



# Chemical variation, modal composition and classification of granitoids

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**Abstract:** Granites (*sensu lato*) come in many types and flavours, defining distinct magmatic series/suites/types. A good classification not only gives generally accepted and understandable names to similar rocks but also links the bulk chemical composition to the stoichiometry of the constituent minerals and, potentially, also to the likely source, magmatic evolution and tectonic setting.

The ‘ideal’ granitoid classification should be based on chemical criteria amenable to an objective treatment. Statistical analysis helps to identify the most discriminant variables. The key properties are (1) acidity/maficity, (2) alkalinity (balance of Na + K v. Ca), (3) aluminosity (balance of Al v. Ca, Na and K), (4) Fe/Mg balance and (5) Na/K balance and K contents at the given SiO<sub>2</sub> level. These are used by successful classifications, e.g. the I/S dichotomy is based mainly on aluminosity, while the Frost *et al.* (2001; ‘A geochemical classification for granitic rocks’, *Journal of Petrology*, **42**, 2033–2048, <https://doi.org/10.1093/petrology/42.11.2033>) classification includes all but Na/K. Even though it is commonplace to use weight percentages of oxides, we suggest that a better strategy is to employ simple atomic parameters (e.g. millications-based) that can be directly linked to modal proportions and compositions/crystal structure of individual rock-forming minerals. This facilitates a petrological interpretation, which, in turn, can be related to petrogenesis and, ultimately, to likely tectonic setting(s).

**Supplementary material:** A database of global granite compositions, used for the statistical analysis, the R code written to generate Figures 8–14, and many diagrams in the supplementary material, including the 3D animation of the first three components from the linear discriminant analysis (LD<sub>1</sub>–LD<sub>2</sub>–LD<sub>3</sub>) are available at <https://doi.org/10.6084/m9.figshare.c.4765277>

Granite (*sensu stricto*) is a plutonic rock with a simple felsic mineralogy of quartz + alkali feldspar + plagioclase, with more than 20% quartz and plagioclase representing 10–65 vol% of total feldspar. In addition to granite, intrusive rocks with over 20 vol% of quartz are commonly referred to as ‘granitoids’, and they share many characteristics with granites *sensu stricto*. Granitoids typically span silica contents from c. 58 wt% (tonalite) to 78 wt% (alkali feldspar granite); more siliceous compositions correspond to altered rocks.

Despite this apparently simple mineralogy, granites are varied in terms of their modal and chemical

compositions, genesis and mode of occurrence. It was very soon recognized that granite can be produced by different processes or combinations thereof, even though the dispute on origin of granites is still ongoing (Young 2003; see Janoušek *et al.* 2019 for a review). In principle, one end member is direct melting of felsic crustal lithologies, and the other one is igneous differentiation of mafic melts by various types of igneous processes, with or without fluids (e.g. Clarke 1992; Pitcher 1993; Clemens 2012; Brown 2013). The diversity of granites certainly echoes this diversity in processes, and the way granites are described or classified by

different authors does reflect as much their views and opinions on granite formation as the actual range of rocks in existence.

A ‘classification’ can have different meanings, or purposes. First, there are ‘comprehensive’ classifications, whose aim, to quote Streckeisen (1976), is to give ‘each plutonic rock its proper name’. These should be objective: that is, usable without any interpretation or prior assumption. Such is, for instance, the modal QAP classification (Streckeisen 1976), or various versions of the total alkali–silica (TAS) diagram (Le Bas *et al.* 1992; Middlemost 1994). Useful as they are as descriptive and communication tools, these classifications do little in terms of helping to understand the origin of a rock, if only because of the more or less arbitrary boundaries between rock types (Glazner *et al.* 2019).

Granites tend to be associated with other igneous rocks – plutonic as well as volcanic – to form cogenetic, but not always comagmatic, batholithic ensembles, named ‘units’ and ‘superunits’ in the Peruvian Coastal Batholith by Pitcher *et al.* (1985), or ‘suites’ and ‘supersuites’ in southeastern Australia by White *et al.* (2001). Their silica contents range from 48 up to 78 wt%, thus including basic rocks, like gabbro and diorite, and intermediate rocks, besides granites. A second approach therefore relies on the identification of key properties common for the whole igneous association in the given geological unit, irrespective of the exact rock type – for instance the S/I classification of Chappell & White (1974), ilmenite/magnetite series of Ishihara (1977) or the elaborate multi-tier schemes of Debon & Le Fort (1983, 1988) and Frost *et al.* (2001). Most of the time, this approach is predicated on a specific model or paradigm for granite formation and thus may be difficult to apply beyond the original case study.

The tools used for classification have also evolved through time. By necessity, the early work was based on observations of the quantitative mineralogy (i.e. modes dominated by felsic minerals). After major-element analyses became more readily available, a second set of classifications has evolved based on either raw data or some chemical indices, including norms. The descriptive classification efforts culminated in the 1970s–80s, with the publication of comprehensive schemes based on modal compositions (Streckeisen 1976) or whole-rock chemistry (De la Roche *et al.* 1980; Le Bas *et al.* 1992). However, the combination of both approaches has never reached a consensus.

Roughly at the same time, the focus of many workers had shifted away from comprehensive, naming classifications towards the more interpretative kind (e.g. Chappell & White 1974; Debon & Le Fort 1983, 1988; Barbarin 1999; Frost *et al.* 2001). Trace-element analyses also became

sufficiently widespread to be used as a routine tool but – with the remarkable exception of the work by Pearce *et al.* (1984) – were never widely used in the granitoid classifications, certainly not to the point we know from mafic igneous rocks. The interpretation of trace-element variations is not straightforward. Many of them are controlled by saturation and fractionation of various accessory phases; in fact, truly incompatible elements are rare in granitic systems (Janoušek *et al.* 2016 and references therein). Moreover, as opposed to lavas, plutonic rocks may not represent chilled liquids but can be rather (partly) cumulative.

Even with the current advancement of analytical techniques and flood of trace-element and isotopic data, the importance of major elements has not diminished. As the key crystallochemical components, they have to reflect modal percentages of the main rock-forming minerals and thus remain important for classification purposes. In coarse-grained plutonic rocks, one of the first observations that can be made in the field is the mineralogy of the rock and it is therefore advantageous to work with the classification schemes that can be directly applied on the outcrop, without the need for laboratory analyses.

The aim of this paper is to provide a critical overview of the main types of modal and whole-rock geochemical classifications, discussing their advantages, shortcomings and interrelations. Particular emphasis is on the link between the whole-rock geochemistry and crystal chemistry of the main rock-forming minerals, and between the modal and whole-rock geochemical compositions. The purpose is not to review all systems introduced into literature, and arguably only the most influential and/or useful schemes are presented. But we explain how diverse classifications relate to or oppose each other, and how they represent, more often than not, different attempts to depict the same underlying petrochemical properties.

In fact, we observe that all the successful classifications end up using similar chemical indicators, reflecting the relatively few underlying variables defining the geochemistry of granitoids, and their best graphical representation is sought using the available diagrams or projections. This allows a statistical evaluation of a large database of major-element analyses from well-studied granitoid suites of contrasting petrology, geochemistry and petrogenetic position to be proposed that shows the difference between various granitic types. Therefore, we do not propose a new classification scheme but, rather, illustrate how some of the existing ones are actually rather efficient at depicting these variations and parameters. Finally, we offer some hints on what we regard as desirable features of any useful scheme. This could hopefully represent a stepping

stone to any new, comprehensive classification system established in the future.

## The link between modal mineralogy and major-element chemistry

### *Modal classification (the IUGS nomenclature)*

*The QAP system.* All granite types contain felsic minerals (i.e. quartz + feldspars  $\pm$  muscovite), mafic minerals (e.g. biotite, amphibole or pyroxenes) and accessory minerals (e.g. apatite, Fe–Ti oxides, zircon, monazite or allanite). As the felsic minerals constitute the dominant assemblage, the IUGS modal system is based on volume proportions of quartz, alkali feldspar (including albite with An <5) and plagioclase (An >5), plotted into the QAP ternary diagram (Streckeisen 1976; Le Maitre 2002).

The first petrologist to propose the diagram was Johannsen (1917). His system was precursory to the IUGS classification but albite was considered as plagioclase, although Johannsen was aware that some albite molecules enter the K-feldspar crystal structure. He developed the quartz–K-feldspar–albite–anorthite tetrahedron, with the 5, 50 and 95 wt% quartz dividing planes. Johannsen refined his system in 1920 (Johannsen 1920), before publishing his four classical books (Johannsen 1931, 1932, 1937, 1938).

Lyons (1976, 1977) and Bateman (1977) debated the possibility that the IUGS QAP classification of granitic rocks could not be widely accepted by British and American geoscientists and will be ‘doomed to oblivion’ (Lyons 1977, p. 254). Eventually, 40 years after the Lyons–Bateman discussion, the IUGS classification of plutonic rocks has gained wide acceptance worldwide, including much of the Anglophone community (but see also Glazner *et al.* 2019 and the related discussion).

*Problems with the QAP classification.* The IUGS-recommended QAP diagram of Streckeisen (1976) is based on objective criteria (proportion of minerals) rather than interpretations (origin of the magmas). As such, it should be stable in time, as progress in geology is unlikely to lead to new names. It is (more or less) usable in the field and it does not require further analyses. Therefore, in our view, the QAP diagram should be the reference for all classifications.

However, standard point counting is time-consuming but image analysis or, better still, dedicated automated mineralogical/petrological systems could remove much of the manual work (Janoušek *et al.* 2014; Hrstka *et al.* 2018). The colourless minerals may be difficult to recognize but the feldspars can be stained (Gabriel & Cox 1929), and optical cathodoluminescence (Marshall 1988) or backscattered electron imaging can be employed.

Unfortunately, the QAP ternary diagram is of no use if the mineral mode cannot be determined because of the texture (too fine-grained rock). Likewise, this technique is problematic for rocks possessing a strong fabric or modal layering, or for strongly porphyritic rocks when the size of a standard thin section clearly does not suffice (Chayes 1954).

To improve the statistical relevance, one can prepare polished rock slabs or even point count directly on the outcrop (Hutchison 1974). Alternatively, one can use mathematical algorithms to recombine the typical mineral compositions obtained by electron microprobe to match as closely as possible the observed whole-rock composition. The least-squares or linear programming approaches are called upon to obtain the ‘best’ mineral proportions in wt% (Wright & Doherty 1970; Albarède & Provost 1977; Le Maitre 1981; Janoušek & Moyen 2014).

Alternatively, the powder homogenized from a large volume of rock can also be utilized for Rietveld (powder X-ray diffraction) quantitative phase analysis (Maniar & Cooke 1987; Kleeberg 2009). The Rietveld analysis uses crystal structures of phases present in the sample to calculate a theoretical diffraction pattern that is then compared with the observed one. The difference is reduced through least-squares minimization (Madsen & Scarlett 2009 and references therein). However, there are many factors limiting the accuracy of Rietveld analysis: micro-absorption effects, preferred orientation of some phases (e.g. micas), chemical substitutions in minerals (notably Fe–Mg) or structural defects.

### *Linking chemistry and mineralogy: the concepts of norm and millications*

In addition to the modal analyses, bulk-rock major-element data have been acquired since the nineteenth century. One of the first attempts to describe igneous rocks through their chemical compositions was the normative system designed by Cross, Iddings, Pirsson and Washington (CIPW) (Cross *et al.* 1902). The standard mineral composition, called norm, constitutes the basis for this nomenclature. The CIPW norm comprises exclusively anhydrous minerals, and thus lacks micas and amphiboles, but includes some phases not occurring in the granitic rocks. Therefore, for granitoids, the CIPW norm tends to be different in many respects from the actual mode.

Cross *et al.* (1912) created four important adjectives: salic and femic for CIPW-normative minerals as opposed to felsic and mafic for actual minerals. The classification system itself was admittedly rigorous and logical but too complicated to gain a wide

acceptance. Regardless, and with some amendments for consistency and computer use, the CIPW norm remains an important tool for use in igneous nomenclature and genetic interpretation (Hutchison 1975; Le Maitre 2002; Verma *et al.* 2002).

Other petrologists, like Lacroix and Niggli, retained the principle of chemical indices for their own nomenclatural schemes, now largely obsolete. Still, it is worth stressing that it was Niggli (e.g. Niggli 1923, 1948) who came with the idea that the bulk major-element analyses of igneous, metamorphic and sedimentary rocks could be recast to cationic values, so that the elemental contents and relationships can be directly compared to the stoichiometry of the main rock-forming minerals. He also defined some simple cationic ratios still much used in the interpretation of igneous rocks, such as Mg# ( $Mg/(Mg + Fe^T)$ ) or  $k$  ( $K/(K + Na)$ ) (for details, see Müller & Braun 1977).

Finally, the work of Niggli and Lacroix laid foundations to more elaborate multicationic parameters of the French authors (De la Roche *et al.* 1980; Debon & Le Fort 1983, 1988), based on major elements converted to atom proportions expressed as ‘millications’. Millications are calculated from raw data (i.e. contrary to TAS diagram, not recast to 100 wt% on anhydrous basis) as  $n_\alpha \frac{C_\alpha}{MW_\alpha} \times 1000$ , where  $C_\alpha$  is the concentration of the given oxide  $\alpha$  (in wt%),  $MW_\alpha$  is its molecular weight and  $n_\alpha$  is the number of cations in the oxide formula (e.g. 2 for  $Al_2O_3$ ).

### *Major-element-based proxies to the modal QAP system*

Major elements are the key components that build the main rock-forming minerals. Thus, in theory, knowledge of the modal assemblage of a rock should be exactly equivalent to that of its major-element bulk composition. Practically, however, many rock-forming minerals, such as amphiboles, are complex solid solutions. Moreover, some phases may show complex zoning or be of distinct generations. So, in order to calculate a chemical composition of a rock (wt%), one needs not only knowledge of the modal proportions of the minerals (vol%) but also their mineral chemistries and densities, which is rarely practicable.

Although tightly connected, modal and chemical classifications cannot be easily superimposed, because each of the systems is based on a subset of more or less simplified parameters. For example, the modal scheme based on felsic minerals cannot be simply portrayed by a chemical system using silica, calcium and alkali oxides, as some mafic minerals contain these elements as well (e.g. biotite or amphibole). The major flaw of the QAP

classification is that it only considers felsic minerals and ignores other ones that contain felsic elements (Glazner *et al.* 2019).

*Adopting the total alkali–silica (TAS) diagram for plutonic rocks?* For the sake of combined information value and simplicity, the total alkali–silica (TAS) chemical diagram (Le Bas *et al.* 1986, 1992) was recommended by the IUGS for classification of unaltered fine-grained or glassy volcanic rocks (Le Bas *et al.* 1986, 1992; Le Bas & Streckeisen 1991). However, no consensus was reached for plutonic rocks, such as granites.

Several attempts have been made (e.g. Cox *et al.* 1979; Middlemost 1985, 1994; Bellieni *et al.* 1995) to adapt the TAS to plutonic rocks. However, as already pointed out by Le Bas *et al.* (1992), this may be not be practicable as the textures in coarse-grained rocks show evidence of mineral sorting and accumulation, with adcumulus growth of minerals and pore-fluid migration. Hence, the resulting coarse-grained rocks may have bulk compositions that depart from those of quickly cooled fine-grained rocks, in the genesis of which these gravity- and deformation-driven processes are admittedly minor.

Middlemost (1994) stressed that, for the same major-element chemistry, the volcanic and plutonic modal compositions may be not identical, because some minerals stable in volcanic rocks are unstable in their plutonic equivalents (heteromorphism: Lacroix 1920). The key factors responsible seem to be the contrasting cooling rates and the variable degree of re-equilibration with fluids or melts. For instance, andesites commonly contain clinopyroxene, while tonalites, their chemical counterparts, are typically amphibole-bearing. However, heteromorphism may occur even among granites; a classic example cited by Middlemost (1994) represents the dichotomy between high-temperature granites containing a single ternary feldspar (hypersolvus granites) and the more common, lower-temperature, subsolvus granites with alkali feldspar and plagioclase as separate phases, whereby excess potassium partitions into biotite.

*Plotting the CIPW normative data on the QAP diagram.* Various approaches have been attempted to replace modal proportions of felsic minerals by their normative equivalents for plotting directly onto the QAP diagram. If CIPW normative compositions are to be used, one faces many pitfalls. First, it is vital to convert the wt% of normative minerals to vol%; this can be readily done using their densities. The other problem represents the necessity of the distribution of Ab between alkali feldspar (A) and plagioclase (P). Following the recommendation of Le Maitre (1976), alkali feldspar can be set to

$Or \times T$  and plagioclase to  $An \times T$ , where  $T = (Or + Ab + An)/(Or + An)$ . The last problem that remains is the degree of iron oxidation that directly influences the normative calculations. In the CIPW system, every molecular unit of  $Fe_2O_3$  is combined with  $FeO$  to make magnetite or is even assigned to hematite, releasing an extra molecular unit of  $SiO_2$  free to form silicates. This ultimately leads to a more siliceous and less alkaline norm. So, if only total iron data are available or the samples were subject to secondary oxidation, the  $FeO/(FeO + Fe_2O_3)$  ratio needs to be adjusted (Le Maitre 1976; Middlemost 1989). However, with some care, the modal QAP values based on the original IUGS definition as proportions of felsic minerals can be amended to correspond more closely to the actual rock chemical compositions, also reflecting the presence of mafic phases.

The most abundant minerals bearing the salic elements Si, Al and K in granitoids are micas. Muscovite may be broken down into the following CIPW-normative minerals:

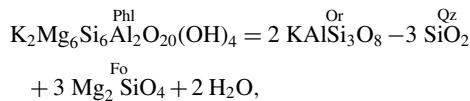


where Or is the salic component:

$$\begin{aligned} \rightarrow 796 \text{ g muscovite} &= 556 \text{ g orthoclase} \\ &+ 204 \text{ g corundum} + 36 \text{ g water} \\ \rightarrow 1 \text{ cm}^3 \text{ muscovite} &= 0.70 \text{ cm}^3 \text{ orthoclase} \\ &+ 0.25 \text{ cm}^3 \text{ corundum} + 0.05 \text{ cm}^3 \text{ water}. \end{aligned}$$

Then, 1 vol% of muscovite corresponds to 0.50A, shifting rock composition towards the A vertex of the QAP triangle.

