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## Research Paper

## Exploring the metamorphic consequences of secular change in the siliciclastic compositions of continental margins

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## ABSTRACT

Shale and greywacke compositions from the Archean to Phanerozoic record a secular change in the siliciclastic material that comprises much of Earth's continental margins, past and present. This study explores the metamorphic consequence of these compositional changes, by comparing phase equilibrium models constructed for average Archean, Proterozoic, and Phanerozoic shale and greywacke compositions equilibrated along two Barrovian-type geotherms: 1330 °C/GPa (A) and 800 °C/GPa (B). Our models show that Archean siliciclastic rocks can retain up to 4 vol.% water at middle to lower crustal conditions, nearly twice that of Proterozoic and Phanerozoic compositions. The increased ferromagnesium content of Archean siliciclastic rocks stabilizes chlorite to higher temperatures and results in a biotite-rich assemblage at solidus temperatures. Accordingly, water-absent biotite dehydration melting is predicted to play a greater role in the generation of melt in the metamorphism of Archean aged units, and water-absent muscovite dehydration melting is of increasing importance through the Proterozoic and Phanerozoic. This secular variation in predicted mineral assemblages demonstrates the care with which metamorphic facies diagrams should be applied to Archean compositions. Moreover, secular changes in the composition of shale and greywacke is reflected in the evolution of anatectic melt towards an increasingly less viscous, Ca-rich, and Mg-poor monzogranite.

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## 1. Introduction

Evolution of the continental lithosphere composition is echoed in secular compositional changes of supracrustal rocks, which are an archive that can be used to understand and quantify geological processes through Earth's history (e.g. Engel et al., 1974; McLennan, 1982, 2001; McLennan and Taylor, 1991; Condie, 1993; Rudnick, 1995; Veizer and Mackenzie, 2003). The 3.0–2.5 Ga transition between the Neoproterozoic and Proterozoic marks an important period during which sediments underwent significant compositional changes. Archean sediments are generally more ferromagnesium-rich (i.e. total Mg + Fe) compared with Proterozoic and Phanerozoic sediments (Condie, 1993). These changes are generally interpreted to be the consequences of the progressive onset of modern-like plate tectonics (e.g. Dhuime et al., 2012; Dyck et al., 2015; Nicoli

et al., 2016; Palin and White, 2016); continental growth and increase in the amount of subaerial land mass (Flament et al., 2013; Dhuime et al., 2017); and changes in atmospheric conditions (i.e. chemical weathering, surface oxidation) (Condie, 1993; Johnsson, 1993; Barley et al., 2005; Campbell and Allen, 2008). The presence of shale (or pelite) and greywacke in the sedimentary record can be tracked back to ca. 3.5 Ga (Veizer and Mackenzie, 2003). When metamorphosed, these two lithologies commonly comprise mineral assemblages dominated by water-rich minerals such as biotite, muscovite, staurolite and amphibole. Accordingly, they represent important crustal water reservoirs which, when buried within collisional orogenic settings, may exert an important control on melt fertility, and mass transfer (e.g. Clemens and Vielzeuf, 1987; Le Breton and Thompson, 1988; Douce and Johnston, 1991; Stevens et al., 1997; Sawyer et al., 2011; Nicoli et al., 2017). Variation in the amount of mineral-bound water may have profound impact on the rheology of the crust (e.g. Kolb, 2008), metamorphic fluid production (Walther and Orville, 1982; Ague, 2011), as well as the thermal state of the crust (e.g. Stüwe, 1995; Depine et al., 2008).

In this study, we focus on the equilibrium assemblages of buried and metamorphosed shale and greywacke in an orogenic setting.

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**Table 1**  
Average shale and greywacke composition (in wt.%) from the Archean, Phanerozoic, and Proterozoic after [Condie \(1993\)](#).

Lithology	Shale			Greywacke		
	Archean	Prot.	Phan.	Archean	Prot.	Phan.
SiO <sub>2</sub>	60.9	63.1	63.6	66.1	65.4	66.3
TiO <sub>2</sub>	0.6	0.6	0.8	0.6	0.7	0.7
Al <sub>2</sub> O <sub>3</sub>	17.5	17.5	17.8	15.3	15.5	15.5
FeO <sup>T</sup>	7.5	5.7	5.9	5.5	6.1	6.2
MgO	3.9	2.2	2.3	3.5	2.2	2
CaO	0.6	0.7	1.3	2.5	2.5	3.2
Na <sub>2</sub> O	0.7	1.1	1.1	2.9	3	3.1
K <sub>2</sub> O	3.1	3.6	3.8	2	2.4	2.3
P <sub>2</sub> O <sub>5</sub>	0.1	0.12	0.14	0.12	0.15	0.14
Mg#	0.34	0.28	0.28	0.39	0.27	0.24
A/CNK	2.4	1.9	1.8	1.4	1.3	1.2

Mg# = Mg/(Mg + FeO); A/CNK = Al/(Ca + Na + K).

Although this study is based on the secular change in shale and greywacke compositions, the timing of metamorphism of these sedimentary rocks is not strictly relevant and we recognize that Archean compositions may have been metamorphosed and/or reworked through Proterozoic and Phanerozoic orogenesis. However, average Archean compositions were determined using rocks from terrains with little to none Phanerozoic and Proterozoic overprint ([Condie, 1993](#)). We investigated the influence of rock composition on water retention in hydrous minerals below solidus temperatures and the chemistry and degree of melt generated at suprasolidus temperatures in water-absent conditions.

