



Letter to the Editor

Fluids, Melting, Granulites and Granites: A Controversy – Reply to the Commentary of J.D. Clemens, I.S. Buick and G. Stevens


The commentary of Clemens, Buick and Stevens on our papers [Aranovich et al. \(2014\)](#) and [Newton et al. \(2014\)](#) contains many interesting arguments and is a useful contribution in helping to formulate the framework of what we believe to be an important on-going controversy. In question is whether volatile components, especially H₂O, have been commonly infiltrated into sites of deep-seated metamorphism and granitic magma genesis, or whether granitic magmas, and their supposed residues, the granulites, more commonly resulted from fluid-absent (closed system) melting processes. This is an old debate in petrology, coined by N.L. Bowen nearly a century ago as the “Pontiffs versus the Soaks”, i.e. those who adhere religiously to established dogma (in Bowen’s time restricted to simple low-pressure experimental melting diagrams) versus those who indiscriminately invoke the action of exotic fluids, or “ichors”, for any and all petrogenetic purposes, free from the embarrassing restrictions of any hard data of physical chemistry.

The authors of “Commentary” are quite frank in espousing their fluid-absent point of view. In their “Preamble” they state: “The authors (of our two papers) attempt to show that the view of these processes as dominantly fluid-absent should be re-appraised. We disagree—”. Throughout their paper [Clemens et al. \(2016\)](#) reiterate their belief that the bulk of analytical and experimental evidence favors the fluid-absent hypothesis of deep-crustal melting and that granulite facies metamorphism takes place, for the most part, in the absence of active fluids.

We do not agree that our point of view is a re-appraisal of a near-consensus opinion. Over the past two decades many workers have judged that field evidence in various terranes where partial melting is inferred to have taken place, including granulite facies terranes, favors the view that infiltration of exotic volatiles, including H₂O, was active (summarized by [Aranovich et al., 2013](#)). Moreover, many workers have amassed evidence that fluids of low H₂O activity were influential in high grade metamorphism of the deep crust (summarized by [Touret and Nijland, 2013](#)). In succeeding paragraphs we discuss issues raised by [Clemens et al. \(2016\)](#) concerning (1) the temperature ranges, H₂O contents, H₂O activities and possible melt yields in deep-crust source regions of granitic magmas, (2) characteristic H₂O activity in granulite facies metamorphism and partial melting, (3) the reputed possibility of efficient melt extraction, (4) geochemical differentiation of the crust and (5) the significance of fluid inclusions in granulites. Since the separate critiques of our two papers by [Clemens et al. \(2016\)](#) have considerable overlap, we deal with their issues jointly.

The recent paper of [Collins et al. \(2016\)](#) showed, using large data sets of Ti-in-zircon thermometry of granites and experimental data of zircon saturation temperatures in granitic magmas, that the

temperature scale of magma genesis of subduction zone, or I-type, granites should be shifted downward from previous estimates, with necessarily greater H₂O contents of the granitic liquids. Sufficient experimental data are available to address this problem; that is, to estimate the temperatures, H₂O contents and H₂O activities of granitic melts which could coexist with intermediate gneisses at deep-crustal conditions, either by fluid-absent dehydration melting or with additional input of externally-derived H₂O. [Fig. 1](#) shows the H₂O activity at liquidus temperatures of an alkali feldspar-quartz melt from dry melt to saturation with an aqueous fluid at 1.0 GPa, according to [Aranovich et al. \(2013\)](#) and [Clemens and Vielzeuf \(1987\)](#). Shown also are the H₂O activity values defined by the model granulite facies assemblage phlogopite-quartz-K-feldspar-enstatite, in equilibrium according to the reaction:



These model granulite facies H₂O activity values are reversed data ([Aranovich and Newton, 1998](#)) and should be considered more definitive than the thermodynamic extrapolations from much lower-pressure experimental data of previous studies. They require revision of phlogopite stability, despite the protests of [Clemens et al. \(2016\)](#). Detailed defense of this assertion is given in [Aranovich and Newton \(1998, p. 195\)](#) and [Berman et al. \(2007, p. 146\)](#). The two H₂O activity versus temperature trends intersect at 830 °C. This is the temperature at which a simple granitic liquid could coexist with a model biotite-orthopyroxene (granulite facies) intermediate gneiss. The corresponding H₂O activity is 0.36, corresponding to a H₂O content of the magma of 5.2 wt.%, according to the reversed haplogranite liquidus composition measurements of [Makhlof et al. \(2014\)](#), shown in [Fig. 2](#).