Phlogopite, the Mg end member of biotite, may be broken into the following CIPW-normative minerals:



in which Or and Qz are the salic components:

$$\begin{aligned} \rightarrow 834 \text{ g phlogopite} &= 556 \text{ g orthoclase} \\ &- 180 \text{ g quartz} + 422 \text{ g forsterite} + 36 \text{ g water} \\ \rightarrow 1 \text{ cm}^3 \text{ phlogopite} &= 0.75 \text{ cm}^3 \text{ orthoclase} \\ &- 0.25 \text{ cm}^3 \text{ quartz} + 0.45 \text{ cm}^3 \text{ forsterite} \\ &+ 0.05 \text{ cm}^3 \text{ water}. \end{aligned}$$

Annite, the  $Fe^{2+}$  end member, yields nearly the same result. Then, 1 vol% of biotite corresponds to

(0.75A–0.25Q), and the resulting coordinates in the QAP ternary diagram become:

$$\begin{aligned} Q^* &= (\text{modal Qz} - 0.25 \text{ modal Bt}) / (\text{modal Qz} \\ &\quad + 0.5 \text{ modal Bt} + \text{modal Afs} + \text{modal Pl}) \\ A^* &= (\text{modal Afs} + 0.75 \text{ modal Bt}) / (\text{modal Qz} \\ &\quad + 0.5 \text{ modal Bt} + \text{modal Afs} + \text{modal Pl}) \\ P^* &= \text{modal Pl} / (\text{modal Qz} + 0.5 \text{ modal Bt} \\ &\quad + \text{modal Afs} + \text{modal Pl}). \end{aligned}$$

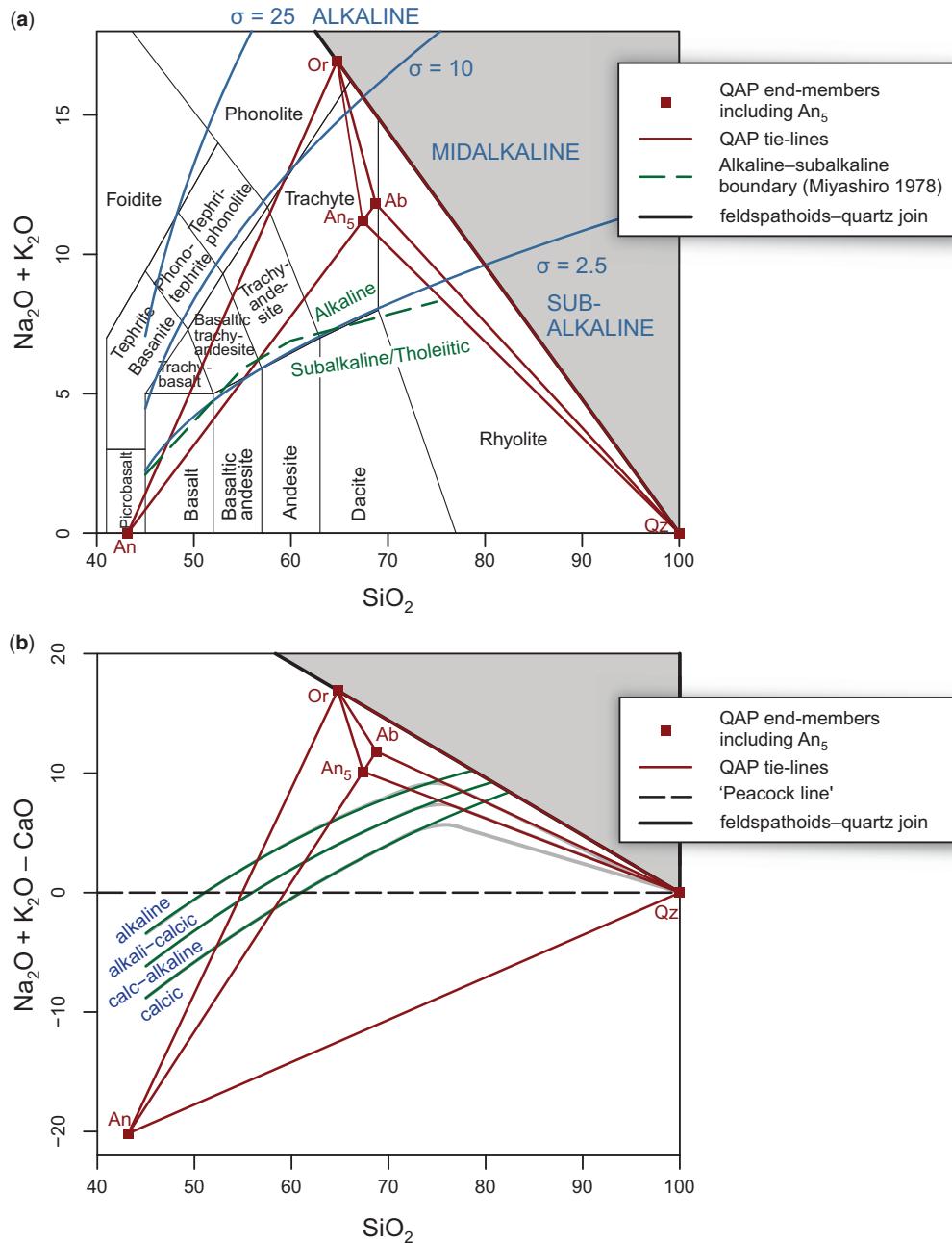
Tonalite, with (almost) no K-feldspar and K stored only in biotite, may contain 20–40 vol% of biotite. Taking the biotite effect into account, the representative point in the QAP triangle moves from the tonalite field to the quartz monzonodiorite or monzonodiorite fields. If biotite is even more abundant, say up to 60 vol%, the representative point is shifted to feldspathoid-poor monzonite. This effect will also be important in mica-rich rocks, such as melagranite, where it may account for more than 20 vol% of the mode.

Regarding amphibole and clinopyroxene, Ca-bearing and Na-rich species are Al-deficient to Al-free and correspond to insignificant amounts of CIPW-normative salic minerals (plagioclase, albite and quartz). Thus, they exert small to null effects on the QAP classification.

*Alternative norms (and their projections) more suitable for granitic compositions.* Streckeisen & Le Maitre (1979) established the Q'–ANOR binary diagram, in which  $Q' = 100 \times Qz / (Qz + Or + Ab + An)$  and  $ANOR = 100 \times An / (Or + An)$ . The authors stress that for calculation of normative Q, Or, Ab and An, molecular norms are to be preferred, such as the Barth–Niggli molecular norm. Although not frequently used, this diagram yields a very close correspondence with the modal QAP triangle and the best separation of the distinct modified alkali-lime index (MALI) types of Frost *et al.* (2001) (e.g. Whalen & Frost 2013).

Molecular norms tailored specifically for granitic rocks offer a promising alternative. Several attempts have been published, including the improved granite mesonorm (Mielke & Winkler 1979), which also incorporates biotite and amphibole. Modal norm (PERANORM), containing muscovite, biotite, garnet, cordierite and sillimanite, was developed specifically for peraluminous granitoid suites (Usdansky 1986). There also exists a modification of the classic CIPW norm including wt% of biotite and amphibole (Hutchison 1975).

A recent approach was offered by Enrique (2018), with the empirical  $2 \times Qz - (Or + Ab) - 4 \times An$  (CIPW normative minerals) ternary diagram. Instead of 17 fields as in the QAP, it displays 21



**Fig. 1.** QAP triangle converted into chemical systems. **(a)** Total alkali–silica (TAS) diagram (Le Bas *et al.* 1986, 1992). Squares: Q, P and A (alkali feldspar, plagioclase An < 5) vertices, red tie-lines: sides of the QAP triangle. Dashed green line is the alkaline–subalkaline boundary of Miyashiro (1978); solid blue lines correspond to Rittmann’s serial index ( $\sigma = (\text{Na}_2\text{O} + \text{K}_2\text{O})^2 / (\text{SiO}_2 - 43)$ ) values of 2.5, 10 and 25 separating the subalkaline, mid-alkaline and alkaline domains (Rittmann 1957). **(b)** Silica–MALI diagram (Frost *et al.* 2001). ‘Peacock line’, horizontal line with MALI = 0 corresponding to the original definition of alkali–lime index by Peacock (1931). Abbreviations of mineral names are after Whitney & Evans (2010).

fields. It has the advantage over the modal system of avoiding the problem of albite defined as An < 5 (so that the sodic series evolves in this projection continuously from the An to the Or + Ab vertices) and displaying distinct diorite ( $5 < \text{An} < 50$ ) and gabbro ( $\text{An} > 50$ ) fields.

## Current classification schemes

Existing schemes for granite classification can be, roughly, separated into three families: classification/naming diagrams for individual samples; diagrams that show the key geochemical properties (possibly also their evolution in time or space) of rocks/rock suites and petrogenetic (or interpretative) diagrams.

### Classification/naming diagrams

As a consequence of problems with determining the modal analyses (and of the advent of whole-rock chemistry), a range of diagrams was devised that use major elements for nomenclature purposes.

*The total alkali–silica (TAS) diagram(s).* The TAS diagram (Le Bas *et al.* 1986, 1992; Le Bas & Streckeisen 1991), including its modifications for plutonic rocks (Cox *et al.* 1979; Middlemost 1985, 1994; Bellieni *et al.* 1995), takes into account only three major-element oxides ( $\text{SiO}_2$ ,  $\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$  in wt%), while felsic minerals contain  $\text{CaO}$  in addition. In the standard TAS diagram, the P vertex of the QAP triangle can be remapped by the anorthite–albite  $\text{An}_5$  segment and the A vertex by the K-feldspar–albite  $\text{An}_0$ –albite  $\text{An}_5$  triangle (Fig. 1a).

*Cation-based nomenclature diagrams.* A better approach to rock-naming diagrams, therefore, should include more elements. In the realm of basaltic rocks, Yoder & Tilley (1962) established that four components are sufficient to depict at least the most meaningful variations. A similar approach can be applied to granitic rocks (although, as we discuss below, five components are required at least). Nonetheless, the  $R_1$ – $R_2$  binary diagram (De la Roche *et al.* 1980) is built on this idea (Fig. 2). It incorporates all major elements expressed as millications. In addition to the QAP triangle, it portrays the Yoder & Tilley (1962) tetrahedron (Fig. 2a–b). Designed for both volcanic and plutonic (Fig. 2c) rocks, it displays a curvilinear, instead of rectilinear, grid. In addition to its intrinsic complexity, the major concern with the  $R_1$ – $R_2$  system is that K-feldspar and albite projection points coincide, and granite, the most abundant rock in the crust, thus occupies a fairly restricted area (Fig. 2c). This is in contrast to the less common silica-undersaturated rocks that take up 75% of the

available space, and for which the diagram is thus much more suitable.

Following the same idea and also using millications, a complex classification scheme was developed by Debon & Le Fort (1983, 1988). Its core, the P–Q ('nomenclature') diagram (Fig. 3a–b), a chemical equivalent of the QAP plot, is defined by Q ( $\text{Si}/3 - (\text{Na} + \text{K} + 2 \times \text{Ca}/3)$ ), a measure of quartz abundance, and P ( $\text{K} - (\text{Na} + \text{Ca})$ ), an indication of the proportion of K-feldspar among feldspars.

The cation-based approach leads to identification of further key chemical indicators. Although not part of the naming scheme per se, they are nevertheless commonly used in granite petrology and will be dealt with in the following subsection.

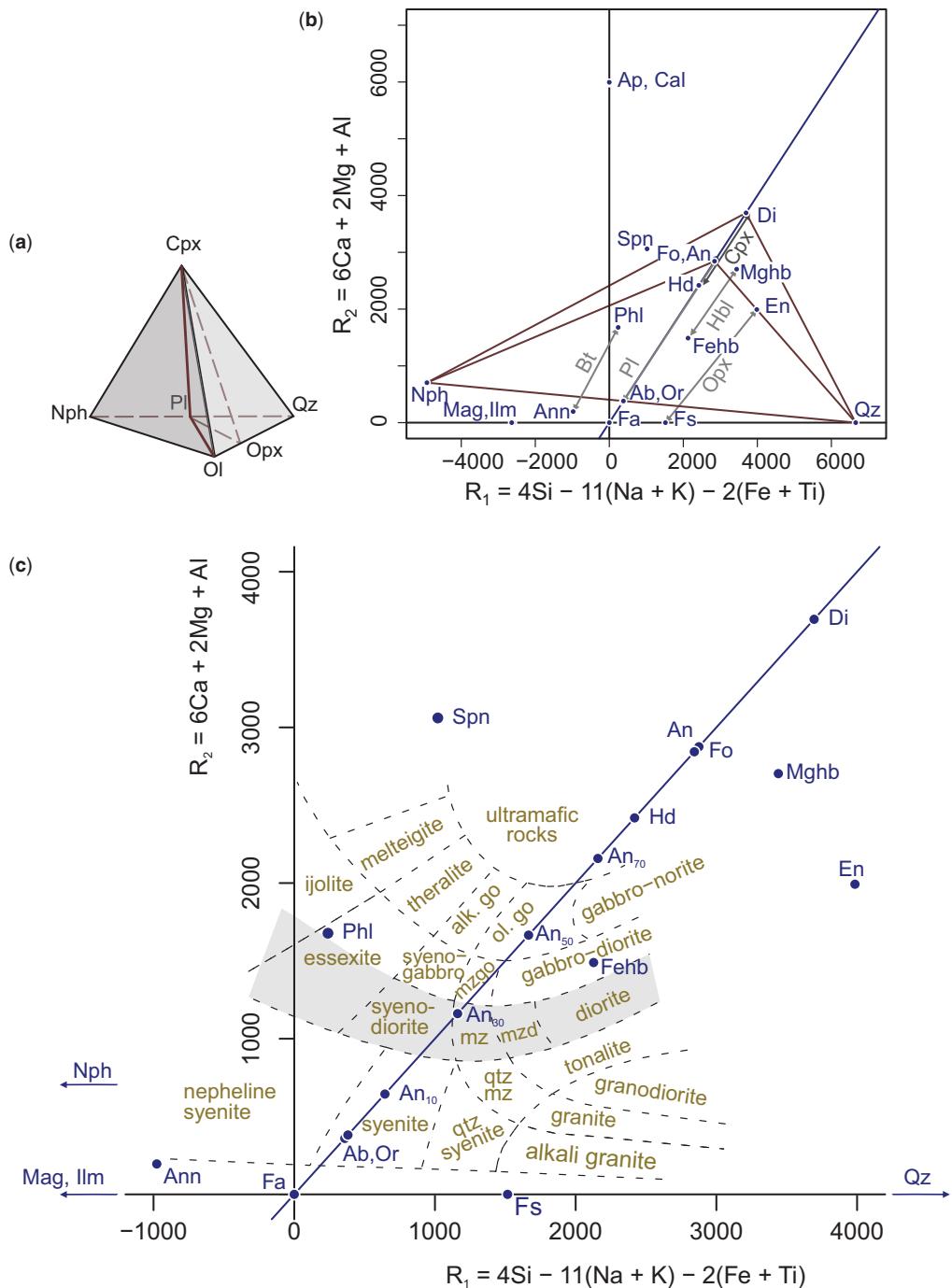
### Diagrams that show the key chemical properties of granitoids

*Aluminosity as the key property of granitoids.* As early as 1927, S.J. Shand proposed two still widely used and useful parameters (Shand 1927): A/CNK (molecular ratio of  $\text{Al}_2\text{O}_3/(\text{CaO} + \text{Na}_2\text{O} + \text{K}_2\text{O})$ ) and A/NK (molecular ratio of  $\text{Al}_2\text{O}_3/(\text{Na}_2\text{O} + \text{K}_2\text{O})$ ). The two values are built to reflect the feldspar stoichiometry, as both anorthite ( $\text{CaAl}_2\text{Si}_2\text{O}_8$ ) and the alkali end members albite and orthoclase ( $(\text{Na}, \text{K})\text{AlSi}_3\text{O}_8$ ) have A/CNK = 1; the alkali feldspars in addition show A/NK = 1. The reader's attention is drawn to the fact that the feldspar stoichiometry can be expressed as mol% oxides as A/CNK =  $\text{Al}_2\text{O}_3/(\text{CaO} + \text{Na}_2\text{O} + \text{K}_2\text{O})$  or in mol% atoms as A/CNK =  $\text{Al}/(2 \times \text{Ca} + \text{Na} + \text{K})$  – an endless source of confusion for students, and occasionally in the literature.

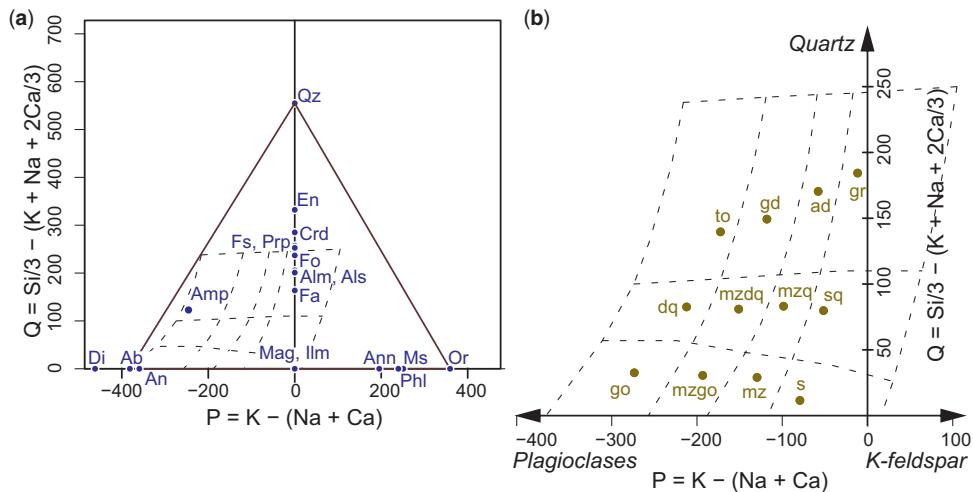
The A/CNK value is sometimes corrected assuming that apatite accommodates all phosphorus and part of calcium in the rock. But in the Al-rich and Ca-poor magmas, monazite hosts part of phosphorus as well. Importantly, apatite solubility is dramatically elevated due to the formation of aluminium–phosphate complexes (Mysen *et al.* 1999 and references therein). Consequently, apatite may not form at all, and then all P is incorporated into alkali feldspars (Piccoli & Candela 2002).

Shand's diagram A/CNK v. A/NK (Maniar & Piccoli 1989) is often used to reflect the excess, or deficit, of Al relative to feldspar, and thus the need to crystallize Al-excess (muscovite, aluminosilicates, cordierite, garnet, topaz and/or tourmaline) or Al-deficient (amphiboles, pyroxenes or magmatic epidote) minerals (Clarke 1992).

*Combining aluminosity and other properties.* It is however, seldom realized that Shand's diagram depicts another property, namely the balance between Ca and  $(\text{Na} + \text{K})$ , roughly reflecting the



**Fig. 2.** The  $R_1$ - $R_2$  multi-element system, where  $R_1 = 4\text{Si} - 11(\text{Na} + \text{K}) - 2(\text{Fe} + \text{Ti})$  and  $R_2 = 6\text{Ca} + 2\text{Mg} + \text{Al}$  in millications (De la Roche *et al.* 1980). **(a)** Yoder & Tilley (1962) tetrahedron. **(b)** Basic principles. Main rock-forming minerals, including end members of isomorphic series, are plotted. Abbreviations of mineral names are after Whitney & Evans (2010). **(c)** Plutonic rock classification. Please note that additional root names are added to the IUGS classification. Abbreviations used: qtz mz, quartz monzonite; mz, monzonite; mzd, monzodiorite; alk. go, alkali gabbro; ol. go, (olivine) gabbro. The grey field denotes the intermediate rocks.



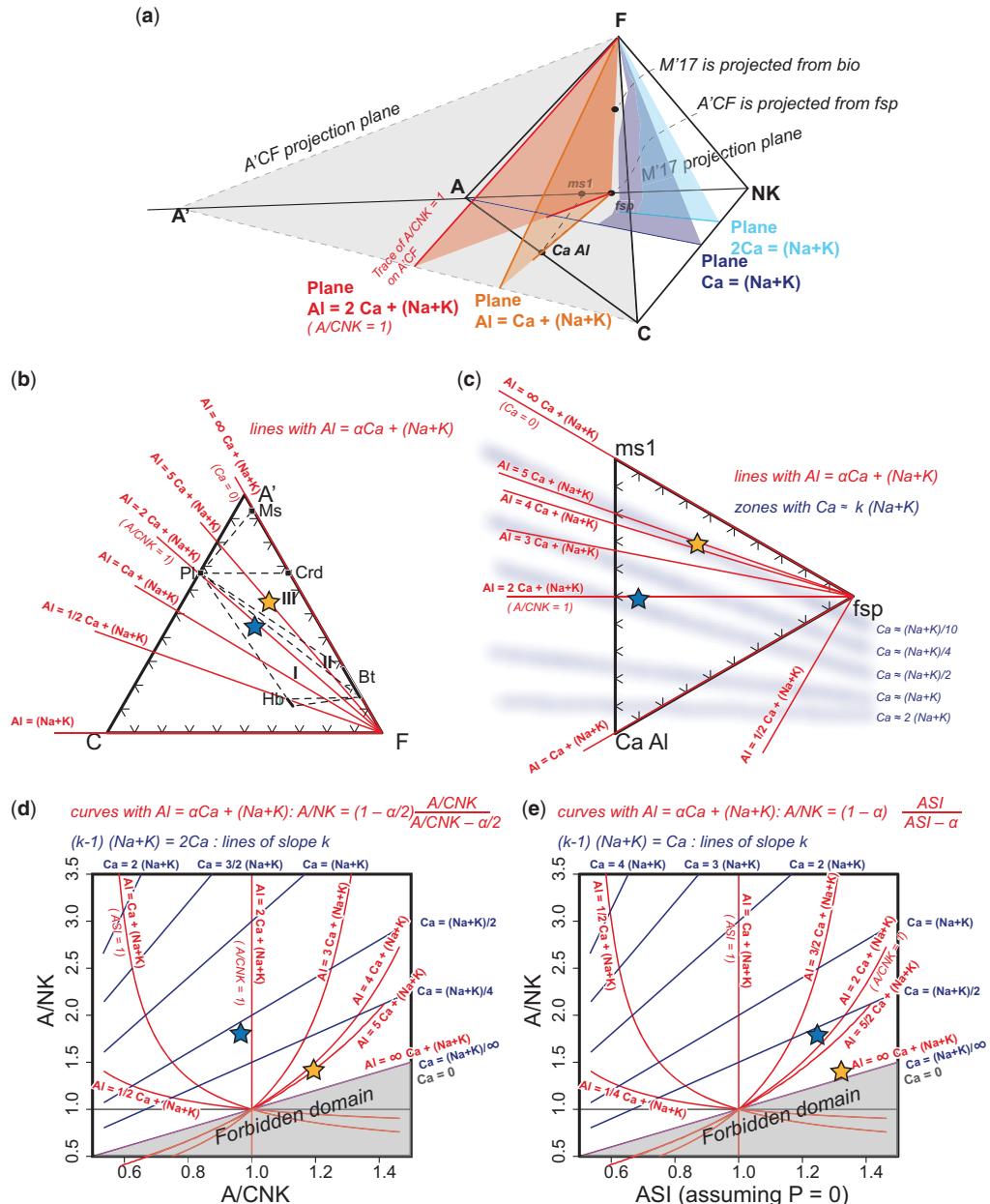
**Fig. 3.** The Debon & Le Fort (1988) system based on millications. Abbreviations of mineral names are after Whitney & Evans (2010). (a) Construction of the P–Q ('nomenclature') diagram. The lower part of the QAP diagram is represented. (b) Zoomed portion of the same diagram normally used. The reference compositions of the 12 petrographical types are, from top to bottom and from left to right: to, tonalite; gd, granodiorite; ad, adamellite (=monzogranite); gr, granite (=syenogranite); dq, quartz diorite; mzdq, quartz monzodiorite; mzq, quartz monzonite; sq, quartz syenite; go, gabbro; mzgo, monzogabbro; mz, monzonite; s, syenite. Note that not all IUGS root names are reported.

plagioclase/K-feldspar proportions. The A/CNK v. A/NK diagram, however, is not a very good projection because it combines the sum of Ca and (Na + K) and Al into a ratio. Geometrically, it amounts to projecting onto curved surfaces. The fact that ratios are used means that it is not possible to plot all minerals (when Na + K = 0, for instance). This diagram cannot consequently be used as a phase diagram (to which the lever rule applies). More elaborate projections (Fig. 4a–c) – that respect this property – can therefore be proposed (they also are, unfortunately, more difficult to plot as they require an extra step of coordinate mapping (Spear 1994) before plotting). They have been designed so that, hopefully, they can be directly linked to the modal compositions and the likely genesis of magmas.

