dated coronadite group mineral samples form several micrometric bands (Figs. 5a-d), consisting of 156 micrometric needles (1µm in diameter and 3–500µm long; Figs. 5b and d; Dekoninck et al., 2016a) growing at 157 the surface of the first epigenetic mineralization stage (Fig. 5d). Potassium contents in these samples, based on 158 EDS and ICP-MS analyses (Dekoninck et al., 2016b), range from 0.43 wt. % to 4.88 wt. % K₂O. The inner (C) 159 and outer (EX) parts of two colloform aggregates were investigated in order to estimate the growth rates (IM98 160 and ECH-101-b; Fig. 5b). Five experiments were duplicated (18TIF02, 18TIF03, IM41d, ECH-101-b-C and ECH-161 101-b-EX). Pyrolusite belongs to the second stage of mineralization (Figs. 4 and 5d). Quartz and dolomite grains

are occasionally disseminated in the Mn oxides (Fig. 5a and d). Late calcite veins cut across Mn oxides (Figs. 5a, c and d). The texture and composition of the K-Mn oxides were checked using a JEOL 6010 LV and 7500-F Scanning Electron Microscopes (SEM) coupled with an ULTRA MINI-CUP Energy Dispersive Spectrometer (EDS) at the Microscopy Service of the University of Namur. 40 Ar/ 39 Ar analyses were performed on single millimeter-scale fragments of Mn oxides using a CO₂ laser probe coupled to a MAP215 mass spectrometer. Details of the 40 Ar/ 39 Ar methodology are in electronic supplementary material (ESM S1). The ages are summarized in Table 1. Raw data are available in ESM S3.

169 2.2. Lead isotope geochemistry

170 Lead isotope were analysed on thirty-seven whole rock samples at the Laboratoire G-Time (Université Libre de 171 Bruxelles, ULB, Belgium) with a Plasma II HR-Multi-Collector Inductively Coupled Plasma Mass Spectrometer 172 (Nu Instruments). These include: (i) the Neoproterozoic volcanic and plutonic basement rocks of the northern 173 Anti-Atlas, (ii) the late Neoproterozoic Mn vein deposits of the northern Anti-Atlas, (iii) the Cambrian basalts, 174 (iv) the Cambrian-Ordovician shales of the Atlas, (v) the Triassic CAMP basalts and interbedded red evaporitic 175 sandstones, (vi) the CT dolostone hosting the Tasdremt and Imini Mn mineralization and (vii) the Mn ores of the 176 Imini and Tasdremt areas. We have collected the least altered samples in outcrops but basalts were subjected to 177 alteration/weathering. We additionally leached basalt samples (Triassic and Cambrian) to avoid any 178 alteration/weathering effects on the pristine isotopic signatures (ESM S2). See ESM S1 for analytical details and 179 ESM S4 for geochemistry of the samples.

180 **3. Results**

181 **3.1.** ⁴⁰Ar/³⁹Ar dating

The complex age spectra obtained in this study reflect the coexistence of several radiogenic components of different ages in the samples analysed. This may be at the origin of many geologically significant ages. A detailed analysis of the ⁴⁰Ar/³⁹Ar data, based on the works of Vasconcelos et al. (1995, 1999), Ruffet et al. (1996), Hénocque et al. (1998), Colin et al. (2005), Beauvais et al. (2008), De Putter et al. (2015) and De Putter and Ruffet (2020), is therefore essential to detect, identify and possibly validate these radiogenic components.

187 The age spectra of two samples from the Timkit site (IM37 and IM39), which have very pronounced saddle shapes 188 (Fig. 8g-h), have to be rejected. The convex trajectory of the degassing steps in the inverse isochron diagram 189 reflects an intense recoil of 39 Ar_K during irradiation. The presence of a contaminant or inherited component cannot

be excluded.

With the exception of samples IM98-EX, 18TIF03 or 18TIF13, all age spectra show abnormally high apparent ages in the low temperature steps, which rapidly decrease in the first 20% of 39 Ar_K degassing (Figs. 6, 7, 8 and 9). Their possible significances, not relevant to this study, have been discussed in detail by Dekoninck et al. (2021). Three groups of 40 Ar/ 39 Ar analyses can be distinguished allowing the identification of three radiogenic components: (i) late Cretaceous to early Paleogene, (ii) Priabonian and (iii) Burdigalian-Serravalian (Table 1). We first present the Priabonian age group because it provides crucial information for the characterisation of the other two .

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3.1.1. Priabonian radiogenic component

This group (Fig. 6) consists of Mn oxides growing on goethite (Fig. 5c). Samples were collected in the C3 orebody at Tiffersine (18TIF03, 05 & 12) and from the Plateau site (18PL12b; Fig. 2a). 40 Ar/ 39 Ar experiments are characterized by (i) 37 Ar_{Ca}/ 39 Ar_K ratios in the range of 0.5-2 (Fig. 6b-e), significantly higher than those of the other samples from this study, (ii) high atmospheric contamination rates and (iii) low K-content. The main radiogenic component is Priabonian in age as shown by the plateau age at 36.3 ± 1.1 Ma (Fig. 6c).

