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Statistical modelling of compositional trends in sediments

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Abstract

Trends in compositional diagrams are frequently used to describe and interpret the imprint of certain processes on sediment composition. Most of these trends are intuitive and "hand-drawn", although a numerical description of compositional changes involved with these processes would allow for a quantitative assessment of the observed sediment compositions and the proposed processes. This paper suggests a statistical approach to model compositional linear trends, which allows for a mathematically rigorous description of changes in sediment composition related to a specific process or a combination of processes. Three case studies dealing with different aspects of the sedimentary cycle demonstrate the usefulness of the approach. In the first example, a modelled average global chemical weathering trend is used to calculate an appropriate measure for the degree of chemical weathering of the suspended load from some of the world's major rivers. In the second example, data on precipitation and relief from different watersheds are quantitatively related to the petrographic composition of fine-grained sands from low-order streams derived from these watersheds. The third example is an attempt to quantify the interdependence of chemical sediment composition and grain size in the absence of chemical weathering. Superposition of individual compositional linear trends allows for modelling sediment composition in more complicated case studies. Although primarily a forward modelling approach, it can be also used inversely, and, therefore, has strong potential for tracing back the sediment-forming processes in order to reconstruct the original source rocks of the sediment.
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1. Introduction

The composition of clastic sediment is generally characterized by its physical features (most important: grain size distribution), chemical composition, and petrographic composition (texture and mineral phases of individual sediment grains). This overall composition is controlled by a broad range of parameters starting with the composition of the source rock(s) and followed by the various processes that modify the sediment all along the way from the former source area to the present-day situation in outcrop or in the subsurface (the so-called sedimentary cycle). The

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most important modifying processes are chemical weathering and erosion in the source area, abrasion and hydrodynamic sorting and mixing during transport and deposition, as well as diagenetic processes during the burial stage (e.g., Johnsson, 1993). Sedimentary rocks may be exhumed again to the surface and are then exposed to another round within the sedimentary cycle. This sediment recycling further complicates the reconstruction of the processes forming sediment composition.

All these processes change the composition of the sediment relative to the initial source rock composition, and one of the main tasks in sedimentary petrology is to figure out if observed changes in sediment composition (e.g., through time or within space) are related to climate changes, changes in the source rocks, diagenetic processes, or some other process or combination of processes.

The evolution of sediment compositions under given circumstances are usually illustrated by trends in compositional diagrams, e.g., ternary diagrams (e.g., Cox and Lowe, 1995). Such trends are often interpreted as being caused by a specific geological process such as changes of source rocks through time, sediment recycling, or chemical weathering (Fig. 1). Regardless whether these controlling processes are correctly interpreted or not, trend lines are usually "hand-drawn" and there is mostly no statistical or other numerical rationale behind the interpreted trends. Statistical modelling of such trends allows

for a quantitative description and analysis of the observed trend. Furthermore, if the compositional trend related to a specific process is already quantified, it can be used to test whether an observed trend can be explained by the proposed process or not.

The aim of this paper is to briefly outline a mathematical approach for analysing and testing such changes in sediment composition with statistical rigour. Statistical rigor means here that the specific nature of compositional data and their appropriate sample space (the so-called simplex; Aitchison, 1986) has to be taken into account. The approach largely consists of finding a statistically derived equation of a straight line within the simplex which best describes the observed compositional evolution of a sample suite. This line is called a compositional linear trend (for details of the mathematics involved, see von Eynatten et al., 2003). In essence, fitting linear compositional trends in the simplex is equivalent to linear regression in the real space. Compositional linear trends provide an objective and consistent alternative to the common hand-drawn trend lines.

