Supplementary Document S1: Analytical methods

1. Whole rock analyses

1.1 Hudson (1975) thesis

Analyses were performed by Neil Hudson at the University of Edinburgh. Major elements were determined from X-ray fluorescence spectroscopy using a Phillips PW 1212 automatic spectrometer. Glass discs for analysis were prepared by fusion of 1 part rock powder, 1 part La_2O_3 and 6 parts $Li_2B_4O_7$. FeO was determined using the method of Wilson (1955), Na_2O was determined by flame photometry, and Loss on Ignition (LOI), assumed to equate to H_2O in these carbonate-free rocks, was determined by fusion. Concerning determination of FeO, dissolution and other difficulties were noted on pages 79 and 80 of Hudson (1975), from which he concluded that the FeO values determined by the method of Wilson (1955) "can be considered at best as only an approximation".

1.2 This study

Samples were analyzed commercially for bulk chemical composition by X-ray fluorescence (XRF) at two commercial labs: Geochemical Laboratories, Earth & Planetary Sciences, McGill University, Montreal, Quebec; and ALS (Vancouver) Geochemistry Lab. Analyses at McGill were were performed on a Philips PW2400 3kW automated XRF spectrometer system, and at ALS on a PANalytical Axios FAST automated XRF spectrometer system. In both labs the analyses were performed using fused beads. All iron was assumed to be in the ferric state.

2. Major mineral analyses

2.1 Hudson (1975) thesis

Analyses were performed by Neil Hudson using (1) a Cambridge Instruments Geoscan electron microprobe in the Department of Mineralogy and Petrology, Cambridge University, and (2) a Cambridge Instruments Microscan electron microprobe in the Grant Institute of Geology, University of Edinburgh, using wavelength-dispersive spectroscopy and the methods of Sweatman & Long (1969). Standards included orthoclase (K), jadeite (Na) and, for all other elements, stoichiometric oxides and pure elements.

2.2 This study (= Goldsmith, 2017 thesis)

Analyses were performed by Shantal Goldsmith using wavelength-dispersive spectroscopy on a JEOL JXA-8200 electron microprobe at the University of Calgary, using standard operating conditions (15 kV; 20 nA; focused beam) and a range of well-characterized natural and synthetic standards (see Nicholls & Stout, 1988). Results were subjected to matrix corrections based on the ZAF method (Reed, 1996).

3. Monazite analyses

3.1 Microprobe analyses

Partial monazite analyses from nine samples (see Figure S12 and Tables S12 and 13) were obtained by Shantal Goldsmith (Goldsmith, 2017) using wavelength-dispersive spectroscopy on a JEOL JXA-8200 electron microprobe at the University of Calgary. Monazite was identified by mapping full thin sections for Ce, and P using electron dispersive spectrometry (EDS) on the microprobe. Measurements of P, Ce, La and Y were obtained using an accelerating voltage of 15 kV, a Faraday cup current of 50 nA and an electron beam defocused to a 5 μm diameter. A set of natural and synthetic standards were used for calibration, and ZAF matrix corrections (Reed, 1996) were applied. Measurement conditions and analytical settings for U, Th and Pb are given in table 2 of Yang & Pattison (2006). The monazite analyses in Table S12 are partial analyses; the rest of the undetermined elements were treated as Nd (Yang & Pattison, 2006).

3.2 U-Pb LA-MC-ICPMS analyses

Backscattered electron (BSE) imagery was used to characterize the textural setting of monazite grains (see Supplementary Figure S11). BSE imaging of monazite in the nine samples in Table S13 did not show any evidence of internal zoning, and the grains were so small (most <20 μm) that only a single measurement could be made on two thirds of the grains identified.

U-Pb monazite data was collected by Shantal Goldsmith and Andy Dufrane (U. Alberta) using laser ablation multi collector inductively coupled mass spectrometry (LA-MC-ICPMS) at the Canadian Centre for Isotopic Microanalysis at the University of Alberta, Edmonton, Canada using procedures modified from Simonetti et al. (2005). The analytical setup consists of a New Wave UP-213 Nd:YAG laser ablation system interfaced with a Nu plasma MC-ICPMS equipped with three ion counters and 12 Faraday cups allowing static collection of both U and Pb isotopes. The laser was operated at 4 Hz with a beam diameter of 8-15 um, depending on the grain size of the monazite (see above and Fig. S12), which yielded a fluence of ~2 J/cm2. Ablations were conducted in a He atmosphere at a flow rate of 1 L/min through the ablation cell. Output from the cell was joined to the output from a standard Nu plasma desolvating nebulizer (DSN-100). On peak gas + acid blanks (30s) were measured prior to a set of analyses. Data was collected statically consisting of 30 1s integrations. Before and after each set of analyses, in house monazite reference Madagascar (Heaman, unpublished data, Simonetti et al., 2006) and 44069 (Aleinikoff et al, 2006) were repeatedly analyzed, to monitor U-Pb fractionation, reproducibility, instrument drift, and to assess data quality. Standard 44069 when analysed as an unknown gave an age of 424.5 ±2.0 Ma, in excellent agreement with the published TIMS age of 424.9 ±0.4 Ma. Mass bias for Pb isotopes was corrected by measuring ^{205}Tl/^{203}Tl from an aspirated Tl solution (NIST SRM 997) via the DSN-100 desolvating nebulizer using an exponential mass fractionation law and natural ^{205}Tl/^{203}Tl of 2.3871. All data were reduced offline using an Excel-based program. Unknowns were normalized to Madgascar monazite as the primary reference and 44069 was treated as an unknown to assess data quality. The uncertainties reported are a quadratic combination of the internal measurement precision and the overall reproducibility of the standards during an analytical session. The long term 2-σ reproducibility for the standards is estimated to be ~1% for ^{207}Pb/^{206}Pb and 2% for ^{206}Pb/^{238}U. The data are not corrected for common Pb due to the difficulty in resolving transient contributions of ^{204}Hg present in the Ar gas from ^{204}Pb present in either the crystal and/or the acid + gas blank. Thus, reported ^{204}Pb values are for informational purposes only, but can be useful for identifying and rejecting samples that have obvious amounts of common Pb.