Shown also is the temperature range (860–890 °C) over which orthopyroxene and biotite coexist in experimental dehydration melting at 1.0 GPa of a natural tonalite of H₂O content 0.8 wt.% ([Rutter and Wyllie, 1988](#)). The corresponding H₂O activities range from 0.22 to 0.32 ([Fig. 1](#)) and the H₂O contents range from 4 to 4.8 wt.% ([Fig. 2](#)). The difference between the H₂O activity indications in natural and synthetic systems presumably results from neglect of the Fe in natural biotite and orthopyroxene, the Al and Ti in natural granulite facies biotite, and the Na content of alkali feldspar. It is probable that synthetic phlogopite is somewhat less thermally stable than the natural biotite.

The foregoing temperatures, H₂O contents and H₂O activities seem quite plausible for generation of granitic liquids in the deep crust by dehydration melting. Experimental studies on homogenization temperatures of melt inclusions of the Transbaikalian (Siberia) anorogenic granites give liquidus temperatures around 875 °C ([Litvinovskii et al., 1994](#)). This temperature range is consistent with the estimates of [Collins et al. \(2016\)](#) for within-plate

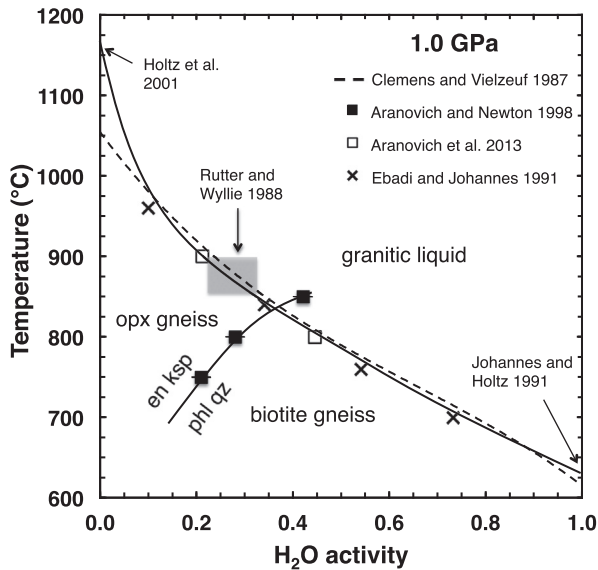


Fig. 1. Liquidus (melt–quartz–alkali feldspar) equilibrium H_2O activity at 1.0 GPa. *Open squares:* H_2O activity determined by equilibration of simple granitic melt with (K, Na) Cl solutions of known H_2O activity (Aranovich et al., 2013; Aranovich and Newton, 1997). *X-symbols:* Liquidus H_2O concentrations of simple granitic melts measured by Ebadi and Johannes (1991) in CO_2 – H_2O fluids, converted to activity–temperature points by activity–concentration data of Aranovich and Newton (1999). *Dashed Line:* Liquidus H_2O activity–concentration function of a simple granitic liquid shown by Clemens and Vielzeuf (1987), attributed to calculations by H. Nekvasil. The agreement of the latter estimate with the other data is remarkably good, except that we adhere to the revised temperature scale of melting of a dry simple granite of Holtz et al. (2001). Shown also are reversed data for the model granulite-forming dehydration reaction of phlogopite + quartz to enstatite + K-feldspar, Expression A, text. The intersection of this trend with the simple granitic liquid H_2O activity–temperature trend gives the temperature at 1.0 GPa (830 °C) at which a simple granitic melt could form by dehydration melting of phlogopite + quartz, either fluid-absent or by input of additional H_2O of external origin. The shaded box is the temperature interval over which biotite and orthopyroxene coexist in the presence of a granitic partial melt of a natural tonalite found by Rutter and Wyllie (1988). The apices of the box give the associated H_2O activity range. The higher temperatures and lower activities indicated for the natural system probably results from greater stability of the Al, Ti biotite than phlogopite.