In particular, this paper ought to demonstrate the usefulness of the approach by some examples dealing with different aspects of the sedimentary cycle. In these case studies, compositional linear trends serve as a basis for inference regarding causal mechanisms. This includes (1) calculation of a process-related measure for the degree to which a specific process was active, (2) evaluation of the presence or absence
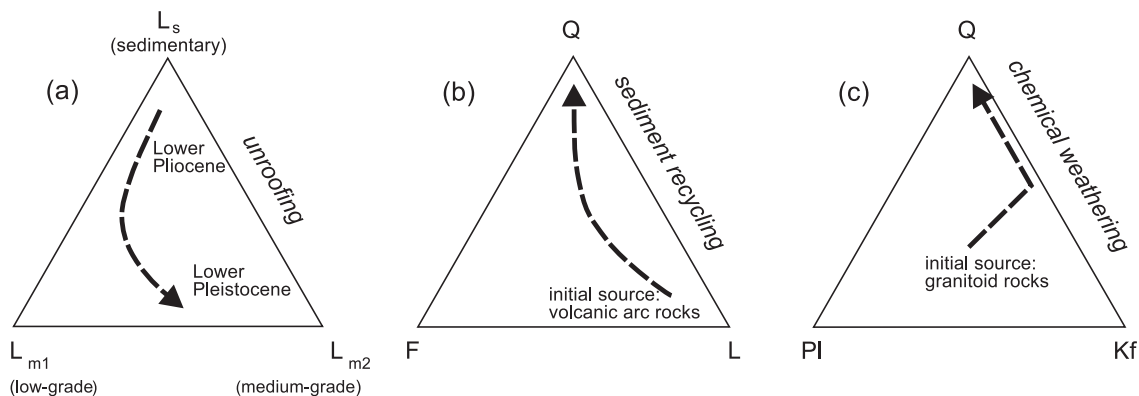

Fig. 1. Some examples from the literature showing compositional evolution of sediments and the proposed processes controlling these compositional trends: (a) unroofing in the source area as recorded by a decrease of sedimentary lithoclasts (L_s) and an increase of progressively higher grade metamorphic lithoclasts (L_m; Dorsey, 1988); (b) cumulative sediment recycling as recorded by a decrease of lithoclasts and an increase of quartz grains in QFL-space (Cox and Lowe, 1995); (c) increasing chemical weathering as recorded by the removal of first plagioclase grains (Pl) and later K-feldspar grains (Kf) from sand-sized sediment (Nesbitt et al., 1997).

of an additional process inferred by deviation of local observations from a general trend, (3) accepting or rejecting proposed causal mechanisms by comparing values which describe the degree of change along the compositional linear trend with values describing the cause (e.g., some kind of weathering index), and (4) evaluation and separation of the effects of multiple causes on sediment composition.

The method is primarily a forward modelling approach, but it can also be used as an inverse modelling approach and, thus, provides a helpful tool in deciphering quantitatively the provenance characteristics from sediments with well-known composition. I refer to the term provenance in a broad sense, which means, encompassing all factors related to the production of the sediment (Pettijohn et al., 1972, 297f).

2. Mathematical approach

Most of the data used to describe sediment composition—either chemistry or petrography or grain size—are compositional data, which means each variable is non-negative and all variables of a composition sums to a constant c (usually 100%). These conditions are known as the constant-sum constraint that raises—among many other pitfalls— serious problems concerning correlation and trend analysis of compositional data (e.g., Chayes, 1960; Butler, 1979). The sample space for d-part compositional data is not the d-dimensional real space $\Re^d$, but the simplex S^d (Aitchison, 1986). Therefore, refined statistical models need to be applied that allow for a statistically rigorous analysis of changes in sediment compositions. If $d=3$, the simplex is graphically represented by the well-known ternary diagram that is used for illustrations throughout this contribution.

The simplex has a vector space structure and mathematical operations are defined that are quite similar to those used in the real space (Aitchison, 1986; Pawlowsky-Glahn and Egozcue, 2002). This paper focuses on mathematical operations that can be combined to define compositional straight lines in the d-dimensional simplex S^d. These trend lines are called compositional linear trends and allow for describing observed trends of compositional changes.

A compositional linear trend is determined by an initial (d-part) composition $\mathbf{a}=[a_1, \ldots, a_d]$, and with direction given by the unitary perturbation vector $\mathbf{p}=[p_1, \ldots, p_d]$. Fig. 2 illustrates the main properties of compositional linear trends for the three-dimensional case ($d=3$). The scalar t determines the position (composition) of a specific sample along the compositional linear trend. Thinking in terms of a process, the scalar t ($t \in \Re$) determines the relative change in composition compared to the original state (composition $\mathbf{a}$) depending on the degree of intensity to which that specific process has been active.