204 The Priabonian radiogenic component is neither the oldest nor the youngest in the analysed samples (Fig. 6a). The 205 low to medium temperature steps of one of the age spectra of sample 18TIF03 (Fig. 6a), and the age spectrum of 206 sample 18TIF12 (Fig. 6d) give pseudo-plateau ages at 35.6 \pm 1.8 Ma, and 35.5 \pm 0.9 Ma, respectively. 207 Nonetheless, the high temperature steps with apparent ages as old as ca. 51 Ma suggest that the Priabonian 208 component is superimposed on an older one (Fig. 6d), likely to be Paleocene to late Cretaceous in age. A younger 209 component is also superimposed on the Priabonian component. This is observed in the steady increase in apparent 210 ages of the second age spectrum from sample 18TIF03 (Fig. 6b). This feature is characteristic of supergene K-Mn 211 oxides, when two distinct age phases co-exist (De Putter et al., 2015). This age spectrum provides an overestimate 212 of the age of the youngest component (< ca. 24.1 Ma), possibly of Miocene age, as well as an underestimate of 213 the age of the oldest component (> ca.33.9 Ma), possibly of Priabonian age. At the same time, the ${}^{37}\text{Ar}_{\text{Ca}}/{}^{39}\text{Ar}_{\text{K}}$ 214 ratios increase from ca.0.8 to ca.1.8, confirming the increasing input of a Ca-rich component, in line with the 215 Priabonian component. The Miocene disturbance is also highlighted by analysis of sample 18PL12b (Fig. 6e). 216 The low to medium temperature steps define a pseudo-plateau at 14.3 ± 0.1 Ma, which is not validated by the isochron calculation [(³⁶Ar/⁴⁰Ar vs. ³⁹Ar_K/⁴⁰Ar*; Hanes et al., 1985; Roddick et al., 1980; Turner, 1971 -217 Atmospheric ratio ${}^{40}\text{Ar}/{}^{36}\text{Ar} = 298.56$ according to Lee et al. (2006)]. The high temperature steps allow the 218 calculation of a pseudo-plateau age of 36.8 ± 0.6 Ma. The age and ${}^{37}\text{Ar}_{Ca}/{}^{39}\text{Ar}_{K}$ spectra have similar shapes. The 219

increase in ${}^{37}\text{Ar}_{\text{Ca}}/{}^{39}\text{Ar}_{\text{K}}$ ratios during degassing, from ca. 0.1 to ca. 0.9, highlights the Ca-rich signature of the high temperature radiogenic component (Fig. 6e). This is consistent with what is expected for the Priabonian component. The frequency histogram (probability density diagram) of the apparent ages (Fig. 6a) synthesizes information about the Priabonian component.

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3.1.2. Miocene radiogenic component

Samples IM98, ECH101b, ECH102b, ECH103 and ECH104, collected in the C1 horizon and the stockwork of the Bou Aguioun area, as well as samples 18TIF02 and 18TIF13 collected from the C3 horizon of the Tiffersine area (Figs. 2a and 7b-h) have dominant Miocene radiogenic components. Most of them have ${}^{37}Ar_{Ca}/{}^{39}Ar_{K}$ ratios in the range 0.001-0.1.

The Miocene radiogenic components span a period of ca. 7 Myr, between ca. 20 Ma and ca.13 Ma, from Burdigalian to early Serravallian periods. Their ages depend on the sampling areas. In the Bou Aguioun area (C1 and stockwork mineralization) (Fig. 7b-d, g-h; Table 1), the samples yield mostly early Burdigalian ages in the range 19–20 Ma with 5 plateau ages supported by the isochron calculation.

233 In contrast, the outer (EX) domain of the IM98 colloform aggregate (Fig. 7b) yields an apparent plateau age at 234 15.0 ± 0.1 Ma, which is ca. 4.7 Ma younger than the inner domain (IM98-C; Fig. 5b). Although validated by 235 isochron calculation, the subtle hump shape of the age spectrum of IM98-EX indicates mixing between two 236 radiogenic components of different ages (Wijbrans and McDougall 1986; Ruffet et al. 1996; Vasconcelos 1999; 237 Beauvais et al. 2008). The older component is mostly expressed in the middle temperature domain of the age spectrum and would be older than ca. 15 Ma, whereas the secondary component, whose influence is perceptible 238 239 in the low and high temperature steps, is younger than ca. 14 Ma. This attempt to evaluate the duration of 240 precipitation is unsuccessful. Duplicate analyses of the ca. 15mm colloform aggregate ECH101-b (Fig. 7h) show 241 that there are no significant differences between the age of the core (C) and the outer rim (EX). All age spectra 242 define a frequency peak of apparent ages at ca. 19.3 - 20.5 Ma (Fig. 7h).

Samples 18TIF02 and 18TIF13 collected at Tiffersine in the C3 orebody (Figs. 2 and 7e-f) provide contrasting data to those previously mentioned, both in terms of age spectra shape and apparent ages. Sample 18TIF02 (Fig. 6e) yields a flat age spectrum (no plateau) with a mean age at 16.0 ± 0.1 Ma which is not validated by the isochron calculation. The two other age spectra from sample 18TIF02 show staircase shapes with apparent ages that increase up to ca. 26.5 Ma. The flat low temperature steps of samples 18TIF02 and 18TIF13 yield two concordant pseudo-plateau ages at 13.0 ± 0.3 Ma and 13.1 ± 0.3 Ma, respectively. The degassing peak associated with the pseudo-plateau of sample 18TIF02 shows that a mineral component would have crystallized in the early Serravallian (ca. 13 Ma) (Fig. 7e-f). Its radiogenic component is characterized by ${}^{37}\text{Ar}_{Ca}/{}^{39}\text{Ar}_{K}$ (#Ca/K) ratios in the range of 0.001-0.1. It is also superimposed on a Ca-rich (${}^{37}\text{Ar}_{Ca}/{}^{39}\text{Ar}_{K}$ ratios in range 0.1-1) and older (> ca. 26.5 Ma) radiogenic components (probably Priabonian). We observe the same effect in the high temperature steps for sample ECH101-b (core) (Fig. 7h). The ${}^{37}\text{Ar}_{Ca}/{}^{39}\text{Ar}_{K}$ ratios in range 0.1-1 throughout the degassing spectrum of sample 18TIF13 (Fig. 7f) suggest that the pseudo-plateau age at ca.17.4 Ma may be a mixture between the Serravalian and Priabonian radiogenic components.

Although sampling may introduce statistical bias, the results show that there are two distinct radiogenic components: Burdigalian and Serravallian. Detailed data processing suggests that apparent "intermediate" ages in the range ca.14.3-17.4 Ma have no geological significance (Fig. 7a). The Burdigalian radiogenic component is only observed in the C1 horizon and in the Bou Aguioun stockwork mineralization, whereas the Serravalian radiogenic component is predominantly observed in the C3 horizon at Tiffersine (Fig. 7a) and Plateaux sites (Fig. 6a).