If we project the major-element compositions from quartz and K-feldspar, we can define three pseudocomponents of the so-called A'CF system, the origin of which goes back to Eskola (1915). These are (atomic)  $A' = Al - Na - K$ ,  $C = Ca - 1.67 \times P$  and  $F = Fe + Mg$ . This projection (Fig. 4b) depicts aluminosity in a slightly different way: lines of constant A/CNK cannot be plotted on this diagram but, at least in the case of  $P = 0$ , reference lines such that  $Al = \alpha Ca + Na + K$  are present by construction (of course,  $\alpha = 2$  corresponds to the line where A/CNK = 1 and  $\alpha = \frac{Al - Na - K}{Ca}$ ). These lines unfortunately correspond to curves in Shand's diagram (Fig. 4d), such that a direct

comparison is difficult. The A'CF diagram includes a mafic component (F) that is not visible on Shand's diagram and, therefore, very efficiently represents the main rock-forming minerals (in addition to alkali feldspar) of a granite. It is very good at separating chemical analyses corresponding to granites that are Hbl–Bt-bearing, Bt only, or those containing peraluminous phases such as Crd, Grt, And or Ms (Hine *et al.* 1978; White 1990). This is, in fact, the chemical basis of the S/I classification of Chappell & White (1974) discussed later in the text.

Another attempt has recently been offered by Moyen *et al.* (2017) (Fig. 4c). In this projection, adapted to granitoids (and woefully inadequate for mafic rocks), the sample composition is simplified to four main components (a differentiation component ( $Fe + Mg$ ), Al, Ca and  $(Na + K)$ ). Since this diagram aims to magnify the difference between suites, more than the fractionation history, the data are projected from the differentiation (mafic) component onto the plane defined by the other three. Finally, in order to further enhance the differences between rocks, the compositions are projected from biotite, on a subset of the Al–Ca–Na + K plane defined by the apices ms1 ( $3Al + 2(Na + K)$ ), fsp ( $Al + Na + K$ ) and CaAl ( $Ca + Al$ ). The new coordinates are calculated from millications as follows:  $x_{ms1} = Al - Ca - NK$ ,  $x_{fsp} = -2Al - 2Ca + 3NK - 1/3FM$  and  $x_{CaAl} = Ca$ , where FM = Fe + Mg and NK = Na + K, and plotted in ternary



**Fig. 4.** Projection and representation of chemical properties in different diagrams. Here we explore the quaternary system,  $F = \text{Fe} + \text{Mg}$ ,  $A = \text{Al}$ ,  $C = \text{Ca}$ ,  $NK = \text{Na} + \text{K}$ , all molar. This system can be represented as a tetrahedron (a). Several ternary projections can be derived from this tetrahedron, depending on the projection plane and origin. (b) A'CF projection (with  $A' = A - \text{NK}$ ), projected from alkali feldspar (fsp). (c) Projection proposed by Moyen *et al.* (2017), projecting from biotite (bio =  $A + \text{NK} + 3F$ ) on the plane defined by  $\text{ms1} = 3A + 2\text{NK}$ ,  $\text{CaAl} = A + C$  and  $\text{fsp} = A + \text{NK}$ . (d) The common Shand projection,  $\text{A/CNK} = \text{A}/(2C + \text{NK})$  v.  $\text{A/NK}$ . (e) Subtly different Frost *et al.* (2001) projection,  $\text{ASI} = \text{A}/(C + \text{NK})$  v.  $\text{A/NK}$ . In each diagram the orange star is the average CPG/MPG granite, and the blue star is the average ACG, both as classified in this work. The red/orange planes (on a) and curves (on b–e) represent the loci of compositions for which  $\text{A} = \alpha\text{C} + \text{NK}$ .  $\alpha = 1$  is Frost *et al.*'s (2001) ASI = 1 and  $\alpha = 2$  is  $\text{A/CNK} = 1$ . In barycentric projections, projected from a point that belongs to the planes in question (fsp in b and bio in c), the red planes become straight lines that happen to intersect one apex. On ratio-based

coordinates. This diagram, being projected onto a surface very similar to Shand's diagram, shows comparable features. However, as it is a linear projection, it is possible to plot mineral compositions and to use it as a phase diagram, to which the lever rule applies. In this diagram, different granitic series plot along lines that fan out of the right (fsp) apex (Fig. 4c) (see also Moyen *et al.* 2017; Moyen & Laurent 2018) and correspond to constant values of  $\alpha$ , as defined above. In addition, the Ca/(Na + K) ratio is reasonably well expressed (see details in Fig. 4c).

Debon & Le Fort (1983, 1988), in addition to the Q and P parameters already discussed, defined other key indicators. In the B–A ('characteristic minerals') diagram (Fig. 5a), A (Al – (Na + K + 2 × Ca)) shows the aluminium balance to Ca and alkalis (separating peraluminous v. metaluminous rocks, thus being analogous to the A/CNK index of Shand) and B (Fe + Mg + Ti) measures the maficity. A more detailed subdivision of the peraluminous domain can be achieved in the modified B–A plot of Villaseca *et al.* (1998) (Fig. 5b).

The next step of the Debon & Le Fort (1983, 1988) scheme involves comparison of the studied samples with 12 reference compositions of the corresponding petrographical type(s) in the ternary plot involving weight proportions of quartz, dark minerals and feldspars (not shown). The classification is complemented by an assessment of the balance of alkalis (Na + K: potassic, sodi-potassic and sodic associations; Fig. 5c) and ferromagnesian components (Fe and Mg: magnesian and ferriferous associations; Fig. 5d).

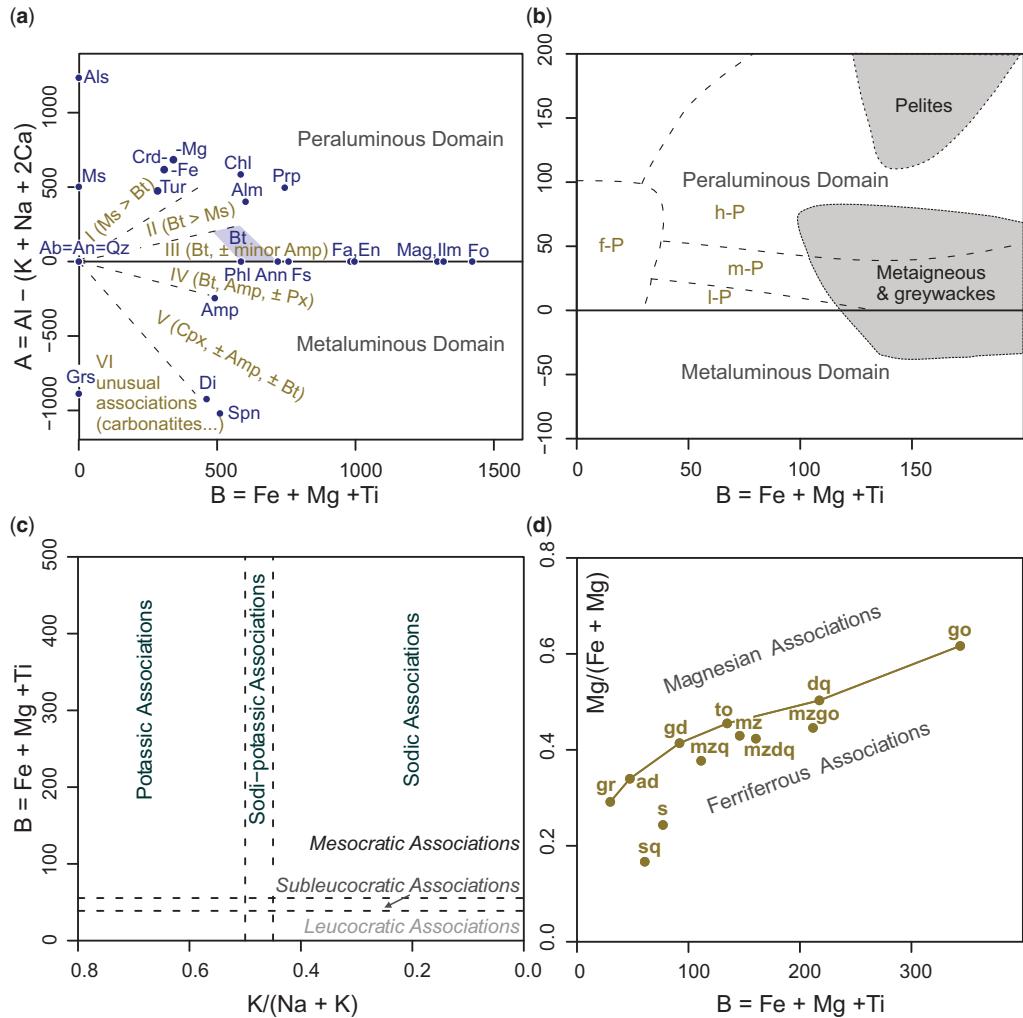
A more recent system is the multi-tiered scheme proposed by Frost *et al.* (2001). The first tier is determined by the Fe-number, defined as  $\text{FeO}/(\text{FeO} + \text{MgO})$ , or by  $\text{Fe}^*$ ,  $(\text{FeO}^\text{T}/(\text{FeO}^\text{T} + \text{MgO}))$ , in which  $\text{FeO}^\text{T} = \text{FeO} + \text{Fe}_2\text{O}_3/1.1113$ . Note that both indexes are calculated as weight proportions, contrary to the common usage of the term 'Fe-number' (as well as 'Mg-number') for molecular proportions. The second tier, also in oxide weight percentages, is the MALI, formulated as  $\text{Na}_2\text{O} + \text{K}_2\text{O} - \text{CaO}$  and plotted on a  $\text{SiO}_2$ –MALI diagram. The third is the aluminium saturation index corrected

for apatite (ASI), in Frost *et al.* (2001) and Frost & Frost (2008) described somewhat confusingly as 'the molecular ratio  $\text{Al}/(\text{Ca} - 1.67\text{P} + \text{Na} + \text{K})$ '. Taking into account the ideal apatite formula of  $3\text{CaO.P}_2\text{O}_5.1/3\text{CaF}_2$ , in our opinion this should be expressed more rigorously in molar proportions of oxides as  $\text{Al}_2\text{O}_3/(\text{CaO} - 3.33 \times \text{P}_2\text{O}_5 + \text{Na}_2\text{O} + \text{K}_2\text{O})$  or in cations as  $\text{Al}/(2 \times (\text{Ca} - 1.67 \times \text{P}) + \text{Na} + \text{K})$ . Finally, the comparatively rare peralkaline type is discriminated from the more common metaluminous type by the alkali index (AI), measured in molecular proportions by  $(\text{Al}_2\text{O}_3/(\text{Na}_2\text{O} + \text{K}_2\text{O}))$  or in cations by  $(\text{Al}/(\text{Na} + \text{K}))$ . Hence, granitoids are distinguished successively into: (i) magnesian and ferroan; (ii) alkaline, alkali-calcic, calc-alkaline and calcic; and (iii) peraluminous, metaluminous and peralkaline types.

*Main components of granitoids.* On the basis of the overview of popular diagrams in common use, it can be concluded that, from a geochemical point of view, the composition of granitoids can be best described in terms of a handful of key parameters:

- The 'degree of differentiation' – since Harker (1909), the most commonly employed proxy for degree of differentiation in granitoids represents  $\text{SiO}_2$  (wt%). Since silica is negatively correlated to most other oxides (in particular mafic oxides such as  $\text{MgO}$ ,  $\text{FeO}$ ,  $\text{TiO}_2$  and, to a lesser degree,  $\text{CaO}$ ), these tend to carry the same information. The differentiation index (DI) of Thornton & Tuttle (1960), or parameters such as the 'si' value of Niggli (1923) or multicationic values Q and B (maficity) of Debon & Le Fort (1983, 1988), are closely related.
- The 'alkalinity' – that is, the balance between the feldspar-forming alkaline and alkaline-earth cations (Na and K, Ca). Relevant indicators include normative amounts of feldspars obtained from various norms, the Ab–An–Or normative diagram (O'Connor 1965), the alkali–lime index (Peacock 1931) and MALI (Frost *et al.* 2001), as well as the P multicationic parameter of Debon & Le Fort (1983, 1988). As shown in Figure 4d, the excess of Ca in the rock can be also shown in Shand's diagram.

**Fig. 4. Continued.** diagrams, however (d & e), these planes become curves. The blue planes (in a) are the loci where  $\text{Ca} = \beta\text{NK}$ . They correspond to lines in (d) & (e). Since the projection point for (b) & (c) does not belong to these planes, they do not correspond to a particular locus. However, (b) is projected from bio that lies close to the blue planes in quaternary space. For reasonable compositions (in or near the bio–msl–fsp–CaAl volume) the projection direction is therefore at a low angle to the blue planes that consequently project as narrow linear bands. The important message here is that each of the four projections depicted in this figure represent the same, or similar, properties but with different geometry and different emphasis. The 'red' property is shown quite clearly in all four diagrams. The 'blue' property is very well expressed in (d) & (e), less well expressed in (c) and not at all in (b). On the other hand, (b) allows properties to be depicted related to the amount of mafic components in the system (hence, related to the  $\text{SiO}_2$  content or the degree of differentiation), information that is lost in the other representations.



**Fig. 5.** The Debon & Le Fort (1988) system II. (a) B-A ('characteristic minerals') diagram. Six fields are represented: the I, II and III fields are for decreasingly peraluminous rocks; the IV, V and VI fields are for increasingly Al-deficient rocks. (b) More detailed subdivision of the peraluminous domain proposed by Villaseca *et al.* (1998). I-P, low peraluminous; m-P, moderately peraluminous; h-P, highly peraluminous; f-P, felsic peraluminous suites. Compositional fields for the possible metasedimentary sources of anatetic granitic magmas are also shown. (c) Binary plot of  $K/(Na + K)$  v.  $B$  for aluminous associations. (d) Binary plot of  $B$  v.  $Mg/(Mg + Fe)$ .

- The 'aluminosity' – that is, the excess/deficiency of alumina with respect to the stoichiometry of feldspars. This is the rationale of many diagrams, using ratios such as  $A/CNK$  with  $A/NK$  (Shand 1927), normative values (normative corundum or aegirine/sodium metasilicate: Cross *et al.* 1902), linear combination of variables (the A cationic parameter of Debon & Le Fort 1983) or more complex representations ( $A'/CF$  system, projection from biotite of Moyen *et al.* 2017 or the  $\alpha$  parameter defined in the current paper – see Fig. 4).
- The balance between Fe and Mg – (see Niggli 1923; Irvine & Baragar 1971; Miyashiro 1974; Debon & Le Fort 1988; Frost *et al.* 2001), particularly for the alkaline (or nearly alkaline) rocks.
- The balance between Na and K – this can be expressed as the  $k$  value of Niggli (1923) or simply by various  $K/Na$  ( $K_2O/Na_2O$ ,  $K/(Na + K)$ , etc.) ratios. Since the  $K_2O$  is generally positively correlated to  $SiO_2$ , its absolute values are not particularly diagnostic. Even the simple  $SiO_2-K_2O$

diagram (Peccerillo & Taylor 1976), which shows how the K<sub>2</sub>O values evolve with rising SiO<sub>2</sub>, effectively captures most of this information.

Clearly, diagrams relating two versions of the same parameter are useless, as they will only show a (nearly) 1:1 correlation. It is more useful to plot diagrams showing how two of these properties are related and evolve as a function of each other. Another implication is that, since the system has five degrees of freedom and only two can be plotted in 2D (three in a ternary plot focusing on relative amounts rather than absolute values), it is impossible to capture all the diversity of granitoids in a single diagram.

### Petrogenetic classifications

Granitic composition, tending to the thermal minimum in the SiO<sub>2</sub>–NaAlSi<sub>3</sub>O<sub>8</sub>–KAlSi<sub>3</sub>O<sub>8</sub>–H<sub>2</sub>O haplogranitic system, or ‘petrogeny’s residua system’, may be produced by two opposite processes: (i) strong fractionation of all silicate melts, controlled by silica-poor assemblages; or (ii) partial melting of silicate rocks. This has been explicitly stressed in the Tuttle & Bowen’s memoir (Tuttle & Bowen 1958). Moyen (2019b) has stated that two competing views on granitoids are (still) present in the literature and in the scientific community: ‘fractionation’ people who tend to regard most granitoids as resulting from fractional crystallization of primitive basaltic melts, and ‘melting’ people who view granites as chiefly melts of continental crustal lithologies. In the first view, differences between granites reflect in part the composition of the primitive basaltic magma, and largely the mechanism of fractionation processes (depth/pressure, temperature, H<sub>2</sub>O, redox state, etc.). In the second view, the compositional differences imply distinct sources and conditions of melting. This is of course a somewhat oversimplified view, as these two models are end-member scenarios. At least it helps to put the debate in perspective.

The combination of petrographical and geochemical data, including trace elements and isotopes, constitutes the spearhead of recent granitic classifications (for reviews, see Barbarin 1990, 1999; Clarke 1992, 1996). These classifications are at least in part interpretative: their aim is to propose a petrogenetic framework for diverse types of granitoids. Thus, they generally focus on one criterion deemed more important than the others, because it is thought to carry the key petrogenetic information. As a result, such classifications work mostly under one paradigm of granite formation. Another possible limitation of these approaches is that they rely on a limited number of categories, and each rock must therefore end up being classified as one of them, leaving no room for other alternatives. These

classification schemes also tend to involve some degree of interpretation, with non-numeric criteria or personal judgement such as ‘rich in...’ or ‘plot mostly in the field of...’. Invariably, these classifications work by analogy – they compare the studied rocks with samples of a presumably well-established origin. We illustrate here some of the most popular genetic classifications; the reader is referred to figure 3 of Barbarin (1999) for a more complete overview.

*The alphabetical (S–I–M–A) classification.* One of the oldest such approaches, still widely used, is the alphabetical (S–I–M–A) classification. In 1974, Bruce Chappell and Alan White introduced a genetic classification of granitic rocks based on their extensive study of the Lachlan Fold Belt in eastern Australia. They described two contrasting suites of granitic rocks, whose major-element compositions are interpreted to largely mimic their sources. These are: (i) relatively sodic, metaluminous to subaluminous mafic–felsic granitoids thought to have originated by the partial melting of meta-igneous rocks (thus termed I-type); and (ii) relatively potassic, peraluminous granitoids with restricted, but relatively high, SiO<sub>2</sub>, presumably of metasedimentary parentage (thus termed S-type) (Chappell & White 1974). Contrasting aluminosity of the two suites is reflected by distinct modal mineralogy, best visualized by the A' CF ternary projection (Hine *et al.* 1978). The metaluminous chemistry of the I-type granites is demonstrated by the presence of hornblende and titanite (field I in Fig. 4b) and the peraluminosity of the S-type granites by muscovite, cordierite, aluminosilicates and monazite (field III). Lastly, the two types show distinct enclave populations (igneous-looking mafic microgranular v. restitic metasedimentary) and Sr isotopic compositions that tend to be more radiogenic in the S-type granites. Over the years, this classification has been further elaborated (e.g. Chappell & White 1992, 2001; Chappell 1999).

Soon thereafter, Loiselle & Wones (1979) recognized a specific group of granitoid rocks generated along rift zones and within stable continental blocks, and coined for them the term anorogenic or, in short, A-type granite. This group was subsequently broadened to accommodate all high-temperature (hypersolvus) granites emplaced in within-plate or post-collisional settings (designated as A<sub>1</sub> and A<sub>2</sub> groups by Eby 1992). Loiselle & Wones (1979), Collins *et al.* (1982), Whalen *et al.* (1987) and Eby (1990) have shown that A-type granites can be distinguished from their I- and S-type counterparts by elevated contents of alkalis, FeO<sup>T</sup>, F, high field strength elements (HFSEs: especially Zr, Nb and Ta) and REEs (except Eu), as well as high Fe/Mg and Ga/Al ratios. Typical features also include low concentrations of CaO and trace

elements compatible with mafic silicates (Co, Sc, Cr, Ni) or feldspars (Ba, Sr, Eu). The chemistry is reflected by the occurrence of characteristic Fe-rich mafic silicates (annite, hastingsite or fayalite), including alkali amphiboles and/or pyroxenes (arfvedsonite, riebeckite, aegirine) in peralkaline rocks. Still, the geotectonic setting and petrogenesis of the A-type granites remain disputed and many models have been proposed for their genesis (see Bonin *et al.* 1998; Bonin 2007 for reviews).