Thus, any d-part composition $\mathbf{y_t}=[x_1, \ldots, x_d]$ on the trend may be calculated by

$$\mathbf{y_t} = \mathbf{a} \oplus (t \otimes \mathbf{p}) \tag{1}$$

or

$$\mathbf{y_t} = C\left[a_1 p_1^t, \ldots, a_d p_d^t\right]. \tag{2}$$

Eq. (1) is the vectorial form using simplex operators, where $\oplus$ and $\otimes$ denote two mathematical operations in the simplex, perturbation and power transformation, that are comparable to addition and multiplication in real space, respectively (Aitchison,
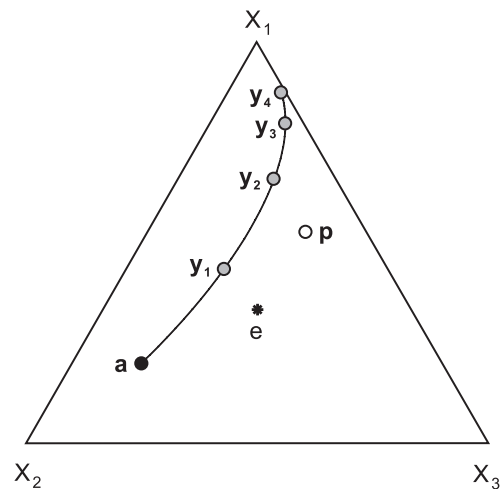

Fig. 2. Ternary diagram illustrating the properties of compositional linear trends in S^3; e denotes the centre (neutral element) of S^3, $\mathbf{a}$ denotes the initial composition, $\mathbf{p}$ denotes the perturbation vector that indicate the direction of the trend, and $\mathbf{y_t}=\mathbf{a}\oplus(t\otimes\mathbf{p})$ indicates specific compositions along the compositional linear trend, here illustrated for $t=1, 2, 3, 4$. Increasing numbers for t indicate increasing degree of change in composition relative to the initial composition $\mathbf{a}$. Modified after von Eynatten et al. (2003).

1986). Note that Eq. (1), describing compositional linear trends in the simplex, is quite similar to the vectorial equation of straight lines in the real space, $\mathbf{y}=\mathbf{b}+\lambda\mathbf{u}$. The position vector $\mathbf{b}$ corresponds to the initial composition $\mathbf{a}$ in Eq. (1), the direction vector $\mathbf{u}$ corresponds to the perturbation vector $\mathbf{p}$ in Eq. (1), and the scalar values λ and t (λ, $t \in \Re$) indicate the position of a specific point on the straight line or a composition on the compositional linear trend, respectively.

Eq. (2) is the explicit form using vector components, where C denotes the closure operation (each component of a vector is divided by the sum of all its components, which means that each composition is normalized to the sum of 1 or 100%).

Given a suite of sediment samples with known composition that is considered to describe the effects of a specific (linear) process, the task is to adjust a compositional linear trend that (1) explains as much as possible of the total variability of the data set, and (2) passes through the initial composition $\mathbf{a}$ (not yet modified by the process), if known. If $\mathbf{a}$ is not known, the leading perturbation vector $\mathbf{p}$ related to the proposed process is adjusted by calculating the first principal component (pc1) or eigenvector of the data set, which captures as much as possible of the total compositional variability. The percentage of the total variability that is explained by pc1 allows for judging on the precision of the approximated linear trend. Standard principal components by definition pass through the centre (geometric mean) of the data set and are only useful if the initial composition $\mathbf{a}$ is not known (in this case, composition $\mathbf{a}$ in Eq. (1) is replaced by the centre of the data set). If $\mathbf{a}$ is known, the leading perturbation vector $\mathbf{p}$ is calculated by noncentral principal component analysis, which means that the variances are calculated relative to the initial composition $\mathbf{a}$ (and not relative to the mean, as usual) and, hence, the resulting noncentral first principal component explains as much as possible of the total variability with respect to $\mathbf{a}$.

Of course, many samples will lie somewhat off the calculated compositional linear trend. Therefore, it may become necessary (1) to calculate a corresponding composition and its t-value on the trend for each of these samples, and (2) to judge on the degree of deviation from the trend (if the deviation from the trend is high, another process or a second superimposed process must be responsible for this specific composi-

tion and its position far off the trend). Concerning (1), the orthogonal projection from the sample in question onto the compositional linear trend allows for determining the corresponding composition on the trend with its specific value of t. This composition is defined by the smallest Aitchison distance to the sample in question (the Euclidian distance is not an appropriate measure, because we are operating in the compositional space S^d, see Aitchison, 1992). Concerning the degree of deviation from the trend, the Aitchison distance again is an appropriate measure, but establishing a critical threshold depends on the case study and its specific questions. Alternatively, an array may be defined along the trend based on the variability of the data with respect to the compositional linear trend at a certain level of confidence.