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3.1.3. Late Cretaceous (to early Paleogene?) radiogenic components

263 Age spectra (Fig. 8b-h) from samples from C1-C2 at Timkit (IM36, IM37, IM39 and IM40), C2 at Bou Aguioun 264 (18BA01) and C3 orebodies at the Far West (IM41d) and Lantenois sites (LAN18, LAN57 and LAN59) can be 265 interpreted in the light of the geochronological study of the late Cretaceous mineralization of the Tasdremt Mn 266 district (Dekoninck et al., 2021). These authors have identified four radiogenic components: (i) Turonian (ca. 91.5 267 Ma), (ii) Campanian (ca. 82 Ma to ca. 77.5 Ma), (iii) Late Maastrichtian (ca. 67-65 Ma) and (iv) Late Paleocene 268 (ca.58 Ma). The age spectra (Fig. 8b-f) display a staircase shape. This indicates mixing of two radiogenic 269 components: a secondary component in the low-temperature domain is superimposed on a primary component 270 that degasses at higher temperatures. The apparent or pseudo-plateau ages in the low- and high temperature steps 271 provide over- and underestimates of the ages of the secondary component and the primary component, 272 respectively (Fig. 8b-f). Not all samples in this group are comparable. The primary and secondary components 273 are unlikely to be unique, and there may be several disturbances superimposed on the same sample (Fig. 8a) as 274 observed by Dekoninck et al. (2021). We distinguish two subgroups.

(1) In samples IM36 and IM40 (C1-C2 Timkit; Fig. 8d-e), the primary component could be at least as old as
ca.66.6 Ma, comparable to the Late Maastrichtian component identified by Dekoninck et al (2021). Two later
disturbances are identified by two degassing peaks in the low to mid-temperature domain of the IM36 degassing

278 spectrum. They are superimposed on the early degassing domain of the primary component (Fig.8d). The first 279 disturbance is thought to have occurred around ca. 59 Ma and may be comparable to the Late Paleocene 280 component identified in the Tasdremt district (Dekoninck et al. 2021). The second event is thought to have 281 occurred after ca. 54.7 Ma. Analysis of sample IM40 (Fig. 8e) suggests that this disturbance is younger than ca. 282 47 Ma and could be related to the Priabonian and Miocene radiogenic components.

- (2) The flat high temperature segments of the age spectra of samples collected in the C3 ore at Lantenois (LAN18,
 57 and 59)(Fig. 8c, g), Far West (IM41d)(Fig. 8b), and in the C2 at Bou Aguioun (18BA01)(Fig. 8h), suggest that
 the primary component is at least as old as ca. 80 Ma. It is comparable to the Campanian component identified at
 Tasdremt (Dekoninck et al. 2021). The apparent ages or small segments identified in the low temperature steps,
 in the range ca. 61.6-69.3 Ma, suggest that the secondary component could be of middle Paleocene in age.
- 288

3.2. Pb isotope compositions

289 The Pb isotopic ratios vary from 17.266 to 28.263 (²⁰⁶Pb/²⁰⁴Pb), 15.437 to 16.122 (²⁰⁷Pb/²⁰⁴Pb), 37.571 to 43.688 (²⁰⁸Pb/²⁰⁴Pb; Fig. 9; ESM S2). They have been recalculated to take into account the age of the primary ore 290 deposition at 67 Ma (Figs. 9a- b)(Dekoninck et al. 2021). It implies slight changes of the recalculated ²⁰⁶Pb/²⁰⁴Pb 291 and ²⁰⁷Pb/²⁰⁴Pb ratios up to 0.673 and 0.032, respectively, from the measured ratios. The significant changes of 292 293 the recalculated ²⁰⁸Pb/²⁰⁴Pb ratios up to 90.711 is due to samples having high Th/Pb ratio that brings more radiogenic ²⁰⁸Pb. We do not report the recalculated ²⁰⁸Pb/²⁰⁴Pb ratios. However, the ²⁰⁶Pb/²⁰⁴Pb vs ²⁰⁸Pb/²⁰⁴Pb and 294 295 ²⁰⁷Pb/²⁰⁴Pb vs ²⁰⁸Pb/²⁰⁴Pb are compiled and are reported in ESM S6; they show the same trends defined in Figure 296 9 and do not bring additional value to the following interpretations.

The Pb isotopic compositions of the Imini Mn ores cluster in a narrow ²⁰⁶Pb/²⁰⁴Pb range of 18.614-18.772,
²⁰⁷Pb/²⁰⁴Pb 15.619-15.678 and ²⁰⁸Pb/²⁰⁴Pb range of 38.530-39.229. These data are significantly different from
those of Mn ore veins of Bachkoun and Tiouine hosted in Neoproterozoic volcanic rocksand other Neoproterozoic
mineralization (Bouabdellah et al. 2016; Figs. 9a-b).

Neoproterozoic rhyolite and andesite of the Ouarzazate Supergroup display a wide range of more radiogenic isotopic ratios: (²⁰⁶Pb/²⁰⁴Pb) 7.173-24.921 , (²⁰⁷Pb/²⁰⁴Pb) 20.053-28.263 and ²⁰⁸Pb/²⁰⁴Pb 15.663-16.122. The Neoproterozoic granite and granodiorite have different isotopic compositions; sample 15TAG02 has Pb isotopic ratios falling in the range of the Neoproterozoic volcanic rocks, (²⁰⁶Pb/²⁰⁴Pb) 15.658, (²⁰⁷Pb/²⁰⁴Pb) 20.213 and ²⁰⁸Pb/²⁰⁴Pb 38.313, whereas ASK02 has the least radiogenic signature with values of (²⁰⁶Pb/²⁰⁴Pb) 17.266 ,(²⁰⁷Pb/²⁰⁴Pb) 15.437 and ²⁰⁸Pb/²⁰⁴Pb 38.041. The CAMP Triassic basalts are characterized by a wider range of isotopic compositions: (²⁰⁶Pb/²⁰⁴Pb) 15.61215.712 , (²⁰⁷Pb/²⁰⁴Pb) 18.435-19.638 and ²⁰⁸Pb/²⁰⁴Pb 21.879-38.178 , similar to those obtained by Marzoli et al.
(2019) for the Atlas belt (Figs. 9a-b). Leached basalt samples plot in a narrow field due to removing of the
alteration/weathering minerals (Fig. 9c). They have a different isotopic composition than Triassic basalts with
(²⁰⁶Pb/²⁰⁴Pb) 18.267 and (²⁰⁷Pb/²⁰⁴Pb) 15.641.