The last addition were the M-type granites, defined as having been derived by melting of subducted oceanic crust at continental margins (White 1979). In a broader sense, this group includes granitoids that originated by differentiation of mafic magmas, sourced from the mantle, in island arcs (Alonso-Perez *et al.* 2009). This definition gained less traction than the others, and M-types are hardly used nowadays.

Undoubtedly, the introduction of the S- and I-type classification in the mid-1970s represented a true revolution; from times of largely descriptive petrographical and geochemical studies emerged a coherent, process-orientated granite science. The identification of aluminosity as a key parameter was epochal, and highly influential in recognizing the links between field and petrological types, chemical properties, and possible genesis. Although still widely used, the S–I–M–A classification endured much criticism, as summarized, for instance, by Clarke (1992) and Frost *et al.* (2001).

A first issue, common to all ‘pigeonhole’ classifications, is that it allows only a handful of outcomes (I, S, A ± M). This is further compounded by incorrect use of the classification by many workers, who tend to pick only one or two from the whole set of diagnostic features, neglecting the others. In separating I- and S-type granites, the most commonly abused is the A/CNK threshold of 1.1.

A second, perhaps more fundamental, issue is that the classification relies much on one criterion, the aluminosity of the rock (either expressed directly by chemical indices such as A/CNK, or mineralogically through the presence of metaluminous or peraluminous minerals and/or specific accessories). A peraluminous rock (regardless of the proxy used) is S-type, a metaluminous rock is I-type. But metaluminous compositions include a wide variety of granitoids with contrasting chemical characteristics (ferroan or magnesian, alkalic or calcic, sodic or potassic, etc.) and, most likely, origins.

Another unsatisfying aspect is that the scheme uses inconsistent criteria – inferred source, presumably reflected by the composition of the generated magmas (I, S, M) or geotectonic setting and specific chemistry but with an unspecified source (A) (Creaser *et al.* 1991). This causes confusion: for instance, A<sub>1</sub> granites of Eby (1992) can be produced

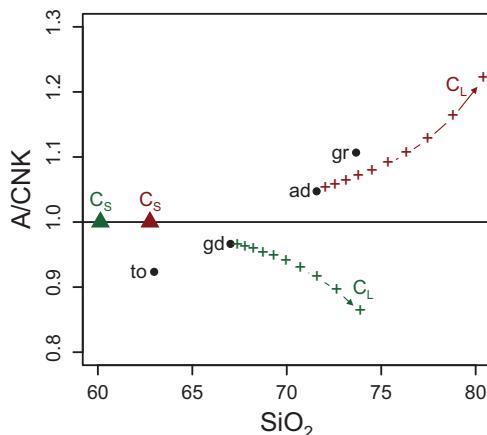
by differentiation of ocean island basalt (OIB)-like/continental tholeiitic basaltic magmas, and thus would be classified either as A- or M-type granites.

Lastly, it has long been realized that the S/I terminology is based on strong model assumptions: S-types are proposed to form by melting of sediments, I-types of igneous sources. This ignores a range of other petrogenetic possibilities. For instance, partial melts of relatively immature greywackes, such as of those deposited close to their magmatic-arc sources, would be likely to acquire an I-type-like chemistry, including relatively low A/CNK values and comparably primitive Sr–Nd isotopic signature. Although technically S-type granites, they will be undistinguishable from some orthogneiss-derived melts, at least solely on the whole-rock geochemical grounds (Sylvester 1998).

*On the origin of peraluminous magmas.* Originally, Chappell & White (1974) assumed that S-type granites are generated solely from mature metasedimentary sources that went through the weathering cycle (stripping them of Na and Ca – hence, the elevated A/CNK values), so mostly metapelites. On the other hand, the peraluminous granites can also crystallize from metaluminous parental magmas that underwent extensive fractional crystallization of metaluminous phases (such as hornblende or pyroxene) (Zen 1986) or can be generated by partial melting of metaluminous sources, felsic–intermediate quartzo-feldspathic (metagreywackes or orthogneisses) sources or, less commonly, mafic sources (Miller 1985; Sylvester 1998; Gao *et al.* 2016 and references therein). It is worth stressing that the initial, just slightly peraluminous or metaluminous character of the melt can be magnified by extensive fractionation of feldspars, despite the fact that these minerals are neutral as regards their Al saturation index (A/CNK = 1) (Fig. 6; see also Zen 1986).

*On the evolution of magmas.* The potential crustal sources are inherently heterogeneous. In addition, the magma may assimilate country-rock material, and coeval contrasting felsic and mafic magmas may interact, producing hybrid granitoids. The latter problem has been recognized early (Didier *et al.* 1982), eventually leading Castro *et al.* (1991) to define H-type granitoids, variously hybridized mantle-derived and pure crustal end members (M- and S-type granites).

*On the controls of melt chemistry.* Melt compositions are controlled by more than their source. A wide spectrum of granitic compositions may originate from the same crustal rock, reflecting distinct pressure–temperature–composition (*P*–*T*–*X*) conditions and mechanisms of melting (vapour-present, vapour-deficient or vapour-absent). These factors,



**Fig. 6.** Binary plot SiO<sub>2</sub> (wt%) v. A/CNK portraying the evolution of two hypothetical granitic suites evolving from slightly metaluminous granodiorite (green) and weakly peraluminous (=monzogranite or 'adamellite' – red) parental compositions by up to 50% fractional crystallization of feldspars (30% of Or and 70% of Pl An<sub>40</sub> v. 50% of Or and 50% of Pl An<sub>30</sub>), respectively. Model compositions of rocks are from Debon & Le Fort (1988). Note that both trends are distinctly curved and result in magnification of the originally small Al<sub>2</sub>O<sub>3</sub> v. CaO + Na<sub>2</sub>O + K<sub>2</sub>O (molar) disproportions.

in turn, control the degree of melting and melt viscosity, two of the crucial parameters determining how easily the melt can separate from unmelted residua and peritectic phases, whose occurrence can modify profoundly the chemistry of the resulting magma (Chappell *et al.* 1987; Stevens *et al.* 2007; Villaros *et al.* 2009; Clemens *et al.* 2011; Clemens & Stevens 2012). In turn, in terms of modal and major-element compositions, similar granites may originate from variable combinations of sources and processes, often reflecting the tendency of granitic suites to evolve towards the minimum compositions in the Qz–Ab–An ternary system. However, this is an inevitable problem of all classification schemes.

Taken together, despite some criticism, the alphabetical classification is still far from being obsolete. The terms S-, I- and A-type granites remain useful in the scientific communication, when referring to the pure and typical end members close to the original definition. Like all classification terms, they are mostly appropriate when employed in the context in which they were originally devised (i.e. geodynamic settings dominated by melting of crustal lithologies: collision/post-collisional settings, inverted back-arcs, etc.) but much less relevant in other situations.

*Links between chemical composition and geotectonic setting.* The difference in composition between

granites from different settings was recognized very early on (see Pitcher 1987; Pearce 1996b for reviews).

Starting with major elements, the  $R_1$ – $R_2$  projection (De la Roche *et al.* 1980) also has geotectonic and petrogenetic implications for granitoid suites (Batchelor & Bowden 1985). First of all, there is a systematic change in the  $R_1$ – $R_2$  parameters through the orogenic cycle, and fields of different geotectonic settings can be delineated: mantle fractionates (e.g. oceanic plagiogranites), destructive plate margin, post-collision uplift, late-orogenic, syn-collisional (i.e. S-type granites), post-orogenic and anorogenic. The diagram allows either compositions of ideal phases (Fig. 2b) or real mineral chemistries from the studied igneous suite to be plotted. The operation of processes such as partial melting, fractional crystallization or binary mixing then can be readily recognized and quantified, and the lever rule is applicable. Vectors are linear, unless there is a change in composition of the fractionating solid assemblage (e.g. increasing An in plagioclase in the course of progressive melting). On the other hand, the diagram fails to distinguish felsic syn-collisional, anatetic (S-type) (leuco-)granites from strongly fractionated members of the other suites, as all fall into the ternary minimum of the Qz–Ab–An system.

The same logic of identifying coherent suites of rocks, rather than focusing on the nature of individual samples, was followed by Debon & Le Fort (1983, 1988). Their multicationic plots, although not actually geotectonic, distinguish three main types of magmatic associations, namely cafemic (broadly corresponding to I-, A- and M-type granitoids), alumino-cafemic and aluminous (broadly corresponding to S-type granitoids). The most important criteria are the position and slope of evolutionary trends in the B–A plot (Fig. 5a). This includes something very similar to the S/I dichotomy (peraluminous v. metaluminous) but allows several associations within each broad realm to be identified. Using other graphs, each of the cafemic and alumino-cafemic associations can be further subdivided into tholeiitic, calc-alkaline, subalkaline (monzonitic) and alkaline. The aluminous associations are further classified based on the quartz content, colour index, K/(Na + K) ratio, and the relationships between quartz, dark minerals and alkalis (e.g. see Fig. 5c). The whole classification scheme, even though in many respects undeniably useful, was proven to be too complex to become popular, especially in North America.

However, a collection of relatively simple diagrams was proposed by Maniar & Piccoli (1989) for major-element-based tectonic discrimination of granitic rocks into oceanic plagiogranites (OP), island-arc granitoids (IAG), continental-arc granitoids (CAG), continental-collision granitoids (CCG),

post-orogenic granitoids (POG), rift-related granitoids (RRG) and continental epeirogenic uplift granitoids (CEUG). The set of six diagrams is based on major-element oxides in wt%, except for molar A/NK and A/CNK values of Shand (1927). The rationale is that only the OP have  $K_2O < 1$  wt%; compared to the CEUG and RRG, the IAG, CAG and CCG are  $Al_2O_3$ -rich and  $FeO^T$ -poor, both in terms of absolute  $FeO^T$  contents and their relationship to  $MgO$ . The IAG are metaluminous–subaluminous, CAG with OP variably metaluminous–peraluminous and CCG weakly to strongly peraluminous. CEUG and RRG are metaluminous to peralkaline.

Four trace-element diagrams for the discrimination of geotectonic environment of granitoid rocks, proposed by Pearce *et al.* (1984), gained unprecedented popularity. Based on a combination of five trace elements (Y, Nb, Rb, Yb and Ta), the following geotectonic settings may be deduced: ocean ridge granites (ORG), volcanic-arc granites (VAG), within-plate granites (WPG) and syn-collisional granites (syn-COLG). The authors specifically stressed that the post-collision granites cannot be easily discriminated, as they originate by the interaction of magmas coming from contrasting crustal and mantle sources, some subduction-modified and others of intraplate character depending on the composition of the continental plates involved and the collision geometry (Pearce 1996b). Because the source is not unique, the setting is not uniquely characterized and the data points may enter the VAG and WPG, or even syn-COLG, domains (Pearce *et al.* 1984).

The Hf–Rb/30–Ta  $\times$  3 ternary diagram produced by Harris *et al.* (1986) is partially successful in distinguishing post-collisional granitic suites. It identifies the following groups: (i) pre-collision calc-alkaline (volcanic-arc) intrusions, which are mostly derived from mantle modified by a subduction component and are characterized by selective enrichments in large ion lithophile elements (LILEs); (ii) syn-collisional peraluminous intrusions (leucogranites), which may sample the hydrated bases of continental thrust sheets, and are characterized by high Rb/Zr and Ta/Nb with low K/Rb ratios; (iii) late- or post-collision calc-alkaline intrusions which may come from a mantle source but undergo extensive crustal contamination and can only be distinguished from volcanic-arc intrusions by their higher Ta/Hf and Ta/Zr ratios; (iv) post-collision alkaline intrusions that are derived from mantle lithosphere beneath the collision zones and which carry high concentrations of both LILEs and HFSEs. The problem of distinguishing late-orogenic, post-orogenic and anorogenic granites, and constraining their sources and petrogenesis, has attracted considerable attention, especially in

1990s (Sylvester 1989, 1998; Bonin 1990, 2004; Rogers & Greenberg 1990; Bonin *et al.* 1998).

The diagrams of Schandl & Gorton (2002) are also based on a combination of four presumably immobile trace elements (Ta, Yb, Th and Hf) with the aim of deciphering the geotectonic setting of felsic volcanic suites, specifically those associated with the volcanogenic massive sulfide (VMS) deposits. The Th/Yb v. Ta/Yb and Th/Ta v. Yb diagrams are divided into three fields: oceanic arcs, active continental margins (ACM) and within-plate volcanic zones (WPVZ). The remaining two diagrams demonstrate that the ACM are, compared to WPVZ, characterized by higher Th and Th/Hf at the given Ta and Ta/Hf.

The geotectonic diagrams presented above are largely empirical, and boundaries were fitted by eye. More recently, statistical treatment invoking (linear or quadratic) discriminant analysis and aiming at the identification of the part of the compositional space occupied by granites from certain tectonic environments was seemingly more objective (Vermeesch 2006). Unfortunately, this often led to a rather abstract and mechanical use of multivariate statistics in an attempt to discriminate the geotectonic setting of igneous rocks. For acid–intermediate igneous rocks, including granitoids, numerous discriminant functions were proposed, either major- (Verma *et al.* 2012) or trace-element-based (La, Ce, Sm, Nb, Th, Y, Zr and Yb) (Verma *et al.* 2013), later implemented in the program Tec-DIA (Verma *et al.* 2015). The trace elements were chosen primarily as they are relatively immobile, but the inclusion of three light REEs (LREEs) – that are likely to give mutually comparable information – is questionable. More worrying is that, in felsic magmatic systems, abundances of trace elements would be governed by saturation and fractionation of accessories, especially zircon, allanite, monazite/xenotime, apatite and rutile (Janoušek *et al.* 2016 and references therein).

Although some rather extreme views have been expressed in the literature (most explicitly by Li *et al.* 2015 for basaltic rocks), we believe that geotectonic diagrams are not evil. They represent useful projections because melts at similar tectonic settings are likely to have formed by the same combination of sources and processes. To be successful, the real geotectonic setting of the given igneous suite needs to match one of the several simple settings predicted by the plate-tectonic paradigm. These are inevitably idealized end members, and transitional or complex settings are by no means rare (Frost *et al.* 2001).

As stressed already by Pearce *et al.* (1984), the whole-rock chemical analysis has to be precise and mimic the composition of the pure melt (i.e. not affected by excessive fractional crystallization,

crystal accumulation, assimilation, magma hybridization or restite/peritectic phase(s) entrainment). Also, the rock has to be unaltered or, at least, essentially immobile elements have to be utilized.

The most useful materials for determination of tectonic setting are typically basic lavas, as they come from primary melts (i.e. are directly derived from a relatively well-defined source, the Earth's mantle; Pearce 1996a). During differentiation, purely incompatible elements are not fractionated, such that differentiated melts largely inherit their parent's features. A powerful tool used mainly by the mantle-melting community (Wood *et al.* 1979) is the spider plot, originally designed to interpret REE patterns (Coryell *et al.* 1963). By arranging the elements in order of decreasing incompatibility relevant to the presumed petrogenesis (e.g. depleted mantle melting, forming mid-ocean ridge basalts (MORBs)) and normalizing to the putative source (the mantle), any anomaly (i.e. deviation from a smooth pattern) could be interpreted as reflecting the effect of specific minerals: for instance, Ti-oxides affecting Nb and Ta.

Unfortunately, this approach becomes less useful for crustal melts. The chemistry of crust-derived granite mimics the available magma source(s) in the (often repeatedly) recycled continental crust, rather than the true geodynamic setting (Arculus 1987; Förster *et al.* 1997; Frost *et al.* 2016). For instance, a granitic magma that had originated by the partial melting of an arc-related metasedimentary or meta-igneous rock would be likely to inherit an arc signature of a potentially much older subduction event. Regardless of the real setting, it will be characterized by a strong depletion of HFSEs relative to LILEs and thus conspicuous negative Nb-Ta anomalies, for example, in the N-MORB-normalized spider plots (Janoušek *et al.* 2010; Konopásek *et al.* 2018). In fact, such a signature is typical of the continental crust as a whole (Rudnick & Gao 2003; Taylor & McLennan 2009 and references therein) and, hence, also of anatetic melts coming from such Nb-Ta-depleted sources.

Thus, in order to interpret anomalies as reflecting the role of specific minerals, one should, in fact, normalize the chemistry of the studied granitoids to the composition of their potential source, or at least to something not too remote (for instance, the average continental crust). Ideally, one should even reorder the elements in order of decreasing incompatibility during a 'reference' crustal melting scenario, which would have to be re-ordered for any change in source – assuming that the source could be determined. This is hardly ever done, if at all.

*Diagrams plotting experimental melting data.* The interpretation of granitoid compositions in

geodynamic terms actually relies on a rather complex chain of reasoning (Pearce *et al.* 1984; Wilson 1989; Rollinson 1993; Pearce 1996b; Moyen & Laurent 2018). The chemistry of any melt reflects a complex interplay of several factors, including the modal and chemical composition of the source (as expressed, for instance, in Shaw's melting equation:  $C_L = C_0/[F + D(1 - F)]$ , where  $C_0$  and  $C_L$  are the concentrations in the source and in the melt, respectively,  $F$  is the fraction of melt formed and  $D$  is the bulk distribution coefficient) (Shaw 1970), the mechanism of melting (water-present v. dehydration), melting conditions ( $P-T$ , oxygen fugacity, etc.) and any of the subsequent differentiation processes (fractionation, assimilation, magma mixing, etc.) that affected the magma. The geotectonic links span from the empirical observation that some tectonic sites are more likely to put the right rocks in the right place. For instance, a collisional orogeny (with considerable crustal thickening) or accretionary orogeny (especially back-arc domains with considerable crustal thinning) are the most likely places to induce melting of metasediments (Collins 2002; Collins & Richards 2008; Villaros *et al.* 2018; Collins *et al.* 2019; Nabelek 2019). Thus, a geodynamic interpretation implicitly relies on two successive layers of reasoning: (i) linking a type of granite to its plausible source (and melting conditions); and (ii) linking these inferred source and melting conditions to a geodynamic site in which they were both most likely to occur.

A slightly more objective approach, therefore, is to try to link granitoid composition to its protolith (but not directly to the tectonic site of formation). This works essentially under the 'crustal melting' paradigm. Several diagrams compare the studied compositions with those of experimentally obtained granitic melts from various starting materials. They can range from simple binary plots of major-element oxides (Roberts & Clemens 1993) through major- and trace-element-based ratios (Sylvester 1998; Patiño Douce 1999; Gerdes *et al.* 2002) to more complex multicaticonic projections (Villaseca *et al.* 1998). For instance, for strongly peraluminous granites (*sensu* Sylvester 1998; i.e. with  $A/CNK > 1.1$ ), the  $\text{CaO}/\text{Na}_2\text{O}$  ratios can serve to distinguish between melts derived from clay-rich, plagioclase-poor pelitic sources and clay-poor, plagioclase-rich psammitic sources. The  $\text{Al}_2\text{O}_3/\text{TiO}_2$  ratios then represent a proxy for temperature of melting (Sylvester 1998; Jung & Pfänder 2007). A recent development, aiming to discriminate sources of granite melts, is the triangular diagram of Laurent *et al.* (2014) with apices  $\text{Al}_2\text{O}_3/(\text{FeO}^\text{T} + \text{MgO})$ ,  $3 \times \text{CaO}$  and  $5 \times \text{K}_2\text{O}/\text{Na}_2\text{O}$  (all in wt%). It is based on a compilation of a large database summarizing compositions from the experimental melting of low- or high-K mafic rocks, tonalites and metasediments.

*The synthetic classification scheme of Bernard Barbarin.* Arguably, the most comprehensive so far has been the classification of Barbarin (1999), updated from a previous scheme by the same author (Barbarin 1990). Barbarin's classification acknowledges the existence of different paradigms for granite genesis (fractionation or melting), and proposes that both apply to different rock types. For this reason, it is not much liked or used, neither by the tenants of exclusively crustal melting models nor by the proposers of pure fractionation. Although this classification scheme ultimately aims to identify the tectonic site of the formation of granites, it is largely based on observations and is relatively objective (i.e. free of tectonic or petrogenetic preconceptions or interpretations). It also includes seven distinct types, obviously covering more variations than simple bi- or tri-modal classifications, and relying on more criteria.