The details of the mathematics are not the focus of this paper and the reader is referred to von Eynatten et al. (2003) for the more theoretical background. For easy computation of compositional linear trends and related calculations, we are currently developing a trend analysis tool for implementation into the Excel-based CODAPack software package. This software is programmed by Santiago Thió Fernández de Henestrosa from University of Girona (Spain), and is freely available under http://ima.udg.es/~thio/#Compositional Data Package. The trend analysis tool will be also freely available as soon as possible.

In case of more complex non-linear processes, compositional trends may be calculated by more sophisticated statistical methods like differential perturbation (Aitchison and Thomas, 1998). In any case, the method described and applied in this paper is considered to allow for a reliable linear approximation of the trend.

3. Case studies

The above-described mathematical approach for modelling trends in sediment composition is applied to three case studies taken from the literature. All examples are largely dealing with weathering processes, both chemical and physical. All data sets used have three components or are amalgamated from more variables to a three-part composition for the purpose of illustrating the results in ternary diagrams.

However, the mathematical approach holds for compositional data of any dimension (von Eynatten et al., 2003).

3.1. Chemical composition of suspended river load

Rocks exposed to and sediments transported on the earth's surface are subjected to alteration processes with chemical weathering being one of the most important. Because feldspar predominates on average in crustal rocks, their transformation to clay minerals and Al-hydroxides is essential to chemical weathering. Basically, this transformation is chemically expressed by the removal of Ca, Na, and K and a relative enrichment of Al and may be nicely illustrated in A–CN–K ternary diagrams (Al_2O_3–$CaO+Na_2O$–K_2O). In order to calculate a global chemical weathering trend within the A–CN–K three-dimensional simplex, a data set compiled by McLennan (1993) is used which is based on estimates of the bulk chemical composition of suspended sediments from several of the world's largest rivers ($n=16$) and of erosional products from some of the world's major denudation areas ($n=7$). Of course, many factors, both natural and anthropogenic, are involved in determining the composition of the sediment in present-day rivers

and may have a certain imprint on sediment composition (see discussion in McLennan, 1993), but none of these is capable of significantly perturbing the general trend of relative Al-enrichment at the expense of alkaline and alkaline earth elements (Fig. 3a). The degree of this enrichment is usually measured by the Chemical Index of Alteration (CIA), starting with values of ~50 for the initial source rock and ending up with values close to 100 for the final, completely weathered residue. In the present case study, CIA-values of river sediment range from 51 (St. Lawrence) to 95 (Niger).

The compositional linear trend is adjusted using noncentral principal component analysis with the average composition of the upper continental crust as initial composition **a** (Taylor and McLennan, 1985). The result is a precise approximation of the global chemical weathering trend in the A–CN–K simplex with 97% of the total variability being explained by the compositional linear trend (Fig. 3). For each river, a corresponding t-value is calculated using the orthogonal projection from the river's sediment composition onto the compositional linear trend. This value describes best the composition of the river sediment along the trend and, hence, the degree of chemical weathering the sediment has experienced
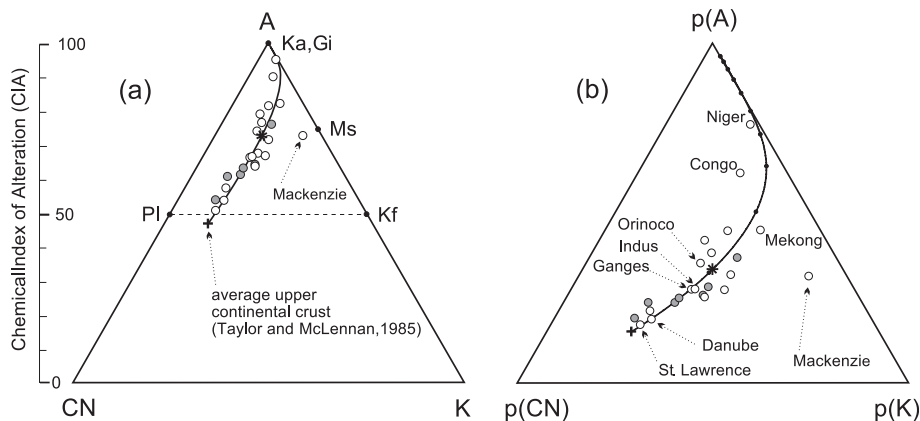