After data reduction, concordia intercept ages in Tera–Wasserburg U–Pb concordia diagrams (Tera and Wasserburg 1972) were calculated using the ISOPLOT version 4.11 Excel add-in of Ludwig (2003), by anchoring to a common Pb value of 0.86 ± 0.06 estimated from the Pb evolution model of Stacey and Kramers (1975).

4. References for Supplementary Document S1

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Supplementary Document S2: Thermodynamic modelling

Chemical systems, thermodynamic databases and activity-composition (a-X) models

The phase diagrams in Figs. 17 and S3 were calculated in the chemical systems MnNCKFMASHTO (MnO-Na_2O-CaO-K_2O-FeO-Fe_2O_3-MgO-Al_2O_3-SiO_2-H_2O-TiO_2), using the measured molar Fe^{3+}/Fe_{total} of the rocks, and in MnNCKFMASHT, in which all Fe was converted to Fe^{2+}. The compositions used to calculate the phase diagrams are provided in Supplementary Table S4. The phase diagrams were calculated using the Theriak-Domino software (de Capitani & Petrakakis, 2010) using three commonly used thermodynamic datasets and allied activity-compositions relations: 1) Holland & Powell (1998; updated to version ds5.5) and a set of a-X models dating mainly from 2005 (hereafter termed dataset "HP5.5", identical to dataset HP1 of Pattison & DeBuhr, 2015); 2) Holland & Powell (2011; dataset 6.2) and the a-X models of White et al. (2014a, b) (hereafter termed dataset "HP6.2"); and 3) the "SPaC14" database and a-X models of Pattison et al. (2002), Spear & Pyle (2010) and Pattison & DeBuhr (2015). The 2005 a-X models for dataset HP5.5 comprise the following: garnet and chlorite, Tinkham et al. (2001); biotite, White et al. (2005); plagioclase, Holland & Powell (2003; ternary feldspar, Cbar1 field); white mica, Coggon & Holland (2002; margarite component omitted); ilmenite, Tinkham & Ghent (2005; ideal ternary); melt, White et al. (2007); all other phases including H2O, Holland & Powell (1998). In the SPaC14 data set, which lacks Fe^{3+}-bearing end members, all Fe was converted to Fe^{2+} and ideal on-sites mixing is assumed for all phases.

When calculating phase diagrams using dataset HP5.5 with Theriak-Domino, molar Fe^{2+} and Fe^{3+} are inputted directly as separate chemical components ("FE" and "F3") in the input file. When calculating phase diagrams using datasets HP6.2, however, Fe^{3+} cannot be entered as a separate chemical component, so that "excess oxygen" is added to the bulk composition to equal the amount of FeO that must be converted to Fe_2O_3 to give the specified molar X_{Fe3+} (= molar Fe^{3+}/total molar Fe). Both ways of inputting the bulk composition, for each rock composition, are provided in Supplementary Table S4.

Calculation of combined subsolidus-suprasolidus phase diagrams

The phase diagrams in Figs. 17 and S3 span the subsolidus and suprasolidus domains. Constructing combined subsolidus-suprasolidus phase diagrams involves a series of steps involving different assumptions about how H_2O behaves. In the subsolidus domain, below the wet melting curve (solidus), H_2O is assumed to be in excess, reflecting the assumption that a free hydrous fluid phase is released during passage through (most) prograde metamorphic reactions. In the absence of observable graphite, a_{H2O} was assumed to be unity.