Synthesizing (then) current knowledge of granite classification and genetic models, Barbarin (1999) defined in total seven granitoid groups based on characteristic minerals (both main rock-forming minerals and accessories), the nature of surrounding country rocks, enclave populations and whole-rock geochemical composition. The chemical parameters comprised absolute and relative contents of  $\text{Al}_2\text{O}_3$ ,  $\text{CaO}$ ,  $\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$ , maficity, degree of Fe oxidation,  $\text{FeO}^\text{T}/(\text{FeO}^\text{T} + \text{MgO})$ , and Sr–Nd–O–S isotopic compositions. The premise was that each of these groups tends to form in an analogous geotectonic environment from comparable sources: ridge ‘tholeiitic’ granitoids (plagiogranites) (RTG), arc ‘tholeiitic’ granitoids (ATG), amphibole-rich calc-alkaline granitoids (ACG), K-rich and Kfs-phryic calc-alkaline granitoids (KCG), muscovite-bearing peraluminous granitoids (MPG), cordierite-bearing, biotite-rich peraluminous granitoids (CPG), and peralkaline and alkaline granitoids (PAG).

The mainly mantle-derived RTG and PAG were assumed to be products of extensive fractionation of tholeiitic melts at ocean ridges or alkali basalt melts in continental doming/rifting settings, respectively. Typical of island arcs are mantle-derived ATG with ACG of presumably mixed crust–mantle origin; the ACG are also most characteristic of continental subduction settings, including the huge Cordilleran batholiths. The previously distinguished peraluminous CPG and MPG (Barbarin 1996) are generated by crustal anatexis in continental collision or early post-collisional settings. Apart from that, the former collision belts (type area being the Scottish Caledonides) often feature voluminous post-collision uplift-related KCG plutons. In principle, CPG (and partly MPG) overlap with S-types, PAG match the A-types, and the other groups are all I-types despite their contrasting features, origins and occurrences.

*Future avenues to improve the genetic and geotectonic classifications?* Trace-element contents and ratios in granitic melts give insights into sources and differentiation processes, which modal and major-element classifications do not (for reviews, see Rollinson 1993; Shaw 2006; Zou 2007; Janoušek *et al.* 2016; Janoušek & Moyen 2019). In particular, the trace and minor elements (such as  $\text{P}_2\text{O}_5$ ) become handy in studies of highly fractionated granites, whose major-element contents change only a little when approaching/reaching the haplogranite composition (e.g. Tuttle & Bowen 1958; Chappell 1999; Frost *et al.* 2016).

For classifications, as well as to study differentiation, trace elements entering the main rock-forming minerals are particularly useful, especially the mica- and feldspar-compatible LILEs (Ba, Rb and Sr) (Hanson 1978). However, the elements incompatible with major phases are unlikely to add more information to any descriptive granite classification, as the majority of them (U, Th, REEs, HFSEs) are stored in accessory minerals. Nevertheless, these elements can be useful in deciphering the geotectonic setting of igneous suites, with the added advantage that they are less mobile than the LILEs (Floyd & Winchester 1978; Pearce 1996a). Increasingly important nowadays have become the trace-element characteristics of the main rock-forming minerals, determined by *in situ* techniques such as laser-ablation inductively-coupled plasma mass spectrometry (LA-ICP-MS), and ion or electron microprobe. The mineral chemistries can be used to constrain the evolution of the magma that they crystallized from, as well as the corresponding  $P$ – $T$  conditions (e.g. Anderson *et al.* 2008; Barnes *et al.* 2017).

Also, isotopic (stable and radiogenic) data offer invaluable additional information. As the radiogenic isotopic ratios are completely transparent to mechanisms of closed-system magmatic differentiation, they can portray the granite sources. Alternatively, they may disclose the operation of open-system processes, such as magma mixing, assimilation or hydrothermal alteration, and help to further parameterize them (Faure & Mensing 2004; Dickin 2005). Fresh insight into the sources and petrogenesis of the granitic magmas is brought by non-traditional isotopic systems (Johnson *et al.* 2004; Teng *et al.* 2017).

## Some problems with classic terms used in classifications

Granite classification is not only an academic exercise but should provide an accepted basis for scientific interpretations and debates. The problem is that the scientific community cannot agree on exact meanings even of the simple terms currently

used in the literature, such as alkaline–subalkaline (Iddings 1895), felsic–mafic (Cross *et al.* 1902), metaluminous–peraluminous–peralkaline (Shand 1927) and calcic–calc-alkaline–alkali-calcic–alkaline (Peacock 1931) groupings (for a short review, see Barker 1981).

### *Felsic–mafic, acid–basic, differentiated–undifferentiated*

Although these pairs of terms are often used interchangeably, they actually refer to different concepts. A ‘felsic’ rock contains mostly light-coloured minerals, whereas an ‘acid’ rock is  $\text{SiO}_2$ -rich, and conversely for ‘mafic’ and ‘basic’. The words ‘mafic’ and ‘felsic’ are essentially abbreviations, introduced by Cross *et al.* (1902) and referring to high contents of dark (Mg–Fe-rich) and light minerals (feldspars, feldspathoids and quartz). This can be quantified, for instance, by using the sum of the modal or normative proportions of light-coloured minerals (e.g. the CIPW-normative differentiation index of Thornton & Tuttle 1960). More recently, Debon & Le Fort (1983, 1988) used parameter  $B = \text{Fe} + \text{Mg} + \text{Ti}$  (in millications) to the same effect; a variant of the same parameter has been nicknamed ‘maficity’ by, for example, Clemens *et al.* (2011).

The terms ‘acid’, ‘intermediate’ and ‘basic’, on the other hand, have well-established definitions with  $\text{SiO}_2$  cutoff values of 52 and 63 wt% (Le Maitre 2002). Therefore, a syenite is felsic but it is not acid.

The term ‘differentiated’ is meaningful in the context of planetary evolution (i.e. the processes that form rocks with compositions strongly differing from the Bulk Earth = chondritic ones) or in the context of differentiation of a magmatic series. In the general case, however, it is not a synonym for ‘acid’ or ‘felsic’. Consider, for instance, the case of sediments melting to form peraluminous granites: writing that a ‘leucogranite is more differentiated than the granite and therefore corresponds to lower temperature melting (lower  $F$ ) of the same source’ is an oxymoron.

Of course, during the course of fractionation from a common parental magma, the more differentiated melts tend to become both acid and felsic. Thus, the proportion of  $\text{SiO}_2$ , the maficity or the differentiation index can all, in this context, serve as a proxy for differentiation within a suite – assuming that this was the process shaping its composition.

### *Alkaline–subalkaline*

The terms are spelled variously: alkali, alkaline or alkalic and subalkali, subalkaline or subalkalitic. The ‘alkali–subalkali’ pair seems to be used preferably before the name of a rock. The terms alkaline and subalkaline date back to the end of the nineteenth

century: defined by Iddings (1895), the concept was discussed by Harker (1909). It refers to rocks that, having the same silica content, contain contrasting – higher and lower, respectively – alkali oxide contents. This distinction was developed in terms of igneous series by volcanologists since the influential paper of Macdonald & Katsura (1964) on Hawaiian basalts. The classical boundary between subalkaline and alkaline basalts (for alternatives, see Rickwood 1989; Rollinson 1993) was extended to felsic rocks, including the alkaline silica-oversaturated ones of the Coombs trend (Miyashiro 1978).

A more restrictive definition was set out by Shand (1927). Considering molar proportions of  $(\text{Na}_2\text{O} + \text{K}_2\text{O}) : \text{Al}_2\text{O}_3 : \text{SiO}_2$ , he stated that, in an alkaline rock, the alkalis are in excess of 1:1.6 – that is, either silica or alumina, or both, being deficient. In this conception, alkaline rocks bring together all silica-undersaturated and peralkaline (having molar  $\text{Al}_2\text{O}_3 < (\text{Na}_2\text{O} + \text{K}_2\text{O})$ ) silica-saturated to oversaturated rocks. Contrary to Miyashiro (1978), the definition excludes non-peralkaline granites, even if they are closely related to peralkaline granites (e.g. see Bonin & Giret 1984; Bonin 2007). Incidentally, in the silica–MALI diagram (Frost *et al.* 2001), syenites and peralkaline granites plot in the alkaline field, and other alkaline silica-oversaturated rocks (including the metaluminous and peraluminous granites) plot in the alkali-calcic field, whereas the subalkaline rocks fall in the calc-alkaline and calcic fields.

In the European community, ‘subalkaline’ became synonymous with ‘nearly alkaline’. As this meaning is in variance with the original definitions of Iddings and Harker, it should be abandoned. ‘Subalkaline’ should stand for rocks plotting below the alkaline–subalkaline boundary line defined by Miyashiro (1978) in the TAS diagram. Although popular, the Irvine & Baragar (1971) dividing line is irrelevant, as it separates silica-undersaturated and silica-saturated rocks within the alkaline field. To cope with the incorrect use of ‘subalkaline’, Lameyre *et al.* (1982) suggested the term ‘transalkaline’, while Middlemost (1991) defined ‘transalkali’ suites, i.e. suites of rocks plotting along the alkaline–subalkaline boundary in the TAS diagram.

### *Misuses of the term ‘calc-alkaline’*

In his classification of igneous rocks, Peacock (1931) introduced this term, together with alkaline, alkali-calcic and calcic. Noting that  $\text{CaO}$  is negatively and  $(\text{Na}_2\text{O} + \text{K}_2\text{O})$  are positively correlated with  $\text{SiO}_2$  (if all are presented in wt%), he defined the ‘alkali–lime index’, which is the critical  $\text{SiO}_2$  value for a given igneous suite in which the best-fit curves  $\text{SiO}_2$  v.  $\text{CaO}$  and  $\text{SiO}_2$  v.  $(\text{Na}_2\text{O} + \text{K}_2\text{O})$  intersected. Differing alkali–lime indices of magma series are

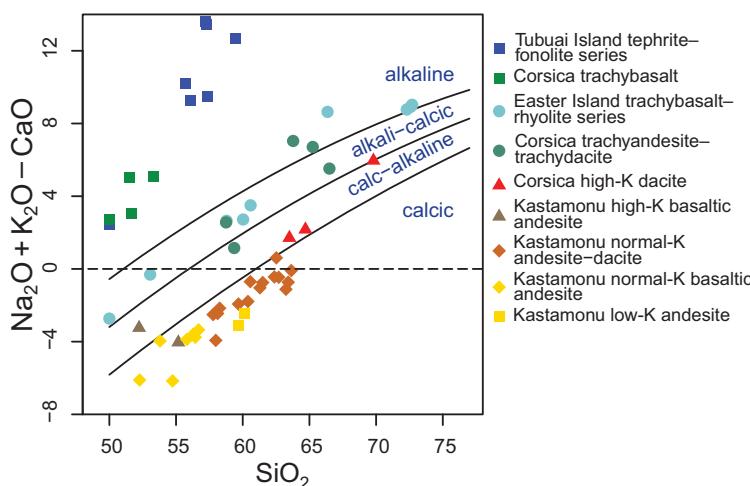
related more to changes in alkalis than in CaO, because the CaO curves of most series are roughly similar but the alkali contents and alkali curves may be highly variable.

The ensemble of alkali–lime indices was subdivided by three thresholds: 51, 56 and 61 wt%, thus defining four igneous rock, or magma, series: alkaline (alkali–lime index <51 wt%), alkali-calcic (alkali–lime index between 51 and 56 wt%), calc-alkaline (alkali–lime index between 56 and 61 wt%) and calcic (alkali–lime index >61 wt%).

Unfortunately, the meaning of the term evolved over time. The first change took place when calc-alkaline and tholeiitic suites were defined as subalkaline suites differing by the absence and presence of iron enrichment, respectively (Nockolds & Allen 1953; Irvine & Baragar 1971; Miyashiro 1974). Later on, ‘calc-alkaline’ became roughly synonymous with ‘orogenic subalkaline’, while ‘tholeiitic’ was given an ambiguous status of low-K as well as high-Fe subalkaline rocks, whether orogenic or not. Such a shift is exemplified by the influential Peccerillo & Taylor (1976) SiO<sub>2</sub> v. K<sub>2</sub>O diagram, depicting low-K tholeiitic, medium-K calc-alkaline and high-K calc-alkaline suites, besides shoshonitic and potassic suites. In the silica–MALI diagram (Fig. 7), all tholeiitic and calc-alkaline volcanic suites of the Kastamonu area, northern Turkey, plot in the calcic field and are, therefore, not in this sense calc-alkaline at all.

The term is currently used for a variety of geochemical characteristics that are not necessarily correlated (e.g. the absence of iron enrichment), and typically have no relationship with ‘calc’ or ‘alkaline’ character (Sheth *et al.* 2002). Moreover, given the association in many geologists’ minds of calc-alkaline suites with active or ancient subduction zones, the former existence of subduction-related environments is frequently, and often erroneously, asserted. The unjustified speculations about plate tectonics on Mars, based on andesitic composition of Pathfinder rocks (McSween *et al.* 1999), constitute a typical example. This is also the case for Archean magmatism (for a discussion, see Moyen & Laurent 2018).

As stressed by Frost *et al.* (2001, p. 2044), ‘the term “calc-alkalic” (should be) used in the manner that it was originally defined – to describe suites of rocks that show moderate enrichment in calcium over the alkalis’. Thus, many suites that are presented in the current literature as calc-alkaline vary from calcic to alkali-calcic. Arculus (2003, p. 934) pointed out that ‘the application of the terms calc-alkaline and calcalkalic is plagued with inconsistency, with the terms having different meanings for different users’. He showed that a majority of the so-called calc-alkaline suites is actually ‘calcic’ and, therefore, that the common usage of ‘calc-alkaline’ for all orogenic subalkaline rocks is misleading.



**Fig. 7.** SiO<sub>2</sub>–MALI (wt%) binary diagram (Frost *et al.* 2001) for some volcanic series. Data sources: Neogene Tubuai alkaline suite (Brousse & Maury 1980); Permian Corsica trachybasalts (Cabanis *et al.* 1990); Easter Island trachybasalt–rhyolite (Baker *et al.* 1974); Permian trachyanandesite–trachydacite and Carboniferous–Permian high-K dacite suites from Corsica (Rossi *et al.* 1993); Eocene Kastamonu low-K (‘tholeiitic’); normal-K calc-alkaline and high-K calc-alkaline suites (Peccerillo & Taylor 1976). The first three suites were considered by the authors as alkaline, and the last six groups as calc-alkaline.

Consequently, according to Le Maitre's (2002) remark, the widely used  $\text{SiO}_2\text{-K}_2\text{O}$  diagram should be employed without adding the term 'calc-alkaline' to the names of the low-K, medium-K and high-K fields. The commonly accepted, or tolerated, use of loose definition of the adjective 'calc-alkaline' results in a net loss of information and can lead to erroneous conclusions. Thus, 'calc-alkaline' should be reserved for suites conforming to the original Peacock (1931) definition.

The  $\text{SiO}_2\text{-MALI}$  plot (Frost *et al.* 2001) constitutes currently the best tool. In order to understand its geometry and limits, within which ordinary granitoids should fall, a modal QAP diagram is projected (Fig. 1b). The P vertex transforms into a long segment and the A vertex into a small triangle. Finally, it should be noted that both TAS and  $\text{SiO}_2\text{-MALI}$  diagrams (Fig. 1a, b) lay emphasis on plagioclase compositions and abundances. Transcribing the QAP diagram within these plots is clearly not a straightforward task and neither was it the purpose of the original Frost *et al.* (2001) classification.

## Re-examining the composition of granitoids

In this section, we take a more quantitative view. Instead of describing existing classification schemes, we examine the composition of a large global database of granitoids to establish the main chemical differences between possible types, and see how the various classifications depict these differences.

### Statistical tools

*Designing the 'best' diagram.* The art of building a useful diagram amounts to finding the appropriate projection, one that best represents the features to be studied. Critically assessing the existing diagrams, we can conclude that they serve two main purposes. They aim to:

- (1) Depict the evolution of a property as a function of differentiation. This is, for instance, the purpose of Harker-type plots involving  $\text{SiO}_2$  as the differentiation index (Harker 1909) but also of diagrams using  $\text{FeO}^\text{T}$  and/or  $\text{MgO}$  (Fenner-type), or Debon & Le Fort's multicationic diagrams invoking the B parameter as the x-axis. Such a diagram will spread the compositions generated by the differentiation process: that is, we project onto a plane that contains the differentiation vector.
- (2) Emphasize the differences between individual igneous associations, irrespective of their internal evolution/degree of differentiation. A diagram of this type should be, as much as possible, projected onto a plane orthogonal

to the differentiation vector. In this way, all rocks of a given suite should cluster together, such that the remaining differences can be ascribed to different series. This is, practically, very difficult to achieve as: (i) the geochemical properties of igneous rocks are almost all correlated to differentiation; and (ii) the differentiation trends are often not straight, if only due to changing solid assemblages during fractional crystallization.

Interestingly, these two representations largely overlap with the two main paradigms for granite formation: differentiation of mafic melts or melting of distinct crustal lithologies. In a differentiation paradigm, chemical differences are viewed chiefly as the result of the differentiation process. In a crustal melting model in contrast, dissimilar chemistries of individual rock types are viewed as mostly resulting from source differences.

*Statistical approach to projection.* In addition to the empirical approach presented above, many statistical tools exist to deal with an  $n$ -dimension population of observations. The whole-rock major-element analyses, like any other compositional data, suffer from the so-called closure (or constant sum) effect (Chayes 1960; Skala 1979; Aitchison 1986; Rollinson 1992; Pawlowsky-Glahn & Egozcue 2006). As a consequence, spurious correlations between various oxides are induced by the fact that all oxides must sum up to 100 wt% and the silica content accounts for as much as half to three-quarters of each analysis. For all major-element oxides, absolute values are, hence, strongly correlated to silica. At the same time, genuine petrological evolutions are superimposed on the correlation caused by the closure effect (Janoušek *et al.* 2016). Therefore, composite major-element-based indicators showing the relative importance of each component are more useful than the raw weight percentages.

The closure effect makes the statistical tools based on the distance between two points of the compositional space difficult to use. Statistical solutions to this problem are relatively well known, the most common involving the 'log-ratio transformation' (Buccianti *et al.* 2006 and references therein). They are not in very common use in the petrological literature, however. This is probably for a range of reasons ranging from the more mundane (the use of such transformations requires an extra step of calculation) to the more fundamental (in a log-ratio diagram, phase-diagram properties such as lever rule are lost), and the closure effect is probably not critical when dealing with a restricted set of compositions (one rock type).

Statistical tools highlighting the differences between samples or their groups include, for

instance, hierarchical clustering, principal component analysis or linear discriminant analysis. Strictly speaking, all these approaches rely on calculating the distance between samples (in compositional space) and thus should not be used in a closed compositional space (i.e. transformations such as log-ratio should be applied beforehand). Hierarchical clustering (Reimann *et al.* 2008) (*k*-means clustering being probably the most common algorithm) is used to identify ‘clusters’ of points with similar properties in the *n*-dimensional space. Principal component analysis (PCA) is also a commonly employed tool that aims to find the variables that carry most variance (i.e. the ‘angle’ from which one should view (project) the dataset to spread it as much as possible on the diagram plane). Applied to igneous rocks, both methods suffer from the same issue: most of the variance (i.e. most of the differences between two rocks) is related to differentiation. In geological terms, this corresponds to the obvious conclusion that a given granite is more similar to any other granite than to a diorite (even if the diorite corresponds to a member of the same suite); a fully predictable, if rather useless conclusion.

A more promising approach is linear discriminant analysis (LDA), a method that allows one to find the projection that best separates several predefined classes of samples (McLachlan 2004). It is related to, although somewhat different from, PCA in that PCA attempts to maximize the *overall spread* of an unclassified dataset, whereas LDA emphasizes the *differences* between predefined classes. A number of new variables, equal to the number of original classes, are calculated from the first one to the last one, and they carry less and less information (i.e. they are less useful in separating the classes). As a by-product, the LDA also yields scores representing the probability that an unknown sample belongs to each of the known classes. In our case, this can be used to find the projection that best shows the difference between distinct types of granitoids.

### Database used

Our database of granitoid compositions includes more than 3000 analyses compiled from: (i) our own data, both published and unpublished; (ii) the GeoRoc database (<http://georoc.mpch-mainz.gwdg.de/georoc>); and (iii) other publications (see ESM 1 in the Supplementary material for the references). The database is built at the pluton level: that is, all of the samples from a single pluton (or a single phase of a composite pluton) are classified as one type. Furthermore, we kept only those plutons for which a reasonable number (>10 typically) of analyses is available. This allows us to focus on the granitic series more than on individual sample compositions. This means also that we do not

consider much differentiation – we instead aim to identify different series, each including members with different degrees of differentiation. Finally, the database is cleaned by keeping only intermediate and acid samples ( $\text{SiO}_2 > 52\%$ ), and by excluding analyses with  $\text{SiO}_2 > 80\%$ , as well as those with a poor total (<98% or >102%). The final dataset includes about 2500 samples, and we face a conundrum here. As noted, granites of all sorts converge to the same restricted range of compositions and do not show large differences – but less differentiated members of their suites are more contrasted. Thus, classification works better on whole suites including intermediate components that are, of course, not granitoids. We adopted a compromise strategy, in keeping the whole database but plotting samples with  $\text{SiO}_2 < 62\%$  with a different symbol (empty v. full symbols, to make them less visible). The full database is supplied as plain-text (TXT) and Excel (XLS) files in ESM 1 in the Supplementary material. R/GCDkit (Janoušek *et al.* 2006) scripts used to plot all the following diagrams are given as ESM 2 in the Supplementary material.