(anorogenic) granites and with the H_2O contents of melt inclusions in quartz of granites compiled by Scaillet et al. (1996). Most of these values are greater than 4 wt.% H_2O and range as high as 7.5 wt.%. The overall negative correlation of the H_2O contents of Scaillet et al.'s (1996) melt inclusions with apparent entrapment temperatures was considered by Clemens and Watkins (2001) to be strong evidence for closed-system melting, but may have other plausible interpretations (Weinberg and Hasalová, 2015a,b).

The main difficulty with the dehydration melting model is the small yields of granitic liquid which could be obtained. Several authors, including ourselves, have expressed doubt that the amount of H_2O resident in biotite and amphibole of intermediate gneisses would be sufficient to produce large amounts of granitic magmas by fluid-absent melting (Rutter and Wyllie, 1988; Johannes and Holtz, 1991; Weinberg and Hasalová, 2015a) unless melting temperatures were generally greater than about 950 °C. The paper by Collins et al. (2016) indicates that this condition was not met for the cordilleran batholiths, and rarely for the within-plate alkaline granites. Moreover, most workers would assume that the lower continental crust at the time of generation of the Mesozoic and Tertiary batholiths was already in the granulite facies, so that only about 2/3 of the potential hydrous minerals (the mafic components) were actually in the form of biotite and amphibole, the remainder being pyroxenes and garnet. If so, the maximum yield of granitic liquid having 5 wt.% H_2O would be only

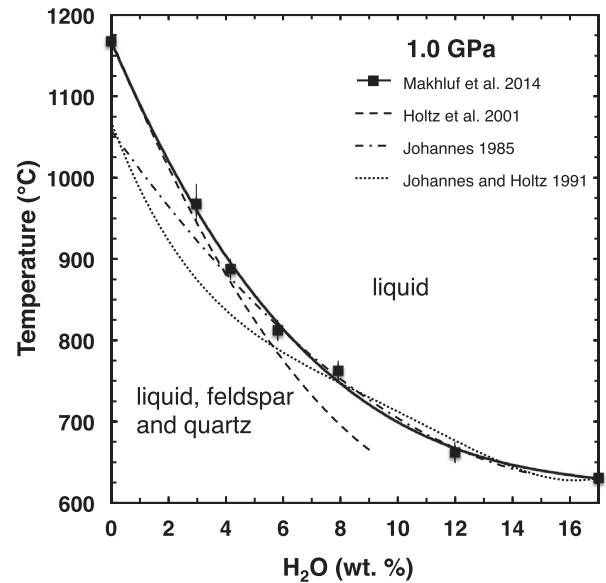


Fig. 2. Reversed liquidus H_2O contents of simple granitic liquids at 1.0 GPa (Makhluif et al. 2014) compared with extrapolation estimates from lower pressures. Our H_2O contents at a given temperature are generally higher than given by other authors, which fact indicates that the yield of granitic liquids by partial melting of an intermediate biotite–amphibole gneiss would be lower than possible with the other liquidus H_2O values shown. (See above-mentioned references for further information.)

around 10% of the country rock by dehydration melting, even assuming complete dehydration and perfectly efficient extraction, both of which seem dubious. If accumulation of granitic liquid is dominantly vertical, it would seem impossible to explain the great thicknesses of some batholiths such as the Transbaikalian granites, which may be generally 15 km thick, based on seismic soundings (Litvinovskii et al., 1994). If, on the other hand, there is the possibility of influx of externally derived H_2O , there is no theoretical limit on the amount of granitic liquid which could be generated, short of melting of the entire crust. According to Figs. 1 and 2, the lower crust could remain in the granulite facies, with the generation of liquids of 4–5 wt.% H_2O , in the temperature range 850–900 °C. If there were high rates of H_2O influx, the lower crust might actually rehydrate, with the possibility of generation of liquids at lower temperatures and higher H_2O contents, as suggested for the origin of the cordilleran batholiths by Collins et al. (2016).