The Triassic sandstone sample overlaps the isotopic field of the Mn ore $(^{206}Pb/^{204}Pb \ 18.824, \ ^{207}Pb/^{204}Pb \ 15.668, \ ^{208}Pb/^{204}Pb \ 38.820;)$. The CT dolostone samples overlap the compositional field basalts and the Mn ore samples $(^{206}Pb/^{204}Pb \ 18.472-18.692, \ ^{207}Pb/^{204}Pb \ 15.610-15.663, \ ^{208}Pb/^{204}Pb \ 38.255-38.647;)$. The Ordovician shale of the Imini anticline $(^{206}Pb/^{204}Pb \ 18.772, \ ^{207}Pb/^{204}Pb \ 15.678, \ ^{208}Pb/^{204}Pb \ 39.229;)$ has a significantly different isotopic composition than Ordovician shales of the High Atlas $(^{206}Pb/^{204}Pb \ 20.565-20.643, \ ^{207}Pb/^{204}Pb \ 15.817-15.839, \ ^{208}Pb/^{204}Pb \ 43.514-43.688;)$ (Figs. 9a- b).

318 4. Discussion

319 4.1. Source rocks for the metals

320 Among potential source rocks, the CT dolostone displays the most similar Pb isotopic compositions to the Imini 321 Mn ore. These rocks cannot be considered as the source of metals given the low primary Mn-Ba-K-Pb content 322 and their narrow thickness (Dekoninck et al. 2016b). Their Pb isotope composition can be impacted by 323 disseminated Mn oxides (Gutzmer et al. 2006; Dekoninck et al. 2016a) that may overprint the pristine Pb isotope 324 of the dolostone. This is supported by slight enrichments in Mn, Ba and Pb in some dolostone samples (see ESM 325 S4). This similar Pb isotope composition can also be due to fluid-rock interaction with the fluid that precipitated 326 the Mn oxides in the host dolostone. A similar pattern is observed in Ordovician shales in contact with the Mn ore that have similar ²⁰⁶Pb/²⁰⁴Pb and ²⁰⁷Pb/²⁰⁴Pb ratios as the Mn ores, whereas samples away from the district show 327 328 a Pb compositions different from that of Mn ore (Fig. 9a).

Triassic CAMP basalts and interlayered red sandstones are voluminous along the Atlas range of Morocco (Marzoli et al. 2019) and are a likely source for the Mn mineralization. Both rock types fall in the same isotopic field as the Mn ore (Fig. 9b). Locally, basalts show a siliceous alteration characterized by clays, chayesite, quartz-jasper (ESM S7) and a higher dispersion to of their 206 Pb/ 204 Pb and 207 Pb/ 204 Pb ratios (Fig. 9c). These basalts typically have low Mn (0.07-0.47 wt.%; avg. 0.18 wt.% MnO), Ba (53-430 ppm; avg. 190 ppm Ba; 0.05), K (0.21-1.68 wt.%; avg. 0.68 wt.% K₂O) and particularly for Pb (2-58 ppm; avg. 4 ppm Pb; SDR1; Marzoli et al., 2019) contents. Given their similar Pb isotope composition (Fig. 9), the red sandstone is likely a second source of metals with basalts 336 (Fig. 9). Therefore, the source of metals is Triassic and is located in the Atlas, which is crucial to understand the 337 significance of the 40 Ar/ 39 Ar ages.

338 4.2. The Imini ore deposit: a ~90 My multistage history

The ${}^{40}\text{Ar}{}^{39}\text{Ar}$ geochronological study shows three significant periods of Mn deposition/remobilization: late Cretaceous (-Middle Paleocene) (ca. >59 Ma), Priabonian (ca. 36.3 Ma) and Burdigalian-Serravalian (ca. 19–20 Ma and ca.13 Ma, Table 1 and Figs. 6, 7 and 8). There is no age difference between the stratabound and stockwork mineralization, which suggest that they were likely formed together. The relative time frame established by Dekoninck et al. (2016a) shows that most of the pyrolusite postdates the dated K-Mn oxides (Fig. 4). Even if pyrolusite is likely connected to stage 2 open space filling rather than stage 1 (Figs. 4, 5a and d), it means that ${}^{40}\text{Ar}{}^{39}\text{Ar}$ ages document only a segment of the entire petrogenetic history.

346 K-bearing Mn oxides are complex materials able to register several mineralization events at the grain scale. Each 347 event is able to completely or partially erase the previous stage(s)(De Putter and Ruffet 2020). Our ⁴⁰Ar/³⁹Ar 348 geochronological study draws the same conclusion. Although the age of the initial stage of mineralization is 349 imprecise, we identify at Imini two sub-stages that appear to be contemporaneous with the stage 2 (~77.5–82 Ma) 350 and 3 (~65–67 Ma) observed at Tasdremt by Dekoninck et al. (2021). These authors also identified early stage 1 351 of Turonian age (~91.5 Ma) in this area. Verhaert et al. (2022) report at Imini goethite (U-Th)/He ages as old as 92.7 \pm 4.6 Ma which suggest that Mn oxide precipitation may have occurred at the same time. The identical 352 petrographic features (Dekoninck et al. 2016a, 2021) and Pb isotope composition of the Mn ores (Fig. 9b) at 353 354 Tasdremt and Imini argue for a contemporaneous Cretaceous age. However, considering the complexity of the K-Mn oxide ⁴⁰Ar/³⁹Ar data at Imini (Figs. 6, 7 and 8), it is likely that some goethite ages of Verhaert et al. (2022), 355 356 in the range 80-50 Ma, are mixtures of different radiogenic components.