Fig. 3. A–CN–K ternary diagrams showing the bulk chemical composition of suspended sediments from several of the world's largest rivers (open circles) and from erosional products from some of the world's major denudation areas (grey circles; all data from McLennan, 1993; A=Al_2O_3, CN=$CaO+Na_2O$, K=K_2O; all in molar proportions). Note the composition of Mackenzie river sediment that lies significantly off the trend. (a) Traditional diagram also showing position of selected mineral compositions (Pl=plagioclase, Kf=K-feldspar, Ms=muscovite, Ka=kaolinite, Gi=gibbsite) and the adjusted compositional linear trend (solid line). CIA-values (Nesbitt and Young, 1982) are displayed on the left. (b) Centred ternary diagram, which means that the centre of the data set (black star) is moved into the centre of the diagram, a technique that allows for better estimating distances between compositional data points (see von Eynatten et al., 2002, for details). Black dots along the compositional linear trend (solid line) indicate compositions for t-values 1, 2, ..., 10.

relative to the initial composition, i.e., the average upper continental crust.

The scalar t ranges from small values for rivers in moderate climate regions like St. Lawrence (0.13) and Danube (0.25) via intermediate values for rivers like Ganges (0.75), Indus (0.78), and Orinoco (1.01) up to high values for rivers in low-relief areas under tropical climate like Mekong (1.87), Congo (2.15), and Niger (3.49) (Fig. 3b). These values express a process-related quantitative measure of the degree of chemical weathering of the suspended load of each river. In contrast to CIA-values, it is a translation invariant measure in the sense that the incremental increase in weathering between, e.g., St. Lawrence (0.13) and Orinoco (1.01) sediments and Orinoco and Mekong (1.87) sediments is about the same. This information is not obtainable from CIA-values (St. Lawrence 51, Orinoco 74, Mekong 82), which is a direct conse-quence of the relative nature of the data and the erroneous use of the Euclidean distance when analysing compositional data (von Eynatten et al., 2002). This point becomes crucial if, for example, immature siliciclastic rocks that already experienced some (paleo-)weathering are presently eroded and weathered again, and the recent degree of weathering is to be compared to the recent weathering of a fresh, e.g., granitic, rock.

For the purpose of provenance studies of fine-grained sediments or sedimentary rocks, the proposed global weathering trend can be used for a first evaluation if a sample fits to the average trend or not, and to what extent the material was affected by chemical weathering (t-value). If a composition plots considerably off the trend, like the Mackenzie river in Fig. 3, a second superimposed process or a basically different process must be considered. In this example, the Mackenzie river is the only Arctic river in this case study suggesting a contrasting weathering behaviour compared to the moderate to tropical climate conditions of all the other rivers. This may be caused by a higher relevance of physical processes (see Section 3.3). The problem of superposition of different processes is schematically illustrated in Fig. 4, and a case study dealing with chemical weathering and superimposed K-metasomatism (Fedo et al., 1995) was already worked out in a previous paper (von Eynatten et al., 2003). I will come back to this point in the discussion chapter.
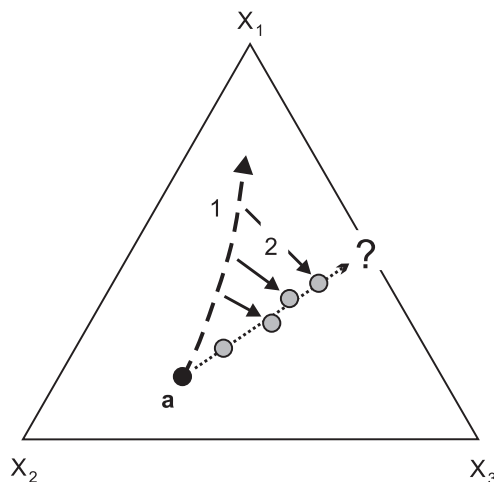

Fig. 4. Synthetic example illustrating schematically the super-position of compositional linear trends. A suite of compositions (grey circles) with initial composition **a** may suggest a composi-tional trend (dotted line) which in reality cannot be explained by a specific (geological meaningful) process but by the superposition of the two processes 1 (dashed line) and 2 (solid line).