Most previous workers have made use of the equilibrium of biotite–quartz–K-feldspar–orthopyroxene to define H_2O activity at the beginning of granulite facies metamorphism, since the appearance of orthopyroxene in felsic gneisses is a definitive isograd of progressive metamorphism. The lower P – T limits of the granulite facies are thought to be around 0.5 GPa and 750 °C. Newton et al. (2014) showed that earlier estimates of H_2O activity of 0.15–0.30 under these conditions should be revised upward to as high as 0.5, in agreement with the thermodynamic analysis of Perchuk et al. (2000) on orthopyroxene-bearing migmatites in Sri Lanka. If SiO_2 activity is lowered, as in anatectic melt, inferred by Perchuk et al. (2000) for the Udadigana, Sri Lanka incipient charnockite occurrence, $a(H_2O)$ could be as high as 0.6.

Clemens et al. (2016) indicate that this revision of the H_2O activity scale is not novel, but makes use of outmoded data. They cite experiments on the H_2O contents of synthetic cordierite grown at granulite facies conditions ($T > 700$ °C; P 0.4–0.6 GPa: Harley et al., 2002) to indicate that granulite facies $a(H_2O)$ may range anywhere between 0.18 and 0.63. This conclusion seems quite at odds with the conclusion of Clemens and Watkins (2001) that the H_2O content of granitic melts produced by fluid-absent melting in the

granulite facies over a small pressure range is narrowly constrained. We do not believe that the cordierite-H₂O barometer scale is calibrated well enough to be a definitive indicator for granulites. The large range of inferred H₂O activities in cordierite granulites found by [Harley et al. \(2002\)](#) would seem to point in that direction.

The accusation of [Clemens et al. \(2016\)](#) that [Newton et al.'s \(2014\)](#) revised H₂O activity scale controverts the experimental work of [Vielzeuf and Clemens \(1992\)](#) on the melting of phlogopite plus quartz is unjustified. [Newton et al. \(2014\)](#) showed that their revised H₂O activity scale of the assemblage phlogopite–quartz–enstatite–K-feldspar actually satisfies the [Vielzeuf and Clemens \(1992\)](#) experimental points better than does the fit to their experiments preferred by [Vielzeuf and Clemens \(1992\)](#).

Another key aspect of the fluid or no-fluid controversy is whether small amounts of granitic melt, limited to less than 20% for ordinary biotite-amphibole gneiss source rocks, can be efficiently separated from the host rock and collected at upper crustal levels to form granite batholiths. The paper of [Sawyer \(1994\)](#) was gratefully hailed as a solution to the extraction problems posed by the high viscosity and low relative buoyancy of partial melts of quartzofeldspathic rocks of low H₂O content ([Wickham, 1987](#)). Qualitative arguments were given by [Sawyer \(1994\)](#) to support the concept that shear deformation during partial melting could aid liberation of small melt fractions, even less than 5%. [Sawyer \(1999\)](#) bolstered his case for separation of low melt fractions citing the common occurrence of interconnected microscopic grain boundary veinlets of feldspars and quartz in granulites, which veinlets he interpreted as frozen grain boundary melts. Alternative interpretation is, however, possible, as discussed below.