Granitoids from the database are initially classified in the following types and subtypes, modified from Barbarin (1999). The classification follows Barbarin’s logic: it is chiefly based on petrological considerations (i.e. rock type, key minerals, etc.), sometimes taking the geological context into account. Subtypes within Barbarin’s types are defined using chemical indicators: that is, on the basis of the position of different suites in the diagrams used in this paper. A shortcoming of our database is that the proportion in which different types are represented does not match their abundance on Earth. No effort has been made to estimate actual proportions, which are changeable over geological time. Instead, we focus here on identifying and describing distinct types, defined in the forthcoming subsections.

Although omissions are unavoidable, we trust that our database includes representatives of most important granitic types on Earth. It contains, for instance, samples from the Lachlan Fold Belt of Australia (CPG and ACG), from Japan and western North America (mostly ACG), from the European Variscan belt (CPG, MPG and KCG), and the Caledonides (KCG). It includes also the original database of Eby (1992) (PAG ( $A_1$ ) and KCG ( $A_2$ )), some Proterozoic granites from Brazil (KCG), cordierite-bearing granites from the Cape Fold Belt of South Africa (CPG) and Archean TTGs from Barberton.

*Ridge tholeiitic granites (RTG).* The RTG are associated either with ophiolites/oceanic crust or with basaltic plateaux. It is an uncommon type (<1% of the database). They overlap with the OP (oceanic plagiogranites) of Maniar & Piccoli (1989) or the

ORG (ocean ridge granites) of Pearce *et al.* (1984), and may correspond to (some) A-types in the S–I–M–A classification.

*Amphibole-rich calc-alkaline granitoids (ACG) and arc ‘tholeiitic’ granitoids (ATG).* The ACG and ATG (32% of the database) are mostly found at active continental margins (Andes, Japan, etc.), where they form the classic Cordilleran batholiths, as well as in somewhat more ambiguous settings (the Lachlan Fold Belt). They are I-types included in both Maniar & Piccoli’s (1989) island-arc granitoids and continental-arc granitoids (IAG and CAG), and are equivalent to Pearce *et al.*’s (1984) volcanic-arc granites (VAG). Several chemical subgroups can be identified: (i) ‘ordinary’ ACG; (ii) relatively alkaline ACG; (iii) tholeiitic ATG; and (iv) relatively alumina-rich ACG. The last group is mildly peraluminous and overlaps with the CPG/MPG; its members correspond, at least in part, to what has been sometimes referred to as ‘transitional S–I type’.

*Cordierite-bearing peraluminous granitoids (CPG) and muscovite-bearing peraluminous granitoids (MPG) (i.e. collisional to post-collisional strongly peraluminous granites).* This group comprises Barbarin’s CPG and MPG (22% of the database). The CPG are the original S-types as defined in the Lachlan Fold Belt, while the MPG are more leucocratic, close to the granite minimum and can therefore occur as differentiated members in all sorts of plutons. In any case, CPG and MPG subtypes are clearly distinct chemically (Barbarin 1996). Both collectively correspond to Pearce *et al.*’s (1984) syn-collisional granites (syn-COLG) and Maniar & Piccoli’s (1989) continental-collision granitoids (CCG).

*K-rich calc-alkaline granitoids (KCG).* The KCG (20% of the database) is, perhaps, the worst-defined group. Maniar & Piccoli’s (1989) continental epeirogenic uplift granitoids (CEUG) are probably the closest match. The KCG are not identified in the original S–I–M–A classification (they do not occur in the Lachlan Fold Belt), and are regarded as I-types when this classification is used because they are not peraluminous. On the other hand, given that KCG are highly potassic (and have a high alkali content in general), some have been described as A-types (‘orogenic A-types’ and ‘oxidized A-types’). To clarify (?) this issue, Eby (1992) proposed a subdivision of A-types into A<sub>1</sub> and A<sub>2</sub>, the latter corresponding to (part of) the KCG. The KCG are also missing entirely from Pearce *et al.*’s (1984) classification.

To further complicate matters, KCG encompass a large range of compositions, occurring mostly in post-collisional to post-orogenic settings (Bonin 1990, 2004). Although they are generally

metaluminous to weakly peraluminous and alkaline or nearly so, we identified several chemical variants: (i) the high-K suite, generally subalkaline and magnesian, associated with mafic rocks (‘vaugnerites’, ‘appinites’ and ‘redwitzites’: von Raumer *et al.* 2014; Murphy 2019), essentially similar to Archean sanukitoids, sometimes described as ‘high Ba–Sr’ granitoids (Fowler *et al.* 2008; Fowler & Rollinson 2012); (ii) the ultra-K suite, alkaline and strongly magnesian (‘durbachites’ of the European Variscides: Janoušek & Holub 2007); (iii) the ‘A<sub>2</sub>’ suite, also alkaline but usually ferroan, and barely peraluminous; and (iv) the geologically related syenites chemically rather unique, although always associated with A<sub>2</sub>. Both high-K and ultra-K suites are post-collisional; the A<sub>2</sub> and their syenites are, in general, post-orogenic.

*Peralkaline and alkaline granitoids (PAG).* Peralkaline and alkaline granites (15%) are typically rift- or hotspot-related. They are, of course, A-types, specifically corresponding to Eby’s (1992) A<sub>1</sub> type, and can be equated to Pearce *et al.*’s (1984) within-plate granites (WPG) and Maniar & Piccoli’s (1989) rift-related granitoids (RRG). They are chemically unique (strongly alkaline and ferroan, metaluminous to peralkaline) and often feature an unusual modal composition with sodic mafic silicates (aegirine, riebeckite). Here they are grouped with the typically coeval (nepheline) syenites that turn out to be clearly distinct from the ‘A<sub>2</sub>’ syenites mentioned previously.

*Archean tonalites, trondhjemites and granodiorites (TTG).* Finally, it turns out that TTGs (10%), which are not part of the Barbarin (1999) scheme, define a type of their own clearly distinct from any other granitoid. Following, for example, Moyen (2011, 2019a), one may separate a more calcic, ‘tonalitic’ series (=LP TTGs) and a sodic, ‘trondhjemitic’ series (=HP TTGs).

### Some general considerations

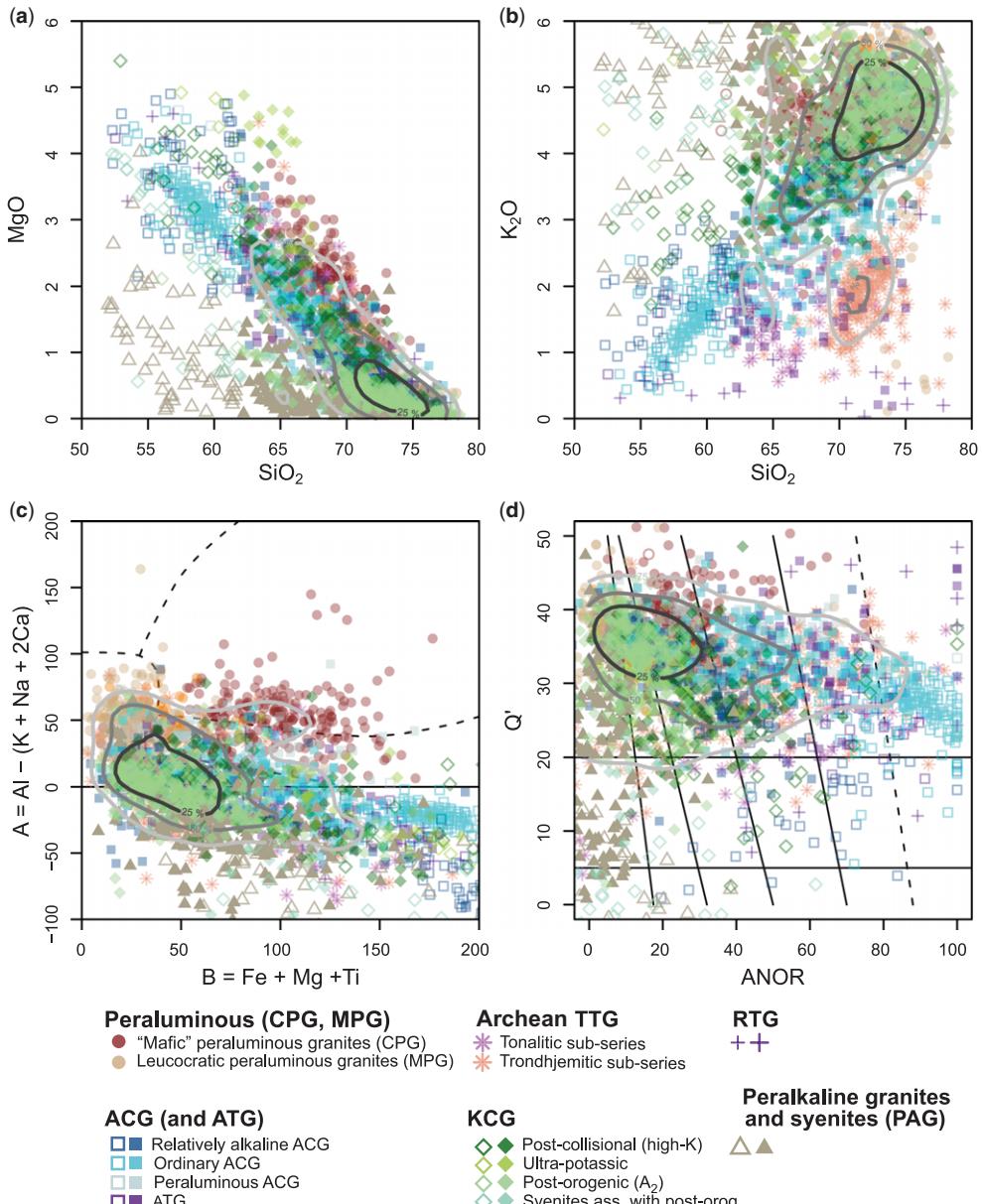
Before embarking into more detailed treatment of this dataset, we can make some preliminary remarks.

*Useful and not so useful diagrams.* A first observation is that not all diagrams are suitable for the purpose of granitoid discrimination. Figure 8 shows the whole database, plotted in two Harker plots and two standard diagrams: one depicting aluminosity, and the other emphasizing the Ca–Na–K (feldspar) and quartz systematics.

Whereas granites of all types are much better separated in the latter two diagrams, they overlap in the Harker plots. This is worse for Harker plots using compatible elements (such as MgO), where the excellent negative correlation (boosted by the

closure effect) between the two oxides means that the whole dataset plots along a tightly constrained array. This applies more or less to every granitoid series on Earth, and such a diagram is therefore of very little

use. Harker plots are differentiation diagrams (and they have been devised by Harker (1909) for this specific purpose): that is, they are meant to show the role of successive minerals that crystallize from



**Fig. 8.** The whole database used in this paper, projected in two Harker-type plots ( $MgO$  and  $K_2O$ : a & b) and two arguably more useful projections, B–A of Debon & Le Fort (1983, 1988) (c) and  $Q'$ –ANOR of Streckeisen & Le Maitre (1979), based on the improved granite mesonorm of Mielke & Winkler (1979) (d). The dark, medium and light grey lines on each diagram include 25, 50 and 75%, respectively, of the granitoids in the database ( $SiO_2 > 62\%$ ), showing that most of the granitoid compositions, irrespective of their types, are actually very similar, corresponding to the ‘granite minimum’.

the magma as it evolves. However, for distinguishing distinct magmatic series (and discussing different sources), they are not very useful. More relevant are projections that try to avoid, or minimize, the differentiation component, and in which individual series define distinct trends.

*The granite minimum.* Secondly, the composition of all granitoids converges into a narrow portion of the diagram, in most projections. In Figure 8, the fields containing 25, 50 and 75% of the acid samples ( $\text{SiO}_2 > 62 \text{ wt\%}$ ) from the database are contoured: they define a composition with  $\text{SiO}_2 \geq 70 \text{ wt\%}$ , subaluminous to mildly peraluminous and  $\text{K}_2\text{O} c. 4\text{--}5 \text{ wt\%}$ . Corresponding rocks are ‘ordinary’ granites, with quartz, K-feldspar, a relatively sodic plagioclase in subordinate amount (about one-fifth to one-third of the total feldspar), biotite as the main mafic silicate and, possibly, some muscovite. In petrological terms, this composition corresponds to the thermal minimum (i.e. either the last liquid formed by fractionation or the first liquid produced by melting). For classification purposes, it is noteworthy that this compositional region contains granitoids of all types and subtypes. This means that in terms of major-element composition, most of the granites (*sensu stricto*) worldwide are identical: they could belong to any association.

More diagnostic, on the other hand, are: (i) the less common compositions, more mafic in general, associated with the granites; and (ii) the whole differentiation trend and the evolution of selected properties. Here, we emphasize that the composition of an individual sample (or even of a small group of similar samples) may not be diagnostic of any association.

#### Statistical approach to granitic types

We performed LDA on our dataset, with the aim of finding the projections that best separate the six

groups previously defined (performing LDA on the subgroups does not significantly change the results). Specifically, the variables were log-centred (Aitchison 1986; Pawlowsky-Glahn & Egozcue 2006) before performing the LDA, and the sign of each LD component was changed if needed to ensure that  $\text{SiO}_2$  always had a positive weight. Table 1 shows the resulting factors, and Figure 9 shows plots of  $\text{LD}_1$  v.  $\text{LD}_2$  and  $\text{LD}_1$  v.  $\text{LD}_3$  (as these diagrams are 2D projections of the underlying 3D space, an animated 3D version is provided as ESM 3 in the Supplementary material).

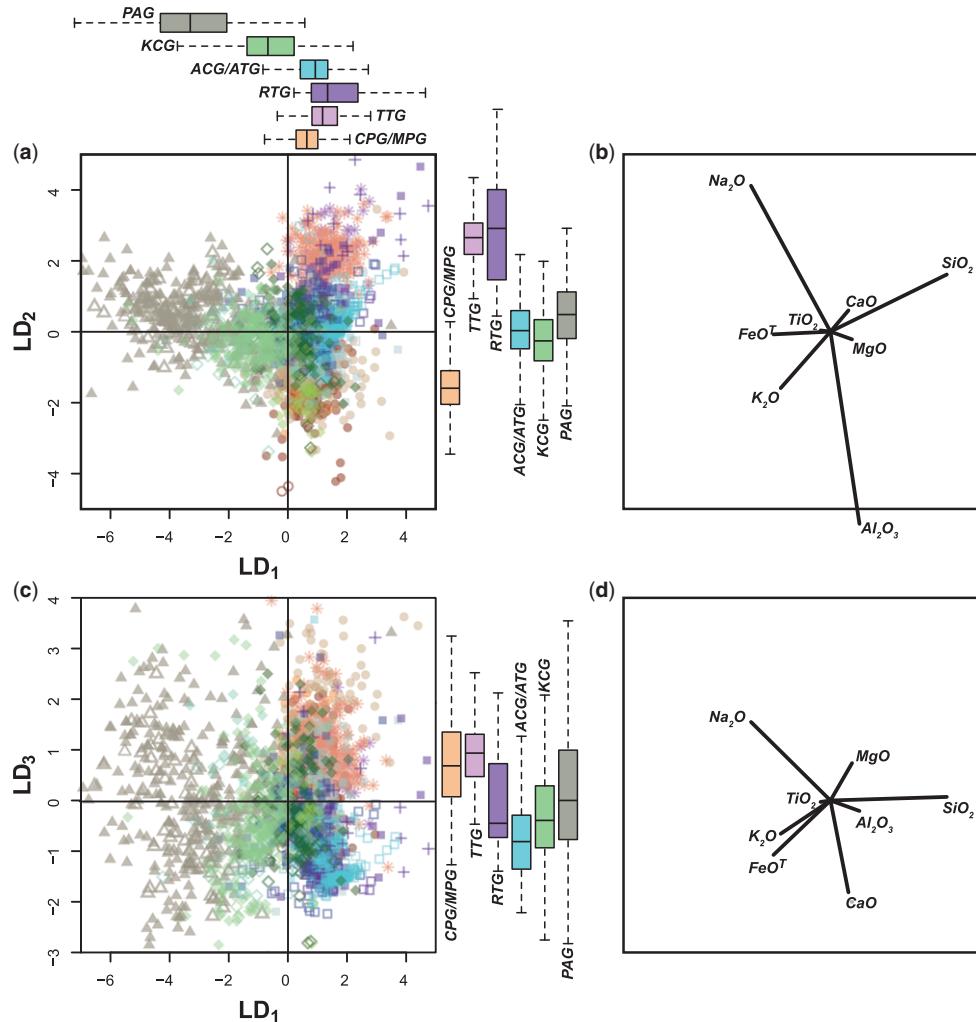
An immediate conclusion is that the LDA efficiently separates our provisional types, with both  $\text{LD}_1$ - $\text{LD}_2$  and  $\text{LD}_1$ - $\text{LD}_3$  diagrams yielding Y-shaped arrays (in 3D, they join apices of a tetrahedron). One of the apices (low  $\text{LD}_1$ ) define the PAG; the second one (high  $\text{LD}_2$  and high  $\text{LD}_1$ ) represents TTG (and RTG); the third one (low  $\text{LD}_2$ ) is occupied by CPG and MPG; and the last one, less marked on the low  $\text{LD}_3$  side, corresponds to ACG. Even in this space, a large proportion of granitoids occupies the central portion (the granite minimum), and in this region it is difficult to decide on the series to which an individual sample belongs.

In more detail, the main LD parameters can be related to conventional petrogenetic indicators.  $\text{LD}_1$  (53% of the variance) is positively linked to  $\text{SiO}_2$ , and negatively to  $\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$ : thus, it separates low-Si, high-alkali from high-Si, low-alkali compositions (i.e. alkaline from subalkaline series). This is demonstrated in Figure 10a–b, which depicts the database with samples colour-coded according to their  $\text{LD}_1$  values. They very clearly separate individual magmatic series in the mesonormative Q’-ANOR plot (Streckeisen & Le Maitre 1979) (cf. Fig. 8d) or the P-Q diagram (Debon & Le Fort 1988) (Fig. 10b). In contrast, the degree of differentiation is not at all, or only slightly, reflected in  $\text{LD}_1$ : samples of similar  $\text{LD}_1$  values plot all along differentiation trends (e.g. Fig. 10a).

**Table 1.** Results of the linear discriminant analysis (LDA)

	LD <sub>1</sub>	LD <sub>2</sub>	LD <sub>3</sub>	LD <sub>4</sub>	LD <sub>5</sub>
$\text{SiO}_2$	<b>3.93</b>	1.61	0.07	1.23	5.85
$\text{TiO}_2$	-0.34	0.03	-0.03	-0.62	-0.57
$\text{Al}_2\text{O}_3$	0.98	<b>-5.42</b>	-0.21	-7.69	-7.23
$\text{FeO}^T$	<b>-1.93</b>	-0.08	-1.07	-1.73	0.90
$\text{MgO}$	0.73	-0.22	0.74	-0.15	0.36
$\text{CaO}$	0.61	0.60	<b>-1.81</b>	0.80	-0.95
$\text{K}_2\text{O}$	<b>-1.68</b>	-1.59	-0.66	1.19	-0.92
$\text{Na}_2\text{O}$	<b>-2.69</b>	<b>4.11</b>	1.55	1.92	-0.34
Proportion of trace	53.59%	32.59%	9.83%	3.36%	0.64%

Each of the LD factors is a combination of the oxides, weighted by certain amounts as indicated in the table. The last row summarizes the proportion of the total variance that is explained by each LD: more than 95 % of the total variance is linked to the first three components. The most important contributors are in bold (see the discussion in the text and Fig. 9).



**Fig. 9.** (a, c) LD<sub>1</sub>-LD<sub>2</sub> and (b, d) LD<sub>1</sub>-LD<sub>3</sub> diagrams. Symbols as in Figure 8, comments in the text. The box and whiskers subplots (a) and (b) show for each group the median (thick line), the interquartile range (box) and the total range without outliers (whiskers). (b) & (d) portray the direction and magnitude of the vectors corresponding to individual oxides (see Table 1).