In the suprasolidus domain, above the wet melting curve, H_2O is assumed to be conserved rather than in excess, reflecting its dissolution into a silicate liquid. A complication is that the H_2O content of silicate liquid decreases as pressures decreases. An effective, if strictly imperfect, solution is to calculate phase diagrams for two fixed values of H_2O content, one at higher pressure and one at lower pressure, and combine them into a single suprasolidus phase diagram. The first suprasolidus phase diagram was calculated using the thermodynamically-predicted H_2O content contained in hydrous minerals at the wet solidus at 4-5 kbar, the exact pressure depending on the specific bulk composition. Owing to the decrease in H_2O content of the melt as pressure decreases, a free H_2O phase of increasing abundance in the suprasolidus domain is predicted as pressure decreases for this fixed H_2O content. This predicted free fluid phase, which is not anticipated in real rocks above the solidus, leads to increasingly erroneous suprasolidus phase equilibria as pressure decreases. Therefore, a second suprasolidus phase diagram for pressures below ~3 kbar was calculated for a lower fixed value of H_2O, using the thermodynamically-predicted H_2O content in hydrous minerals at the wet solidus at 2.0-2.5 kbar. A consequence is that the positions of some reactions common to the two phase diagrams may differ. For example, the two bounding curves for reaction 15 (the Grt-Crd-Kfs-L field in Figs. 17 and S3, that occurs between reactions 14 and 16) differ in the two phase diagrams, resulting in two noncoincident curves for each boundary. The solution was to make single curves for each of the two bounding curves for this field by anchoring their intersections (1) with reaction 14, calculated according to the first (higher) fixed value of H_2O, and (2) with reaction 16, calculated according to the second (lower) fixed value of H_2O. With these and other small adjustments, a single suprasolidus phase diagram results.

The final step is to combine the subsolidus and suprasolidus phase diagrams into one. The subsolidus and suprasolidus phase diagrams have as a common boundary the wet melting curve. The part of the subsolidus phase diagram above the wet solidus is deleted, and the part of the suprasolidus phase diagram below the wet solidus is deleted. When these two phase diagrams are then joined at their common boundary (the wet solidus), a single phase diagram covering the subsolidus and suprasolidus domains results, as shown in Figs. 17 and S3.

Phase diagrams

Figure 17a shows a phase diagram calculated using dataset HP5.5 in MnNCKFMASHT in which all Fe was converted to Fe^{2+}, and Fig. 17b shows a phase diagram calculated using dataset 6.2 in MnNCKFMASHTO that uses the measured value of molar Fe^{3+}/Fe_{total} = 0.15. The complementary diagrams are shown in Fig. S3: Fig. S3a shows a phase diagram calculated using dataset HP5.5 in MnNCKFMASHTO that uses the measured value of molar Fe^{3+}/Fe_{total} = 0.15, whereas Fig. S3b shows a phase diagram calculated in MnNCKFMASHT using dataset 6.2 in which all Fe was converted to Fe^{2+}. Figure S3c shows a subsolidus a phase diagram calculated using the SPaC14 dataset in MnNCKFMASHT in which all Fe was converted to Fe^{2+} (the SPaC14 dataset does not accommodate Fe^{3+} and has no melt model). Figure S3d shows a suprasolidus phase diagram for an average metagreywacke (composition CVGP of Vielzeuf & Montel, 1994) calculated using dataset HP 6.2 in MnNCKFMASHTO that uses the measured value of molar Fe^{3+}/Fe_{total} = 0.08.

Consequences of variations in Fe^{3+}

Use of measured whole rock Fe^{3+} with dataset HP5.5 (Fig. S3a) results in over-estimates of magnetite (1.5-2.1 modal percent in the P-T range 2-5 kbar and 450-800 ^{o}C) in these largely magnetite-free rocks. This is most likely because few minerals in dataset HP5.5 can accommodate Fe^{3+}, and some of the minerals that do are predicted to accommodate less than what is measured in nature (e.g., predicted Fe^{3+}/Fe_{total} in biotite = ~0.03, versus vs ~0.11 measured; e.g., Dyar et al., 1991; Forshaw & Pattison, in press). The result is that incorporation of measured Fe^{3+} using dataset 5.5 raises $Mg/(Mg+Fe^{2+})$ without a counterbalancing increase in the stability of minerals like biotite that contain more Fe^{3+} than predicted. The effect is seen by comparing Figs. 17a and S3a, in which the topologies are similar but the pressure-sensitive reactions, e.g., reaction 7, in the Fe^{3+}-bearing phase diagram (Fig. S3a) are 0.3-0.5 kbar higher pressure. For these reasons, the HP5.5 phase diagram shown in Fig. 17a is the one in which all Fe was converted to Fe^{2+}. Dataset HP6.2 has more minerals that accommodate Fe^{3+}, and the predicted biotite Fe^{3+} contents and modal amounts of magnetite (0.0 to 0.4 modal %) are closer to what is observed. Therefore, the Fe^{3+}-bearing phase diagram calculated using dsHP6.2 is the one shown in Fig. 17b, noting that the difference between the Fe^{3+}-free and Fe^{3+}-bearing phase diagrams (Figs. 17b and S3b, respectively) is relatively small.

Further discussion of the pros and cons of the calculated phase diagrams are provided in the main text of the paper.

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