At Tasdremt, where pyrolusite is absent and the Mn grade is low, the Mn ore deposition occurred only in the late Cretaceous (Fig. 1b)(Dekoninck et al. 2021), when, at Imini, the high-grade Mn deposit resulted from a multistage depositional history spanning from the late Cretaceous to the Neogene (Fig. 10). The causes of this different evolution remain speculative but could be related to the erosion of the Triassic Mn source, or alternatively by a different hydrodynamic regime. The duration of these processes highlights the importance of time in creating high-grade karst-hosted deposits.

363 4.3. Relationship between the Imini Mn ore and Atlasic tectonic

364 From the Cretaceous to the present day, four periods of tectonic activity have built the High Atlas. Our results 365 show that three of these four periods correlate with the K-Mn oxide ages in the Imini Mn district (Fig. 10): the 366 late Cretaceous – Late Paleocene (four sub-phases at ~91.5 Ma, ~77.5–82 Ma, ~65–67 Ma and ~ 58–61.5 Ma), 367 the Late Eocene (ca. 36.3 Ma) and the Early to Middle Miocene (ca. 19-20 Ma and ca.13 Ma). Although the 368 timing of some tectonic phases remains debated, the overall correlation between radiometric ages and tectonic/uplift activity, suggests that mineralization was primarily controlled by the geodynamic evolution of the 369 370 High Atlas. The Quaternary tectonic phase would account for recent or modern mineralization in the Imini area 371 (Dekoninck et al. 2016a)(Fig. 4).

High supplies of meteoric water are needed to sustain fluid-rock interactions with the Triassic series, leaching,
transport and finally precipitating metals. The required humid conditions make climate a key parameter. The North
Africa paleoclimate was supposed to be arid during Mn deposition events (Algouti et al. 1999; El Harfi et al.
2001; Mansour et al. 2021; Zammit et al. 2022). However, higher elevated surface can significantly change the
Earth's atmospheric circulation and alter the regional climate (Ruddiman et al. 1997). Therefore, uplift of the
Atlas can lead to a constant supply of meteoric water, as it is case today (Bell et al. 2022).

378 The building of a significant relief triggers the required hydraulic head (Gutzmer et al. 2006) for the percolation 379 of subsurface fluids. In the Atlas of Morocco, Choulet et al. (2014), Verhaert et al. (2017, 2020) and Bouabdellah 380 et al. (2021), highlighted the primary role of Atlasic tectonic activity in the formation of supergene ore. This study 381 confirms the primary role of regional exhumation in subsurface geological processes, enhancing the advection of 382 mineralizing fluids. According to Gutzmer et al. (2006), Mn mineralization forms from low-temperature (<100°C) 383 hydrothermal events. Goethite hydrogen and oxygen stable isotope data confirm that the mineralizing fluid was 384 likely hot meteoric water that experienced fluid-rock interactions with the host dolostone and other country rocks 385 (Verhaert et al. 2022). These conditions differ from lateritic subaerial systems. The difference is also materialized 386 by high Mn oxides growth rates (>15mm/Myr), unlike those observed in lateritic systems (1-5 mm/Myr; Hénocque 387 et al., 1998). This link between uplift and low-temperature mineralization has been shown in Central Africa 388 (Decrée et al. 2010; De Putter et al. 2015; De Putter and Ruffet 2020), South Africa (Vasconcelos and Carmo 389 2018), India (Beauvais et al. 2016; Bonnet et al. 2016; Jean et al. 2019) and Western Europe (Hautmann and 390 Lippolt 2000; Barbarand et al. 2018; Dekoninck et al. 2019).

391

4.4. Geodynamic-related model of the high-grade Imini Mn ore system

392

4.4.1. The late Cretaceous primary Mn deposition

393 The earliest Atlasic deformation likely led the Tasdremt to Imini area (Fig. 1b) to emerge above sea level during 394 the late Cretaceous. These low stand reliefs were flooded several times and resulted in the deposition of sediments 395 during the Upper Santonian, for the whole district, and Campanian in the Tasdremt area (Algouti et al. 1999). We 396 consider that the primary karst Mn deposition took place at ~91.5 Ma, ~77.5-82 Ma, ~65-67 Ma and ca. 58-61.5 397 Ma (Dekoninck et al., 2021 and this study) when the CT succession was in the vadose zone in a terrestrial 398 environment. The C3 ore formed across the whole Imini-Tasdremt Mn district. Except for the earliest 399 mineralization at ~91.5 Ma, Mn was deposited under a cover of sediments, preserving the thin CT host rock 400 dolostone from erosion (Figs. 10 and 11).

401

4.4.2. The Cenozoic enrichment phases

402 The link between geodynamic events and the ⁴⁰Ar/³⁹Ar ages support Mn deposition when mineralizing fluids 403 migrated from the High Atlas relief and fed temporary aquifers in the CT dolostone (vadose environment). By 404 creating a hydrodynamic gradient, meteoric ground water (Verhaert et al. 2022) transported dissolved metals (Mn, 405 Ba and Pb). The massive Mn oxide ore (especially pyrolusite) is associated with discrete ~N70° structures 406 (Leprêtre et al. 2015), which acted as channels responsible for the orientation of the mineralization (Fig. 2a). The 407 porous and partially dissolved dolostone, led low-temperature acidic and reduced groundwater to be buffered and 408 oxidized in a karst environment. Metal precipitation took place where shallow, O₂-rich, and cold meteoric water 409 resident in the karst system mixed with hydrothermal groundwater (Gutzmer et al. 2006). Dekoninck et al. (2016a) 410 have shown that increasing pH due to dolomite dissolution caused precipitation of Mn oxides. In addition, the ore 411 zone is located where Triassic red sandstones and basalts pinch out (Fig. 11). In this geological setting, and under 412 the influence of overpressure Mn-bearing low-T groundwater due to orogenic head (Gutzmer et al. 2006), Mn-413 rich hydrothermal fluids were forced into the CT dolostone. The Imini fault zone could also act as a preferential 414 drain for mineralization as shown by (i) an eastern zone enriched in pyrolusite (Dekoninck et al. 2016b) and (ii) 415 host of large quantities of barite (Fig. 1b), a mineral also locally observed in the Imini ore (Fig. 4; Dekoninck et 416 al. 2016a) when Ba is also a major element in hollandite.