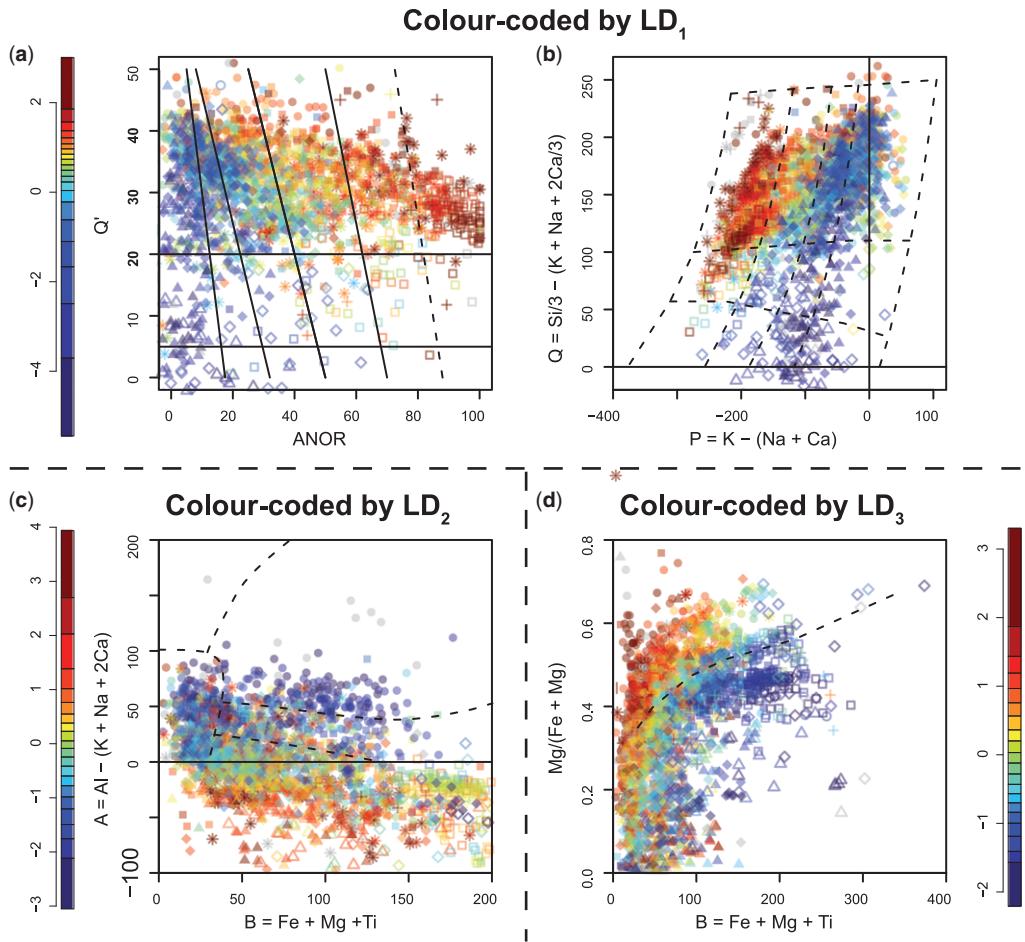
Likewise, LD<sub>2</sub> (32% of variance) is strongly controlled by Al<sub>2</sub>O<sub>3</sub>. Plotting a B-A diagram (Debon & Le Fort 1988), colour-coded by LD<sub>2</sub> (Fig. 10c), shows that different series in this diagram correspond to different LD<sub>2</sub> values. Again, the degree of differentiation is not, or poorly, reflected in LD<sub>2</sub>: LDA allows identification of different groups, irrespective of the degree of differentiation.

Finally, LD<sub>3</sub> has coefficients of opposite signs for FeO<sup>T</sup> and MgO, and Figure 10d confirms that trends of different Mg# correspond to contrasting LD<sub>3</sub> values, irrespective of the degree of differentiation.

This statistical approach confirms not only the efficiency of the LDA method but also the usefulness of Barbarin's (1999) classification scheme and the empirical findings of generations of petrologists: the main chemical parameters that allow distinction between granitic series are the feldspar balance (Ca-Na-K), aluminosity and Mg#.

#### Chemical characteristics of main granitoid types

Each of the six *a priori* types has a consistent set of geochemical features, as depicted in Figures 11–14



**Fig. 10.** Examples of classic classification diagrams colour-coded by LD values. (a) Mesonormative Q'-ANOR diagram (Streckeisen & Le Maître 1979). (b) P-Q plot (Debon & Le Fort 1983, 1988). (c) B-A plot (Debon & Le Fort 1983, 1988). (d) B-Mg# plot of Debon & Le Fort (1988). The points retain their symbols (as in Fig. 8) but are colour-coded by LD values: LD<sub>1</sub> in (a) & (b), LD<sub>2</sub> in (c) and LD<sub>3</sub> in (d).

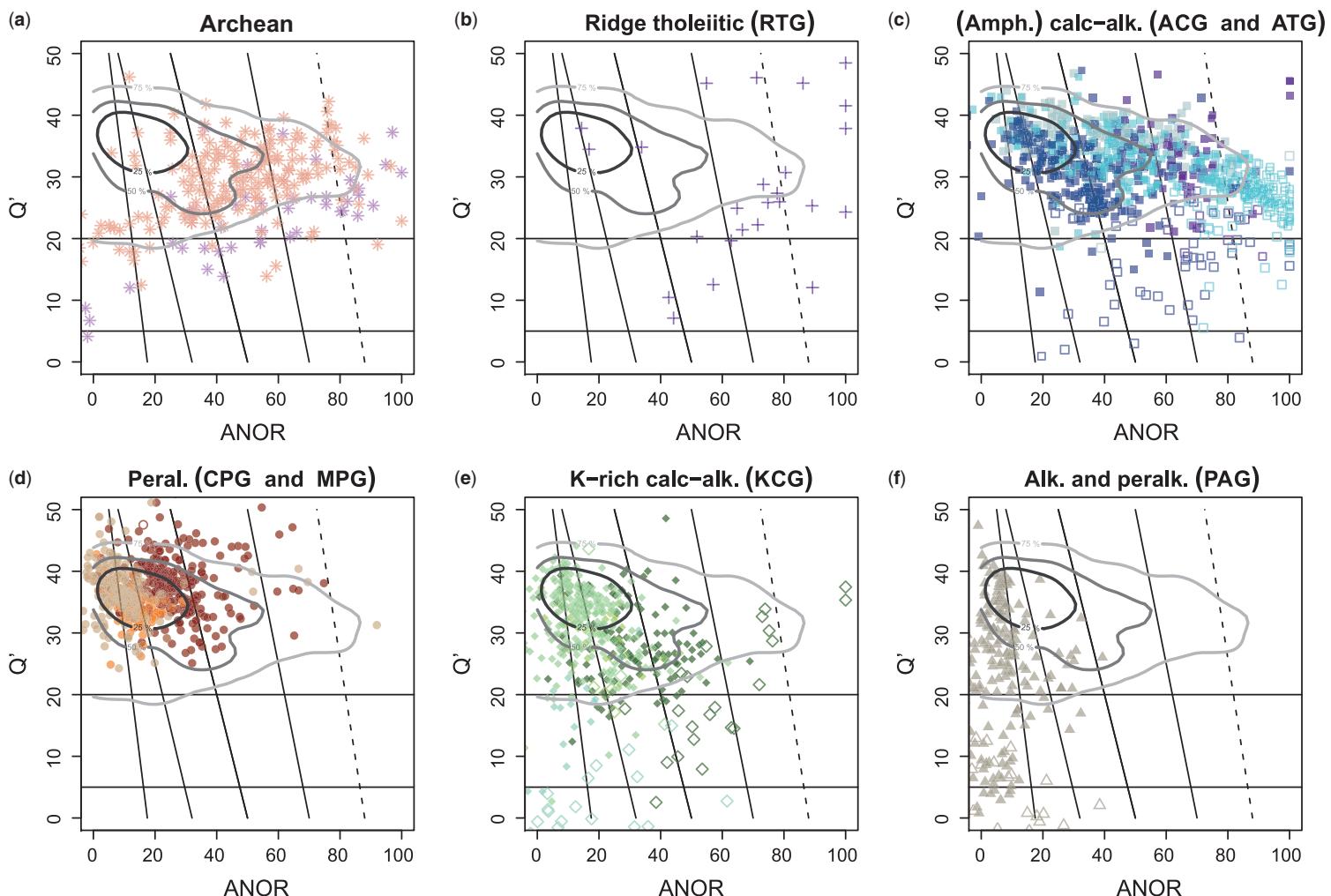
and ESM 4–13 in the Supplementary material. Figure 15 summarizes the distinguishing features.

Archean TTGs define poor trends in most diagrams. They are typically plagioclase-rich, magnesian and metaluminous to only weakly peraluminous ( $1 < \alpha < 3$ ; see ESM 5 in the Supplementary material). This is a unique combination of chemical properties, and no other granitic type really matches all the features of TTGs.

The rare RTG define a strongly subalkaline (tholeiitic) suite, also plagioclase-rich. They tend to be less magnesian than TTGs, straddling the boundary of the ferroan domain; with  $1 < \alpha < 2$  (see ESM 5 in the Supplementary material) they are weakly metaluminous. The main difference from TTGs is that they are poorer in Na: this is best evidenced

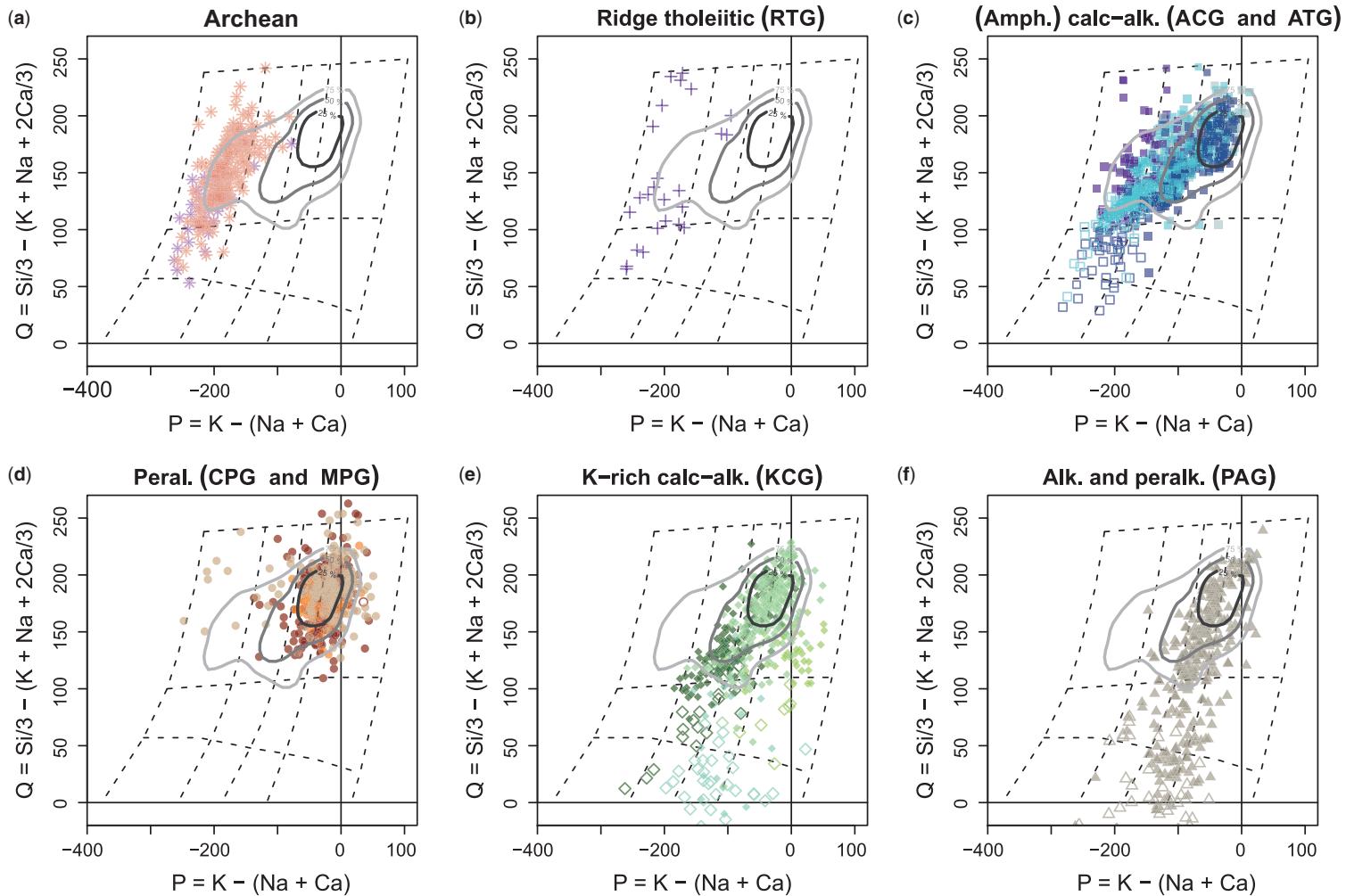
in the O'Connor's triangular diagram (see ESM 6 in the Supplementary material).

The arc-related ATG and ACG are collectively subalkaline and plagioclase-rich to transitional. They fall mostly close to the metaluminous–peraluminous boundary, the more silicic compositions becoming more aluminous (see ESM 5 in the Supplementary material), and magnesian or transitional to ferroan. Different subgroups can be identified. By comparison with ‘ordinary’ ACG, the more alkaline variety defines Ksp-richer differentiation series (Figs 12 & 15). The ATG are somewhat more ferroan (Fig. 14), and the peraluminous ACG are, for the same SiO<sub>2</sub> content, shifted towards more aluminous compositions (Fig. 13; see also ESM 5 in the Supplementary material).



**Fig. 11.** Mesonormative Q'-ANOR (Streckeisen & Le Maitre 1979) diagrams showing the position of samples from each of the six groups used in this paper. Symbols as in Figure 8; grey lines correspond to 25, 50 and 75% of the dataset.

GRANITE CLASSIFICATIONS: PAST AND FUTURE



**Fig. 12.** Same as Figure 11 but using a P-Q diagram (Debon & Le Fort 1983, 1988).

B. BONIN ET AL.

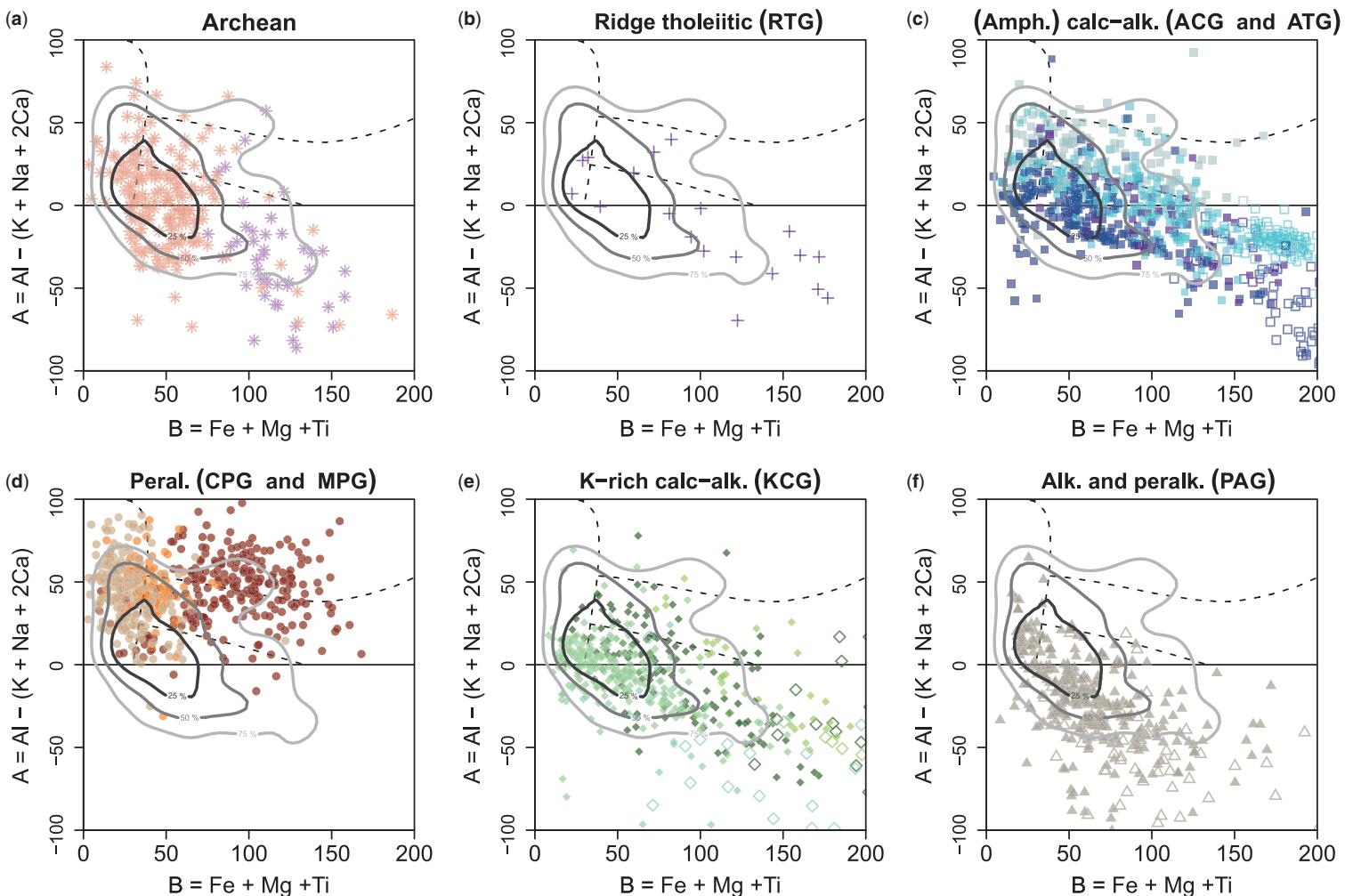


Fig. 13. Same as Figure 11 but using a B–A diagram (Debon & Le Fort 1983, 1988) modified by Villaseca *et al.* (1998).