3.2. Petrography of fine-grained sands in low-order streams

The second example deals with petrographic data of modern fine-grained sands, which are derived from eight different watersheds within a relatively small study area (~16 km²), the Coweeta Basin in the Blue Ridge Mountains, NC, US (Grantham and Velbel, 1988). Although quite small, the study area shows a large variability in annual precipitation ranging from ~170 to ~240 cm, and a quite rugged topography ranging from ~1600 down to less than 700 m. For each watershed, a Cumulative Chemical Weathering Index (CCWI) was calculated based on effective precipitation (annual stream discharge per unit area) divided by the relief ratio as a measure for the average duration of weathering at each watershed. The resulting CCWI values vary from 100 to 274 (Grantham and Velbel, 1988). This means that in this example there exists an external variable that is thought to control the degree of weathering in each watershed, i.e., the composition of the sands in low-order streams close to the base of the watershed.

The source rocks for the sediments analysed are predominantly Upper Precambrian metasedimentary rocks throughout the study area. A major difference in the source rocks lies in the maturity of the sedimen-

tary protoliths. In the northern part of the study area, the protoliths of the present-day metasediments were less mineralogically mature implying a higher contribution from metagreywackes and mica schists, whereas in the southern part, there is a higher contribution from gneisses, meta-arkoses, and quartzites (Grantham and Velbel, 1988). Consequently, sands derived from the northern watersheds contain on average more rock fragments.

Three samples were taken from first- and second-order streams of each watershed. Sand composition was determined by point counting of four grain types (variables): monocrystalline quartz (Q_m), polycrystalline quartz (Q_p), rock fragments (L), and mica (M). In order to account for the compositional changes caused by grain size, samples were analysed separately for three grain size classes: coarse (-1 to 1 ϕ-grade), medium (1 to 2 ϕ-grade), and fine (2 to 4 ϕ-grade) (Grantham and Velbel, 1988).

For the purpose of this study, geometric means of sand compositions for each watershed and grain size were calculated. This data set is now used to (1) model the compositional linear trend that describes best the sand compositions of the different watersheds, and (2) quantitatively relate sand composition to the calculated weathering index CCWI. Because of the strongly different hydrodynamic behaviour of mica grains compared to the other grain types, especially quartz, mica is precluded from considerations. The resulting three-dimensional data set can be illustrated in the ternary diagram Q_m–Q_p–L, with the fine-grained samples showing on average the highest content of Q_m and the coarse-grained samples showing the highest content of rock fragments (Fig. 5a).

To calculate a compositional linear trend, which model the effects of weathering on sand composition, the data set should be restricted to a specific grain size

because otherwise the trend would be biased by the effects of grain size on sand composition. In case of the fine-grained samples, the adjusted trend using the first principal component explains the highest percentage (81%) of the total variability, compared to
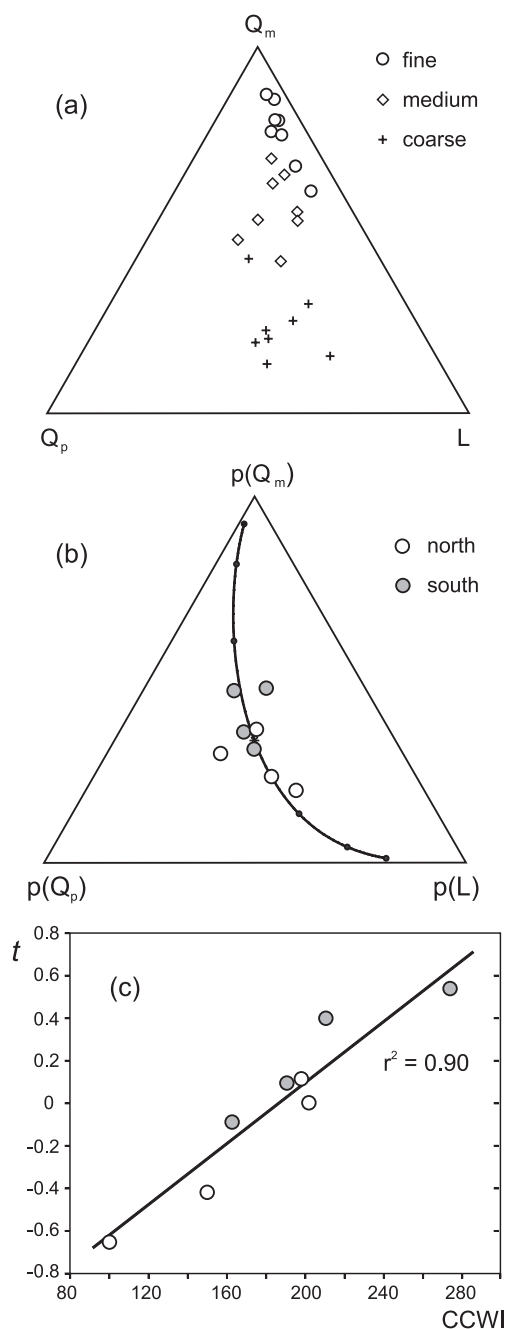