417 Other Mn deposits hosted in CT carbonate karsts in Tunisia (Jebel Aziza, Gattar Member) display similarities 418 with the Imini-Tasdremt Mn ore belt, as they are located at the transition zone between the Tunisian Atlas and the 419 Saharan platform (Garnit et al. 2020). Mn occurrences in this area have an orientation following a NW-SE 420 direction, also suggesting a structural control of the Mn mineralization (Amouri et al. 1991). These occurrences 421 (and maybe others) indicate that Mn mineralization hosted in dolostones could have a larger extension in Northern

422 Africa.

423 5. Conclusion

424 This innovative study combines ⁴⁰Ar/³⁹Ar ages of K-bearing Mn oxides with Pb isotope data from Mn ore samples 425 and of potential regional rock sources to better understand the karst-hosted Mn deposits of the Imini district. Lead 426 isotopic data show that the most likely source of the Mn-Ba-K-Pb metal association in Imini were the Triassic series composed of a thick basaltic pile interlayered in red sandstone. The ⁴⁰Ar/³⁹Ar ages indicate that primary Mn 427 428 formation took place in the late Cretaceous during several pulses (~91.5 Ma, ~77.5-82 Ma and ~65-67 Ma) and 429 probably persisted until the early Paleogene (~58 Ma) along a distance of ~100 km from Imini to Tasdremt due 430 to uplift/tectonic pulses related to the earliest Atlasic deformation. Manganese enrichment during a second and 431 third events at ca. 36.3 Ma (Priabonian), and ca. 19–20 and ca. 13 Ma (Early-Middle Miocene), respectively, are 432 recorded by pyrolusite-rich mineralization in the Imini Mn district. These data establish that the Mn mineralization 433 formed in response to regional tectonic events - similar to many other sediment-hosted deposits such as 434 Mississippi Valley type and supergene deposits. This correlation highlights the topographic control through 435 tectonic or uplift, associated with humid climate. Ultimately, uplift events controlled (1) the fluid circulation 436 pattern, (2) leaching of the source rocks, (3) precipitation/remobilization of the largest Mn deposit of North Africa. 437 This long lasting (~90 My) multistage evolution formed high-grade Mn deposits. The discrete N70° tectonic 438 structures and the pinch out of Triassic series in front of the Imini district led to the final trap. This study shows 439 that ⁴⁰Ar/³⁹Ar dating coupled with Pb isotope data are key methods to build up robust metallogenic models of 440 karst-hosted mineralization, which can apply in the exploration of similar high-grade Mn deposits.

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455 Compliance with Ethical Standards

We declare that this work has been directed without competing interests and is the authors own intellectualproperty.

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700 Figure caption

- Fig. 1 Geological map of the Moroccan High Atlas and its southern foreland (modified after Leprêtre et al.,
- 2018), with location of samples and the Imini and Tasdremt Mn districts (see Fig. 2a for sampling in the Imini
- district). NAF: North Atlas Front; MF: Medinet Fault; TTFZ: Tizin'Test Fault Zone; SAF: South Atlas Front;
- 704 OkT: Oukaimeden Thrust; SFT: Sidi Fars Thrust; AAMF: Anti-Atlas Major Fault; Azg.: Azegour.
- Fig. 2 a Geological map of the Imini foreland showing the extension of the Mn deposits and the location of the
 sampling sites (modified after Leprêtre et al. (2015). b Stratigraphic section of the Cenomanian-Turonian (CT)
 dolostone at Lantenois showing the three Mn orebodies (modified after Gutzmer et al., 2006). c Schematic crosssection of the South Atlasic Front and the Imini foreland showing the general pinch out of Jurassic and Triassic
 rocks and open folds of the Cenozoic cover (modified after Missenard et al. (2007), Zouhri et al. (2008), Leprêtre
 et al. (2015).
- Fig. 3 a Vertical C2 and C3 ore at Timkit near the Imini fold. b C1 Mn ore with stockwork mineralization in the
 Bou Aguioun pit. c Offset of the CT level at Lantenois. A decametric flexure corresponds to the southern limit of
 the mineralization. Barren CT dolostone is observed south when it is mineralized north. d C2 and C3 Mn ore at
 Lantenois. e C3 Mn ore in the Far West area.
- Fig. 4 Paragenetic sequence of the mineralization in the Imini district (modified from Dekoninck et al. 2016a;
 Verhaert et al. 2022). Ho=hollandite, Cry=cryptomelane, Co=coronadite.
- Fig. 5 Petrographic features of Mn oxide samples. a Colloform cryptomelane (stage 2) of C3 ore growing on the
 dolomite (stage 1) at Lantenois. b Colloform cryptomelane (stage 2) of C1 ore growing on dolomite (stage 1) at
 Bou Aguioun. Late calcite veins (Ca) contains reworked dolomite (Do), quartz, feldspar and cryptomelane grains.
 c Brecciated hollandite stage 2 growing on colloform goethite fragment in C3 ore. d Pyrolusite laths (stage 2)
 growing on stage 1 hollandite with dolomite rhomb remnants at Bou Aguioun (C2 ore).
- **Fig. 6** ⁴⁰Ar/³⁹Ar data of the Priabonian Mn phase. **a.** Density probability diagram of resized (De Putter and Ruffet 2020) apparent ages. **b-e** Age and ³⁷Ar_{Ca}/³⁹Ar_K spectra (³⁷Ar_{Ca} # Ca and ³⁹Ar_K # K with ³⁷Ar_{Ca}/³⁹Ar_K = CaO/K₂O/2.179; Deckart et al., 1997). PA and PPA ages assumed to be valid are shown in bold. Unvalidated ages are shown in grey. Apparent age error bars are at the 1 σ level; errors in the J-parameter are not included. PA and PPA ages (1 σ uncertainties including errors in the J-parameter) are given where applicable. Inverse isochron diagrams (³⁶Ar_{atm}/⁴⁰Ar_{atm} vs. ³⁹Ar_K/⁴⁰Ar*) with least squares fitting of the best straight line according to York

(1968) and York et al. (2004). Grey ellipses are excluded from isochron regression. Unvalidated data are shown
in as grey stars. The mean age of the event is the weighted mean of the validated plateau (PA) and pseudo-plateau
(PPA) ages. Detailed data is in ESM S3.