GRANITE CLASSIFICATIONS: PAST AND FUTURE

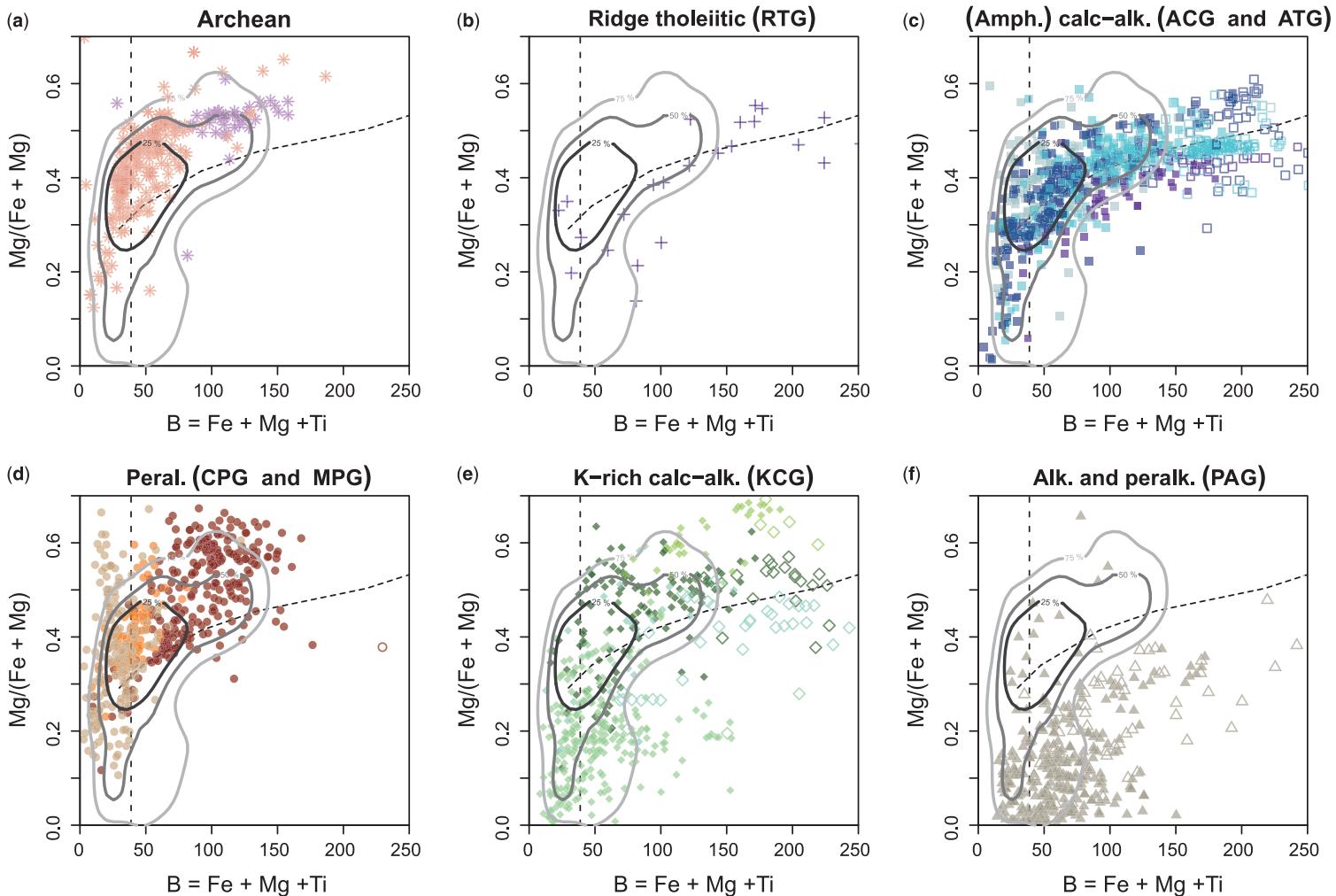
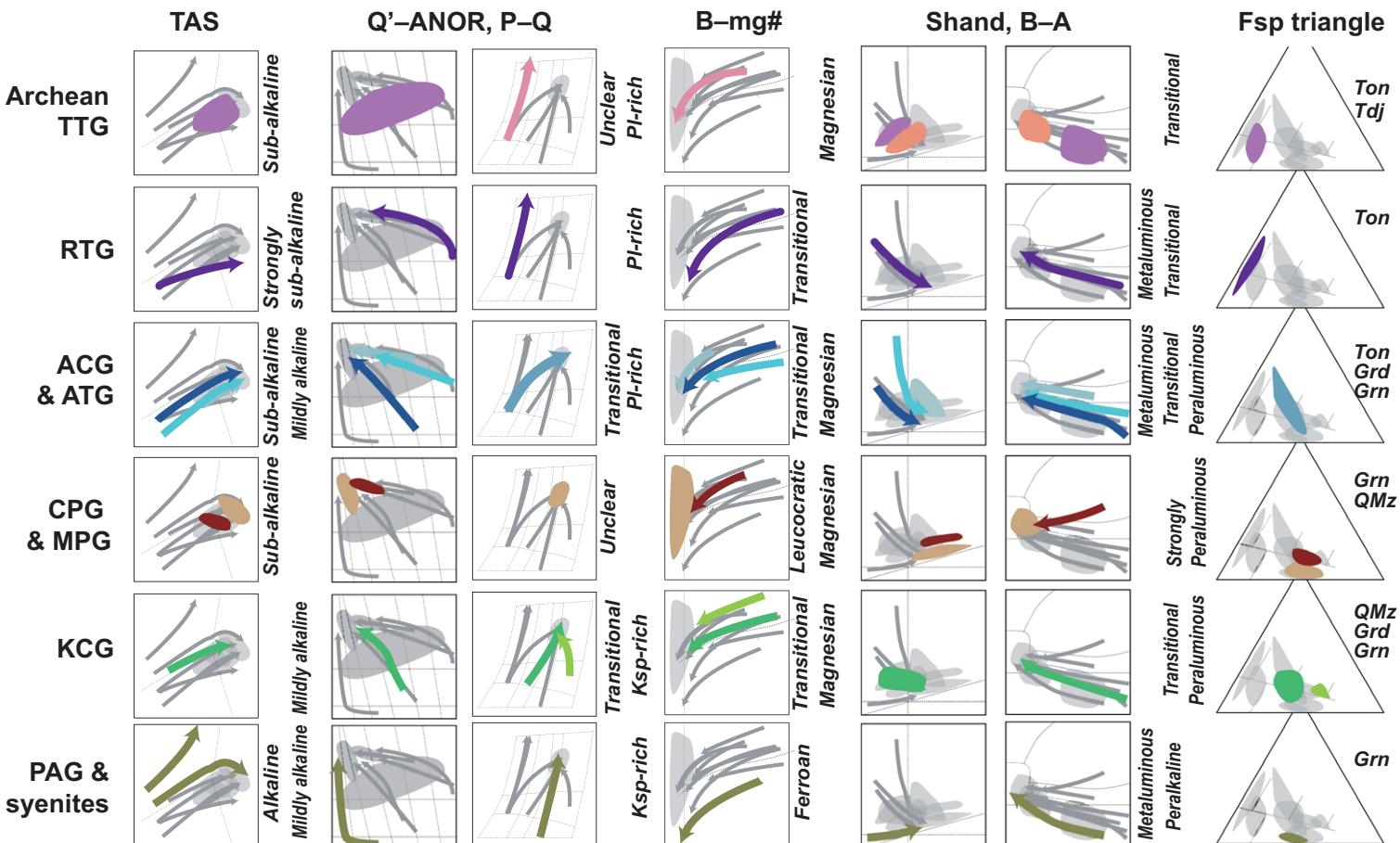


Fig. 14. Same as Figure 11 but using a B–Mg# diagram (Debon & Le Fort 1988).

B. BONIN ET AL.



**Fig. 15.** Summary of the chemical differences between granitic types. For each type, a series of small diagrams (Figs 11–14; see also ESM 4–8 in the Supplementary material) highlight the typical trends or range of values defined by this type. See the discussion in the text.

The collisional to post-collisional peraluminous granites (CPG and MPG) do not define clear trends in differentiation-related diagrams, nor in those reflecting the plagioclase/K-feldspar ratio. They are magnesian, aluminous to strongly peraluminous, with  $\alpha$  values larger (often much larger) than 2 (see ESM 5 in the Supplementary material). They are the only group in which some examples (Villaseca's h-P (highly peraluminous) type, i.e. the original Lachlan Fold Belt S-types) show positive correlations in the B–A diagram (i.e. for which the more mafic rocks are more aluminous).

The KCG are a complex group that appears to be less well defined than the other groups used here. Clearly, these granitoids require more work, even from a purely descriptive point of view, in order to establish a usable and unified classification. Collectively, KCG define poorly constrained subalkaline to slightly alkaline differentiation trends. They are K<sub>sp</sub>-rich, falling on the border between metaluminous and peraluminous domains ( $1 < \alpha < 3$ ), and ranging from ferroan to magnesian. The high-K suite is subalkaline and magnesian. The ultra-K suite is strongly magnesian, generally alkaline, but above all has an extremely high K<sub>2</sub>O content at a given SiO<sub>2</sub> (c. 6 wt% for 65–70 wt% SiO<sub>2</sub>, compared with the high-K suite which has 3–4 wt% K<sub>2</sub>O for the same silica content). The A<sub>2</sub> suite and related syenites are ferroan. The syenites are strongly alkaline, whereas the A<sub>2</sub> proper, close to the granite minimum, are ‘neutral’ to slightly alkaline.

Lastly, the PAG define clearly alkaline trends (with syenites reaching silica-undersaturated alkaline compositions) and are very rich in K<sub>sp</sub>. They are the only type that is consistently ferroan and, of course, metaluminous to peralkaline ( $\alpha < 1$ , except for the most differentiated granites that can attain very high  $\alpha$  values). Altogether, they are the best identifiable group chemically, as evidenced by all the conventional diagrams plotted here, and the LDA results.

#### *Existing classifications in light of granite chemical types*

We can briefly return to some of the commonly used classifications, in light of this analysis. An observation we have made already is that successful classifications all rely on the same small set of chemical variables, or proxies for these variables. The differences between various schemes are: (i) the relative emphasis given to each of the parameters; and (ii) the exact representation chosen to depict them. Based on our analysis, these five variables (degree of differentiation, alkalinity, aluminosity, the Fe/Mg and Na/K balance) describe the diversity of granitoids, in roughly descending order of importance.

The modal QAP diagram, and its chemical derivatives (Fig. 11), effectively plot alkalinity (plagioclase/alkali feldspar ratio) v. differentiation (quartz). By combining the two main controls, it does an excellent job at separating as many igneous types as feasible in two dimensions and, hence, justifies its IUGS-commended use as the basis for rock nomenclature. The TAS diagram (see ESM 7 in the Supplementary material) uses a somewhat less efficient approach in that it involves only the sum of alkalis (wt% Na<sub>2</sub>O + K<sub>2</sub>O) and not CaO, but builds on the same idea. Shand’s A/CNK and A/NK ratios (see ESM 8 in the Supplementary material), and the various cationic parameters of Debon & Le Fort (1983, 1988) (Figs 12–14), can be attributed to the five key variables (Q and B, degree of differentiation; P, alkalinity; A, aluminosity; Mg/(Fe<sup>T</sup> + Mg); and K/(Na + K)). Thus, it is no surprise that they turn out to be an extremely efficient way to identify rock types and, more crucially, rock associations.

The millicationionic  $R_1$  and  $R_2$  values of De la Roche *et al.* (1980) (see ESM 9 in the Supplementary material) follow a different logic: they work analogously to the PCA in that they try to combine all the major-element data in order to spread over the whole diagram. They perform their function but at the expense of the loss of an explicit understanding of the key chemical features associated with each rock type and are more suitable for silica-undersaturated, rather than granitic, rocks.

Turning to the modern ‘petrogenetic’ schemes, we first note that the S–I–M–A classification, and in particular the S/I dichotomy, relies much on the aluminosity parameter. This is the second most significant variable in our LDA analysis and thus clearly important. But of course, the fact that the other parameters are not taken into consideration is limiting its use to the geological settings where, indeed, aluminosity is the key difference – probably accounting for its limited application to, for example, arc magmatism.

Significantly, the classification by Frost *et al.* (2001) uses indicators explicitly correlated to four of the five key variables: SiO<sub>2</sub> (wt%), MALI, ASI and Fe# (ESM 10 and ESM 11 in the Supplementary material). Although we deplore the brute force approach used in the construction of these indicators, as discussed in this text, it remains that this work picks the most relevant features, and is justifiably used a lot. On the other hand, while the diagrams proposed are of widespread use, the whole classification scheme in itself is not. This may be because the original paper has not proposed useful names for each group identified, nor pointed sufficiently to petrogenetic or geotectonic implications.

The seven granitoid groups of Barbarin (1999) were defined on more than geochemical grounds.

Yet, we demonstrate here that they also correspond to clear chemical entities, overlapping largely with the results of analysis by Frost *et al.* (2001) (Table 2). The smaller number of groups in the former essentially means that Barbarin's (1999) work lumped together some of Frost *et al.*'s (2001) groups.

## Concluding remarks and prospects for the future

### *State of the art of granite classification*

In science, classifications provide an important means of communication. Well-designed schemes make us think and show causal relations. Thus, with rigour comes understanding. A good classification scheme should be unambiguous, consistent, cover the whole variability observed in nature, and be model-independent and reproducible.

Therefore, the name given to a rock should not depend on preconceptions on its formation but only on observations. In this respect, the S–I–M–A classification seems unsatisfactory, as granite types are defined (mainly) in terms of rock-forming processes. But even the criteria are not consistent: inferred source, presumably reflected by the composition of the generated magmas (I, S, M) v. geotectonic setting and specific chemistry but with unspecified source (A). In any case, the present-day usage of this classification has moved away from petrogenetic closer to descriptive (focusing on mineralogical and chemical observables) or rather informal, probably as a consequence of the community globally realizing the shortcomings of the model-based approach.

Likewise, the geotectonic classifications (Pearce *et al.* 1984; Maniar & Piccoli 1989), or even the scheme of Barbarin (1999), suffer from a similar flaw: although they are based on objective petrological and/or chemical properties, the names proposed are interpretative. To a point, this is largely a semantic matter: in all these examples, the classification is based on objective criteria, only the name refers to a model.

An ideal classification should be reproducible: that is, any researcher should, unambiguously, arrive at the same name for the same rock. The clearly diagram-based classifications (De la Roche *et al.* 1980; Debon & Le Fort 1983, 1988; Pearce *et al.* 1984; Maniar & Piccoli 1989; Frost *et al.* 2001; Frost & Frost 2008) all fit this criterion. On the other hand, classifications such as S–I–M–A or that of Barbarin (1999) rely on a degree of personal judgement and subjectivity, meaning that different observers may propose different names – clearly not a satisfactory feature.

An underlying problem with any classification of granitic rocks is that individual rock types and

samples do not occur in isolation but as a part of geochemical/petrological trends typically ranging from a common composition (the 'granite minimum') to generally more basic ones. Samples at, or near, the granite minimum are seldom discriminant, as granitic bodies in all settings and of all origins converge towards this composition. Rather, the samples further away from the granite minimum, and the trend connecting them, are more discriminant. To some degree, this makes the classification work more difficult: in any diagram-based classification, petrologically related samples will invariably plot in several distinct fields. Plotting in any of them is not a discriminant feature (especially for the fields close to the granite minimum: granite *sensu stricto*, adamellite, etc., depending on the nomenclature used). Thus, the simple act of naming a sample, even in an objective and reproducible way, is not sufficient to define a useful granitic classification.

We also note that the use of different classifications is not neutral but does actually reflect individual researcher's preconceptions on the prevalent mode of granite formation. For instance, the S–I–M–A classification seems deeply rooted in the crustal melting paradigm, whereas the Frost *et al.* (2001) scheme or the SiO<sub>2</sub>–K<sub>2</sub>O diagram of Pecceirillo & Taylor (1976), that focus on magmatic series connecting mafic to felsic rocks, are intrinsically more relevant to a basalt fractionation paradigm.

All of this highlights an important scientific danger. Since granite classification is not a simple mechanical application of universally accepted criteria, a degree of judgement call is required. It is therefore tempting to consider classification as an end in itself. Thus, it is not uncommon to see publications where the sole or the main conclusion is that 'this granitic body is of S-type', with the often unspoken implication that it is, therefore, formed by melting of sediments in a (collisional or accretionary) orogen. This, of course, is wrong. Proposing a name is useful, if only because it helps comparisons with other, well-studied cases but it cannot replace a proper petrogenetic study identifying geodynamic causes, sources and processes shaping the granitic magmatism.

Lastly, there is undoubtedly a degree of inertia. We are all used to looking at rocks and data in a specific way using our preferred diagrams, and changing to another representation, even one that would be more useful, is challenging. To a point, this is linked to our use of our favourite plotting tools. The relatively restricted abilities of spreadsheets to deal with complex diagram templates (composite projections, fields, etc.) make the introduction of a new diagram a significant investment in time and effort. Using more appropriate software, with more flexible and powerful plotting abilities, helps in smoothing this transition.

**Table 2.** Comparison of the chemical features and classification, under several schemes, of granitic compositions

Chemical features				Classification		
Debon & Le Fort (1983); Frost <i>et al.</i> (2001)	Frost <i>et al.</i> (2001)	Shand (1927); Debon & Le Fort (1983); Frost <i>et al.</i> (2001)	Debon & Le Fort (1983)	Example in Frost <i>et al.</i> (2001)	Barbarin (1999), classified in this work	Granite 'alphabet'
Mg/Fe balance (LD <sub>3</sub> )	Alkalinity (LD <sub>1</sub> )	Aluminosity (LD <sub>2</sub> )	Na/K balance (LD <sub>4</sub> )			
Magnesian	Calcic	Metaluminous	Potassic	Outboard portion of Cordilleran batholiths, island arcs, oceanic plagiogranites	ATG and ACG	M and I
		Peraluminous	Sodic		RTG	M and A
	Calc-alkalic	Metaluminous	Potassic	Main portion of Cordilleran batholiths	ACG (peraluminous)	I
		Peraluminous	Sodic	Not described	ACG (rare KCG)	I
	Alkali-calcic	Metaluminous	Potassic	Main portion of Cordilleran batholiths	TTG	I (or not identified)
		Peraluminous	Potassic	Inboard of Cordilleran batholiths; delamination of overthickened crust	CPG/MPG	S
	Alkalic	Metaluminous or Peralkaline	Potassic	Inboard of Cordilleran batholiths	KCG	I
		Peraluminous	Potassic		CPG/MPG	S
	Ferroan	Metaluminous	Potassic	Not identified	Not identified	I or A?
		Metaluminous	Potassic	No example known	Rare ACG?	
		Peraluminous	Potassic	No example known	Not identified	I
		Metaluminous	Potassic	No example known	Some PAG	A
		Peraluminous	Sodic	Some A-type granites, some granophyres in mafic plutons	RTG	M
		Peraluminous	Potassic	Two-mica granites	Not identified (some differentiated compositions?)	
		Metaluminous	Potassic or sodic	A-types, granophyres associated with mafic dykes and plutons	some KCG (A <sub>2</sub> ), some PAG	A or I
		Peraluminous	Potassic or sodic		Not identified (some differentiated compositions?)	
		Metaluminous or Peralkaline	Potassic or sodic	Within-plate plutons, plutons under alkaline volcanoes	PAG	A
		Peraluminous	Potassic or sodic	No example known	Not identified	

The geochemical indicators are described using the nomenclature of Frost *et al.* (2001) but essentially correspond to the analogous groups of Debon & Le Fort (1983, 1988) and results of our analysis (see the individual LDA components). The combination of the four chemical indicators (differentiation excluded) split into two, three or four groups each, allowing in theory  $(2 \times 4 \times 3 \times 2) = 48$  combinations. However, many of them do not exist (sodic compositions, for instance, are rare outside of the Archean and peralkaline compositions). Frost *et al.* (2001) identified 14 possible combinations. We identified another two (Archean TTGs that are magnesian, calc-alkalic, metaluminous and sodic, as well as a rare ferroan, calcic, metaluminous potassic component). All 16 can be largely mapped to the seven types of Barbarin (1999), and in turn reinterpreted in the 'granite alphabet'.

Indeed, calculations and plotting of rather complex diagrams that were once limiting factors no longer represent a problem. Besides commercial solutions, there are several software packages freely available for this task, including PetroGraph (Petrelli *et al.* 2005), t-IgPet (Carr & Gazel 2017) and R/GCDkit (Janoušek *et al.* 2006, 2016). The last one is even capable of accepting user-defined diagram templates and plug-in modules.

### Towards an ‘ideal’ classification scheme

In general, science is about the communication of ideas – if everybody understands what we mean by the given name, even an inappropriate one, then it serves its purpose. All of us are, to varying degrees, ignorant or careless with regard to the original definitions and proper meaning of even the most elementary terms. However, over time, some of them (e.g. the adjective ‘calc-alkaline’) have become so ambiguous that they should be either redefined or dropped.

We propose that a useful classification of granitoids should:

- Be based on criteria that are more amenable to an objective treatment (plotting in a diagram, algorithmization, etc.) rather than qualitative observations.
- Modal proportions and actual mineralogical descriptions, in principle, should be preferred (as recommended by the IUGS in the QAP classification and useful in the field) but since precise modal analyses are tedious and hardly performed these days, it is unlikely that they will prevail in the future.
- In the absence of modal determinations, they should employ major elements, as they enable linkage to the chemical properties with the main rock-forming minerals present (i.e. we stay close to the observed mineralogy of the rock). Trace elements behave independently of the main phases (they either substitute for major cations in main rock-forming minerals or form accessory minerals of their own) and therefore bring different information.
- Use a simple representation of major-element data that makes linking to the modal proportions and compositions/structure of individual rock-forming minerals easier. In this light, wt% oxides are a poor way to express chemical properties; atomic proportions (millications, etc.) are much more appropriate. On the other hand, while we acknowledge the use of log-transformed values for statistical purposes, we feel that they are too remote from actual petrological observables to be of practical value to a petrologist.

The fact that the compositional space has five degrees of freedom (at least three of which are

needed to fully describe the observed range of compositions) means that one cannot restrict oneself to the application of a single diagram. Thus, an ‘ideal’ classification scheme should additionally fulfill the following two desirable features:

- It should be based on several diagrams that will highlight the key geochemical properties of granitoids, namely (i) acidity (or maficity, or something related), in the form of Si, Fe + Mg, etc.; (ii) alkalinity, related to the balance between Na + K and Ca; (iii) aluminosity, which is the balance of Al with Ca, Na and K, critical for the identification of ‘S-types’; (iv) the Fe/Mg balance (mostly useful for the alkaline or nearly alkaline series); and (v) the Na/K balance and K content at the given SiO<sub>2</sub> level (critical for Archean TTGs, also important in the alkaline or high-K realm).

The reader familiar with the literature on granitoids has, of course, recognized many of the popular diagrams already in existence: they are time honoured for a good reason, and we wish to salute the memory of the petrologists who had the intuition to identify these key variables a century ago previous to the advent of cheap analyses, large databases and desktop computers.

- It should aim to identify trends and associations, and not individual compositions. To our knowledge, despite some attempts in this direction (Debon & Le Fort 1983, 1988; Villaseca *et al.* 1998; Frost *et al.* 2001; Moyen *et al.* 2017), no universally accepted classification scheme taking this aspect into account exists so far, and we suggest that some effort should go in this direction.

For all reasons summarized in this text, it is probably presumptuous to recommend the use of one specific classification scheme over all other ones. Yet, most of our criteria for an ‘ideal’ classification scheme are met by the millication-based multi-element diagrams of Debon & Le Fort (1983, 1988) and their derivatives. In our view, this scheme should be resurrected for the profit of the community (note that the overview of the existing schemes of Frost *et al.* (2001) does not even mention its existence), hopefully becoming a new standard in the granitoid literature.

To sum up, classifications are just one of the tools in our toolbox, the main goal of which is to facilitate the communication and to fuel creative thinking. In deciphering the genesis and geotectonic context of igneous rocks, a holistic approach is to be adopted. Thus, one should combine all available field observations with structural geology, petrology, age, mineral chemistry and complete whole-rock geochemical signature into a single coherent and internally consistent model that does not have to rely on some predefined pigeonholes.

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