An unresolved problem with small melt extraction is the large amount of quartzofeldspathic leucosome (presumably former melt) shown in some well-documented granulite terranes ([Timmermann et al., 2002](#); [Percival, 1991](#); [Morfin et al., 2013](#)). These rocks are all strongly deformed yet managed to hold onto large amounts of melt until temperatures fell below the freezing point. Another point is the non-granitic mineral compositions of supposed thin melt lamellae in some granulites: quartz + K-feldspar in some ([Hansen et al., 1995](#)), quartz + plagioclase or quartz only in some ([Hartel and Pattison, 1996](#)) and two feldspars + quartz in others ([Harlov et al., 1998](#)). The suggestion by some authors that these mineralogic variations may be explained by crystallization–fractionation processes operating at the grain boundary level seems dubious. More probable is the interpretation that the (nearly ubiquitous) grain boundary quartz–feldspar veins in granulites are deposits of very penetrative saline solutions of low H₂O activity, as first postulated by [Perchuk and Gerya \(1992\)](#). Yet another problem is that some mobilizates in granulites have the same (tonalitic–trondhjemitic) compositions as the host rock ([Newton et al., 1998](#)). It appears that the physics and chemistry of partial melting in the granulite *P–T* regime is still too imperfectly understood to appeal to this mechanism as the principal granite (and granulite) forming process.

Another major issue in the granulite debate is whether melt extraction can deplete the large ion lithophile elements (LILE) like Rb and Th, which elements are characteristically low in some, but not all granulites. [Clemens et al. \(2016\)](#) argue that these elements must be depleted by extraction of a granitic melt, since these elements will be partitioned at least to some extent into departing liquids. This does not really address the concern voiced by [Aranovich et al. \(2014\)](#). They note that biotite in the highest grade, LILE-depleted granulites of the South India Archean granulite facies terrane is itself depleted in Rb relative to lower-facies biotites. Since biotite strongly prefers Rb relative to a granitic melt, [Hansen et al. \(2002\)](#) show that the granulite residuum would not

be markedly depleted in Rb unless very large melt fractions (>40%) were removed. This is very unlikely to have been realized in the South India high grade tonalitic gneisses, whose bulk chemistry is not much different than lower grade gneisses in the same terrane ([Hansen and Harlov, 2007](#)). In view of the low melt solubilities of the U and Th carrier minerals monazite and zircon, [Clemens et al. \(2016\)](#) appeal to selective entrainment of accessory minerals in rising magmas as a means of depleting the radioactive elements.

[Clemens et al. \(2016\)](#) express the opinion that the CO₂-rich fluids characteristically found included in minerals of granulite facies rocks merely represent the residues of ambient pore fluids from which most of the H₂O has been dissolved into departing partial melts. This trivial interpretation of the often high CO₂ contents in granulite minerals, notably garnet (up to 1 wt.%: [Srikantappa et al., 1992](#); [Touret and Hansteen, 1988](#)) is disputed by several studies showing that carbon isotopes in granulite facies minerals have a pronounced mantle-origin signature (fluid inclusions in quartz: [Jackson et al., 1988](#); cordierite: [Armbruster et al., 1982](#); scapolite: [Blattner, 1976](#)). [Dunai and Touret \(1993\)](#) found that He isotopes from synmetamorphic CO₂-rich fluid inclusions in deep-crustal garnet granulites from southern India contain He isotopes that can only be explained if the fluids are mantle-derived. The concentrated brine fluid inclusions found also in some granulites and migmatite leucosomes (e.g. Fig. 1 in [Manning and Aranovich, 2014](#)) are similarly dismissed by [Clemens et al. \(2016\)](#) as only residual pore fluids. This interpretation ignores the many reports of elevated Cl (up to 0.8 wt.%) in melt inclusions in minerals of granites ([Dolejš and Baker, 2004](#) and references therein) and the presence of halogen-rich biotite and amphibole in some granulite facies assemblages. Calculations by [Zhu et al. \(1994\)](#) and [Newton et al. \(1998\)](#) have shown that Cl/(Cl + H₂O) ratios in a magnesian biotite as low as 0.015 imply a Cl-rich fluid in equilibrium with the granulite facies assemblages.