731 Fig. 7⁴⁰Ar/³⁹Ar data of the Miocene Mn phase. a. Density probability diagram of resized (De Putter and Ruffet 2020) apparent ages. **b-h.** Age and ³⁷Ar_{Ca}/³⁹Ar_K spectra. Degassing diagram of samples IM98-EX and 18TIF02 732 presenting $({}^{39}\text{Ar}_{\text{K}}/\Delta V_{\text{T}^{\circ}}) / ({}^{39}\text{Ar}_{\text{K}}/\Delta V_{\text{T}^{\circ}})_{\text{Max}}$ vs. $\%{}^{39}\text{Ar}_{\text{K}}$ (see text for explanation). $\Delta V_{\text{T}^{\circ}}$ corresponds to the laser 733 734 power control voltage increment. Weighted age spectrum of sample 18TIF02. Refer to Fig. 6 for explanations. Fig. 8⁴⁰Ar/³⁹Ar data of the Cretaceous to late Paleocene Mn phase. a. Density probability diagram of resized (De 735 Putter and Ruffet 2020) apparent ages. **b-h.** Age and ${}^{37}Ar_{Ca}/{}^{39}Ar_{K}$ spectra. Refer to Figs. 6 and 7 for explanations. 736 737 Fig. 9 Pb isotopic ratios of the Imini and Tasdremt Mn ore samples and potential source rocks. a-b Recalculated 738 (²⁰⁶Pb/²⁰⁴Pb)_{in} vs. (²⁰⁷Pb/²⁰⁴Pb)_{in} at 67 Ma. Lead isotopic data for the Jurassic carbonates and sandstones are from

Bouabdellah et al. (2015). Neoproterozoic Pb isotopic values of galena are from Bouabdellah et al. (2016), and
those of MVT deposits are from Bouabdellah et al. (2012, 2015). Galena Pb isotopic values are not recalculated.
The 67 Ma recalculated values from Triassic basalts are complemented by data of Marzoli et al. (2019). All the
Pb isotopic data were obtained on unleached whole rocks, except for the Triassic basalts. c Pb isotope ratios of
unleached and leached basalts.

Fig. 10 Time-event chart of the Imini Mn ore deposits illustrating the multistage Mn deposition based on ⁴⁰Ar/³⁹Ar geochronological data and of Dekoninck et al. (2021) (Tasdremt area). Sedimentation pulses in the CT and late Santonian stages at Imini are from Rhalmi et al. (2000, 1997) and Algouti et al. (1999). The Paleocene to late Eocene and late Miocene pulses of the western Ouarzazate basin are from El Harfi et al. (2001) and Leprêtre et al. (2015). Early Atlasic doming (D0) and Cenozoic Atlasic uplift (D1-3) are from Leprêtre et al. (2018) and Leprêtre et al. (2015), respectively. Fe mineralization events are from Verhaert et al. (2022).

Fig. 11 Metallogenic sketch illustrating the multistage formation of the Imini Mn oxide deposits and their
relationships to uplift events in the Atlas belt. Geodynamic phases are from Leprêtre et al. (2015, 2018). Mn
transport and deposition setting are from Gutzmer et al. (2006).

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754 Table caption

- 755 Table 1 Summary of significant geochronological data and validated ages (Plateau (PA) and Pseudo-Plateau
- (PPA) ages and Isochron ages based on ${}^{40}\text{Ar}/{}^{39}\text{Ar}$ and ${}^{37}\text{Ar}_{\text{Ca}}/{}^{39}\text{Ar}_{\text{K}}$ spectra and Inverse isochron (correlation)
- 757 diagrams (${}^{36}Ar_{atm}/{}^{40}Ar_{atm}$ vs. ${}^{39}Ar_{K}/{}^{40}Ar^{*}$) of coronadite group minerals in the Imini district. Plateau ages are
- shown in bold type. $({}^{36}\text{Ar}/{}^{40}\text{Ar})_{I}$ is the y-axis intercept of the best straight line using the least squares method
- developed by York (1968) and York et al. (2004). Refer to the text for discussion.