## 2. Starting material

To investigate secular changes in sediment fertility, partial melting and the nature of the generated magma, we used averaged shale and greywacke compositions from the Archean, Proterozoic and Phanerozoic eons ([Table 1](#); [Condie, 1993](#)). The shale compositions represent a global average of shales sampled from cratonic and passive margin successions. The greywacke compositions were assembled from turbidite units within Precambrian greenstone belt and modern volcanic arcs. Archean shales are characterized by a low K content (3.1 wt.%) and a high Mg# (Mg# = Mg/[Mg + FeO<sup>T</sup>]) and A/CNK values (A/CNK = Al/(Ca + Na + K)) of 0.34 and 2.4, respectively ([Condie, 1993](#)). Proterozoic and Phanerozoic shales contain higher concentrations of K, 3.6–3.8 wt.%, and lower Mg# and A/CNK values of 0.28 and 1.8–1.9, respectively. Apart from a decrease in Mg# from 0.39 to 0.24, the major element composition of greywacke does not vary significantly through time ([McLennan, 1982](#); [Condie, 1993](#)). The most significant secular change in greywacke compositions are found in their trace elements compositions ([McLennan and Taylor, 1991](#)), and as such are not likely to exert much control on the primary mineral assemblage.

## 3. Model setup

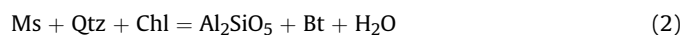
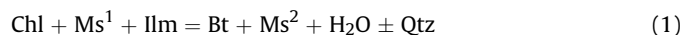
Phase equilibria of greywacke and shale from the Archean, Proterozoic and Phanerozoic were modelled using version 6.7.5 of the *Perple\_X* software package ([Connolly, 2009](#)) in the system MnO–Na<sub>2</sub>O–CaO–K<sub>2</sub>O–FeO–MgO–Al<sub>2</sub>O<sub>3</sub>–SiO<sub>2</sub>–H<sub>2</sub>O–TiO<sub>2</sub>–O<sub>2</sub> (MnNCKFMASHTO). For all calculations, we used the 2004 update of the [Holland and Powell \(1998\)](#) thermodynamic database (<http://www.perplex.ethz.ch/>). This approach assumes equilibrium is maintained between phases across the chosen range of pressures and temperatures ([Connolly and Kerrick, 1987](#); [Connolly, 1990](#)). Solution models used are as follows: silicate melt model after [Green et al. \(2016\)](#); feldspar ([Fuhrman and Lindsley, 1988](#)); chlorite

([Holland et al., 1998](#)); amphibole model after [Diener and Powell \(2012\)](#); white mica ([Coggon and Holland, 2002](#); [Auzanneau et al., 2010](#)); ilmenite ([White et al., 2000](#)); hydrous cordierite ([Holland and Powell, 1998](#)); orthopyroxene ([White et al., 2001](#)); biotite ([White et al., 2007](#)); and garnet ([White et al., 2014](#)). Mineral abbreviations follow [Kretz \(1983\)](#). To investigate the maximum amount of water that shale and greywacke compositions retain under subsolidus pressure and temperature conditions, fluid was considered in excess and treated as a pure H<sub>2</sub>O. Adding CO<sub>2</sub> would lower a<sub>H2O</sub> and reduce the stability of hydrous phases, resulting in a reduction of muscovite, biotite, and chlorite at higher temperatures. As such, maximum water retention values were obtained by summing the water content in the different hydrous phases. For modelling of suprasolidus conditions, bulk-rock fluid contents were individually fixed to be minimally saturated (<0.5 mol.% of free H<sub>2</sub>O) at the point at which the relevant geotherm intersects the solidus. As the focus of this paper is on the metamorphism of siliciclastic metasedimentary compositions in a continental collisional setting, we investigated the modal proportion of hydrous minerals along two Barrovian-type geotherms from 400 to 1000 °C. The two geotherms correspond to the upper and lower bounds of orogenic thermal gradients compiled by [Brown \(2006\)](#) in a global review of peak metamorphic conditions from Archean to Phanerozoic terrains. The lower pressure geotherm (A) was set at 1330 °C/GPa intersecting the solidus at ~675 °C and 0.5 GPa. The higher pressure geotherm (B) was set at 800 °C/GPa, which intersects the solidus at ~675 °C and 0.8 GPa. Along both geotherms, we investigate how secular compositional variations effect the chemistry and modal proportion of silicate melt.

## 4. Results

### 4.1. Subsidius mineral-bound water

The retention of water in metasedimentary rocks that have undergone compaction, diagenesis and low-grade metamorphism is primarily a function of the stability of hydrous minerals ([Walther and Orville, 1982](#)). At lower greenschist facies metamorphic conditions (400 °C) shales and greywackes can respectively retain up to 4.5 wt.% and 3 wt.% mineral-bound water, respectively ([Fig. 1](#)). As temperature increases these minerals devolatilize. The temperature and pressure at which devolatilization occurs is strongly influenced by the equilibria conditions of the following three continuous reactions ([Ramsay, 1973](#); [Ahn and Nakamura, 2000](#)):



Accordingly, the temperature at which geotherms A and B intersect the assemblage field boundaries is markedly different for the different shale and greywacke compositions ([Fig. 1](#)). The higher Fe+Mg content of Archean shale and greywacke results in a greater volume of chlorite, and corresponding mineral-bound water, up until chlorite breakdown at 550–600 °C ([Fig. 1](#)) (for phase proportion along the two geotherms, see [Supplementary Fig. S1](#)). Along the higher pressure geotherm (B), Archean shale retains up to ~4 wt.% water to lower-crustal pressures of 0.75 GPa, twice that of Phanerozoic shale under the same conditions. In Archean shale, the maximum bulk-water content along the higher pressure geotherm (B) drops from 3.8 to 1.5 wt.% over a temperature window of 590–600 °C. By contrast, Proterozoic and Phanerozoic shale compositions undergo two major devolatilization events along

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