[Clemens et al. \(2016\)](#) state unjustly that the experimental and theoretical physical chemistry of immiscible CO₂ and brines at high temperatures and pressures cited by [Newton et al. \(2014\)](#) is not à propos of the granulite problem. We showed that some rocks from the transitional granulite terrane of South India have fluid inclusions in quartz that have compositions, densities and H₂O activities expected of immiscible fluids at the metamorphic *P–T* conditions, and that the two types of fluids co-occur in the same generations of fluid inclusion trails, showing that they were coevally emplaced. The structural position of these incipient charnockites, on the upper-level fringes of the massive granulites, requires that the fluids must have passed through the radiometrically coeval granulites below them. Of course, the ultimate origin of the fluids, particularly the brine suite, remains uncertain, but this fact does not justify their dismissal as irrelevant. The more common preservation of the CO₂-rich suite is explainable by the equation of state of CO₂: its isochores are more nearly parallel to the return *P–T* paths of the granulites from depth than are the isochores of H₂O-rich fluids ([Touret, 2001](#)).

The subject of metasomatism by fluid action is relegated by [Clemens et al. \(2016\)](#) to a role of “local”, but not regional importance in deep-crustal processes. This point of view is certainly not a currently prevalent one. There are many recent descriptions of regional metasomatic alteration, including in granulite facies terranes (for instance the classical terrane of Russian Lapland: [Korikovsky and Aranovich, 2010, 2015](#)). Summaries of regional high grade metasomatism are given by [Touret and Huizenga \(2011\)](#) and [Touret and Nijland \(2013\)](#).

In view of the difficulties and contradictions encountered by the dehydration (closed system) melting hypothesis for the origin of granite batholiths and granulite facies terranes, it seems preferable to suppose, with [Collins et al. \(2016\)](#), that H₂O, in amounts larger

than could have been present in hydrous minerals of high grade tonalitic gneisses, can be supplied by invading basalts, now thought to contain commonly around 4 wt.% H₂O for the subduction zone basalts (Plank et al., 2013). Clemens et al. (2016) agree that invasion of basaltic magmas into the lower crust is the most probable origin of the high temperatures for partial melting and granulite facies recrystallization. However, Bowen (1928) showed that, to melt significant quantities of country rock, a heat-equivalent amount of basaltic liquid must crystallize to supply the latent heat of melting. The H₂O content of a crystallizing basaltic liquid must re-emerge somewhere and, since the hydrous melting temperatures of intermediate rocks are lower than those of mafic rocks, melting of the lower crust must ensue. This scenario is, in principle, the same as that advocated by Litvinovsky and Podladchikov (1993). These authors argue that there might be attendant metasomatic alteration of crustal rocks, with modification of the partial melts. A high H₂O-activity fluid phase would not be produced, though some H₂O could be partitioned into fluid phases dominated by anhydrous volatile components from the basalt, as discussed by Manning and Aranovich (2014), or from sedimentary packets in the thermally activated zone. Alternatively, the anhydrous volatiles might be derivatives of sea water and surficial sediments which could migrate downward as deep as 15 km into the crust (Connolly and Podladchikov, 2004).

Clemens et al. (2016) seem not to take seriously the current active discussion of the importance of metasomatism in deep-crust/upper mantle processes. We believe, on the contrary, that this new avenue of investigation will prove to be more fruitful than the older simplistic view of closed-system crustal evolution.

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L.Y. Aranovich

Inst. of Geology of Ore Deposits, Petrography, Mineralogy and

Geochemistry, Russian Academy of Sciences, Moscow, Russia

Dept. of Geology, Univ. of Johannesburg, Johannesburg, South Africa

A.R. Makhluף

C.E. Manning

R.C. Newton*

Dept. of Earth, Planetary and Space Sciences, University of California at

Los Angeles, Los Angeles, CA, USA

* Tel.: +1 3102062917.

E-mail address: rcnewton@g.ucla.edu (R.C. Newton)

J.L.R. Touret

Inst. de Mineralogie, de Physique des Matériaux, et de Cosmochimie

(IMPMC), Sorbonne Univ., UPMC Univ. Paris 06, UMR-CNRS 7590,

Museum National D'Histoire Naturelle, IRD UMR 206, 4 Place Jussieu,

F-75005 Paris, France

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