760 Appendix A. Supplementary Material

- **ESM S1.** Details of the 40 Ar/ 39 Ar and Pb isotope analytical procedures.
- 762 ESM S2. Lead, uranium, and thorium abundances and lead isotope compositions of the Tasdremt-Imini Mn ores
- and of potential metal source rocks of from Neoproterozoic to late Cretaceous. AA=Anti-Atlas; S=Siroua; S-
- 764 MHA= Southern Marrakech High Atlas; M-MHA= Middle Marrakech High Atlas; MA= Middle Atlas. Initial
- ages of Neoproterozoic volcanic rocks are from Gasquet et al. (2005), Neoproterozoic Mn ores are from Lippolt
- and Hautmann (1995), Cambrian basalts are from Landing et al. (1998), Ordovician shales are from Cornée and
- 767 Destombes (1991), Triassic basalts are from Marzoli et al. (2019), Triassic red sandstones are from El Arabi et al.
- 768 (2006), CT dolostones are from Rhalmi et al. (2000, 1997), Senonian sediments are from Algouti et al. (1999),
- 769 Tasdremt Mn ores are from Dekoninck et al. (2021).
- **ESM S3**. Analytical data and parameters used for calculations of ⁴⁰Ar/³⁹Ar results. Pure K, Ca, and Cl salts; mass
- discrimination; atmospheric argon ratios; J-parameter; and decay constants.
- **ESM S4**. Major and trace element geochemistry.
- 773 ESM S5. K-Ar ages of coronadite group minerals in the Imini deposits. No ages are relevant for this study given
- the complexity of the Ar/Ar spectra.
- **ESM S6**. ²⁰⁸Pb/²⁰⁴Pb vs ²⁰⁷Pb/²⁰⁴Pb and ²⁰⁸Pb/²⁰⁴Pb vs ²⁰⁶Pb/²⁰⁴Pb plots of the Imini and Tasdremt Mn ores and
 potential source rocks.
- **ESM S7.** Altered basalts in the Atlas range. **a** Microscopic view of blue chayesite $[K(Mg,Fe^{2+})_4Fe^{3+}Si_{12}O_{30}]$ under
- polarized light in Triassic basalt at Telouet (Figs. 1b and 2a). b Microscopic view of silicified Triassic basalt with
- chlorite-quartz druses under polarized light at Toufliht (Fig. 1b).

Label	Location	Ore body	PA (*) or PPA (Ma)	±1σ	lsochron age (Ma)	±1σ	(⁴⁰ Ar/ ³⁶ Ar) _i	±1σ
IM98-C	Bou Aguioun	C ₁	19.7*	0.1	19.6	0.1	313.1	7.6
IM98-EX	Bou Aguioun	C ₁	-	-	-	1	-	•
ECH102b	Bou Aguioun	C ₁	19.8*	0.1	19.8	0.1	339.9	47.2
ECH104	Bou Aguioun	C ₁	19.8*	0.1	19.5	0.3	473.9	177.7
IM36	Timkit	C ₁ -C ₂	66.6	0.2	67.0	1.0	158.8	308.4
IM40	Timkit	C_1 - C_2	-	-	-	-	-	-
IM37	Timkit	C ₁ -C ₂	-	-	-	ŀ	-	•
IM39	Timkit	C ₁ -C ₂	-	-	-	-	-	-
18BA01	Bou Aguioun	C ₂	-	-	-	-	-	-
IM41d	Far West	C ₃	61.8	0.3	61.4	1.4	316.5	60.6
LAN57	Lantenois	C ₃	61.6	0.3	61.0	0.8	303.1	6.2
LAN18	Lantenois	C ₃	-	-	-	-	-	-
LAN59	Lantenois	C ₃	-	-	-	-	-	-
18PL12b	Plateau	C ₃	36.8	0.6	36.5	1.3	300.5	6.3
18TIF02	Tiffersine	C ₃	13.0	0.3	12.9	0.3	299.7	2.2
18TIF02	Tiffersine	C ₃	-	-	-	-	-	-
18TIF13	Tiffersine	C ₃	13.1	0.3	13.0	1.2	297.7	2.7
18TIF03	Tiffersine	C ₃	35.6	1.8	36.3	3.0	298.0	1.9
18TIF03	Tiffersine	C ₃	-	-	-	-	-	-
18TIF05	Tiffersine	C ₃	36.3*	1.1	36.0	1.7	298.8	1.3
18TIF12	Tiffersine	C ₃	35.5	0.9	36.0	1.9	297.9	2.2
ECH101b 4 exp.	Bou Aguioun	Stockwork	20.1*	0.1	19.9	0.1	343.7	7.6
ECH103	Bou Aguioun	Stockwork	19.1*	0.1	19.2	0.1	291.4	15.8

Intraplate orogenesis as a driver of multistage karst-hosted mineralization: the Imini manganese case Augustin DEKONINCK^{(1,2)@}, Jocelyn BARBARAND⁽³⁾, Gilles RUFFET^(4,5), Yves MISSENARD⁽³⁾, Nadine MATT

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Oldest radi	ogenic component	Youngest radiogenic component	Comments
			Plateau
			Hump
			Plateau
			Plateau
Maastrichtiar	n #Phase 3 Tasdremt	Paleocene & Priabonian/Miocene	Staircase
Maastrichtiar	n #Phase 3 Tasdremt	Paleocene	Hump
nc	ot identified	not identified	Saddle
nc	ot identified	not identified	Saddle
Campanian	#Phase 2 Tasdremt	Paleocene	Staircase
Campanian	#Phase 2 Tasdremt	Paleocene	Staircase
Campanian	#Phase 2 Tasdremt	Paleocene	Staircase
Campanian	#Phase 2 Tasdremt	Paleocene	Staircase
Campanian	#Phase 2 Tasdremt	Paleocene	Staircase
F	Priabonian	Miocene	Staircase - Ca-rich
F	Priabonian	Miocene	Staircase
			Irregular
F	Priabonian	Miocene	Staircase
2	Paleocene	Priabonian	Staircase - Ca-rich
F	Priabonian	Miocene	Staircase - Ca-rich
			Plateau - Ca-rich
2	Paleocene	Priabonian	Staircase - Ca-rich
Miocene	e (19.3-20.5 Ma)		Plateau
			Plateau

າ YANS⁽¹⁾















Minerals	Sedim-	Diagen	esis	Disso	olution breco	cia and karstifi	cation	Recent surface
	entation	Early	Late I	Dissoluti	ion/ Stage 1	Mineralization Stage 2	Later	processes
Detrital quartz	ø				Epigenesis	Colloform		
Detrital feldspars								
Detrital accessory								
Minerals Aphydrito								
Dolomito microso							1	
	T							
Dolomite rhombs								
Janggunite		<u> </u>			7			
Manganite								
Pyrolusite								
Hollandite				T				
Cryptomelane								
Coronadite								
Lithiophorite					⁷		1	
Romanechite								
Clays			_	Τ	7			
Calcite-Aragonite					7			
Goethite		Y		1			1	
Barite								
Halite								
Time								$\left \right $
						(this study)		١























ESM S1-S6-S7

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