Fig. 5. (a) Q_m–Q_p–L ternary diagram displaying mean sand compositions of the eight watersheds for each of the three grain size classes. (b) Centred Q_m–Q_p–L ternary diagram displaying sand compositions of the eight fine-grained samples (the centre of the fine-grained data set is moved to the centre of the diagram). Data are separated into northern and southern watersheds. The adjusted compositional linear trend passes through the centre of the data set. Black dots along the compositional linear trend indicate compositions for t-values ± 1, ± 2, and ± 3. (c) Bivariate plot of the scalar value t and the corresponding weathering index CCWI for each watershed ($n=8$). Linear correlation is significant at the 1% level with $r^2=0.90$.

~70% for the medium-and coarse-grained samples. Because there is no well-defined starting composition like in the previous example, the perturbation vector is calculated by standard principal component analysis passing through the centre (geometric mean) of the fine-grained data set (Fig. 5b). The orthogonal projection from each composition onto the compositional linear trend is again used to calculate the corresponding scalar value t for each fine-grained sand composition. These t values show a highly significant correlation (at the 1% level) to the cumulative weathering indices CCWI of the eight watersheds (Fig. 5c) suggesting that sand composition is strongly controlled by CCWI. Consequently, it is possible to calculate—under comparable geological conditions—for a given watershed with known discharge rates and relief an approximate composition of the fine-grained sand fraction in the three-part Q_m–Q_p–L simplex. Or vice versa and even more important for provenance studies, we can calculate an appropriate weathering index CCWI for the corresponding watershed (source area) of a given sand composition in first- or second-order streams.

The linear correlation between the t values defining the position on the compositional linear trend and the weathering index CCWI suggest that the contrasting source rock lithologies in the northern and southern watersheds have a relatively low impact on the composition of the fine-grained sand fraction compared to the combined effects of effective precipitation and relief. This is in principal agreement to the conclusion of Grantham and Velbel (1988) that rock-fragment abundance is largely controlled by the CCWI, but appears to be in contrast to Heins (1993) who concluded that rock-fragment abundance from, at least, four of the watersheds (the southern group with more mature source rocks) is largely controlled by the areal proportions of different source rocks. However, this is not really contradictory because the external variables considered are not independent of each other. For example, source-rock type and strength usually has considerable influence on relief (in this case study the highest relief corresponds to the highest areal proportion of a specific rock formation) and vice versa (the lowest relief corresponds to the watershed with the highest areal distribution of Quaternary cover and, not surprisingly, the highest CCWI; Grantham and Velbel, 1988; Heins, 1993).

Compared to the previous studies, the compositional linear trend approach provides a robust solution that is statistically sound, covers (almost) the whole sand composition (here: three compositional variables instead of only rock-fragment percentage), and uses a cumulative external variable that encompasses directly and indirectly a large part the controlling parameters.

3.3. Grain size vs. bulk chemical composition of glacio-fluvial sediments

The grain size of sediments largely results from source rock comminution (mechanical and chemical abrasion) and hydrodynamic sorting. In the previous example, the strong impact of grain size on sand composition was already mentioned (Fig. 5a). Therefore, any evaluation of the influence of a specific process on sand composition must be performed under strict grain-size control, or, when trying to evaluate exclusively the influence of grain size on sand composition, a case study must be selected where other processes like chemical weathering can be more or less precluded. Such an example is presented by a study of the chemical composition of sediments from the Guys Bight Basin, Baffin Island, Arctic Canada, where physical processes greatly predominate over chemical processes (Nesbitt and Young, 1996). Mechanical abrasion occurs primarily within the glacial regime, whereas selective sorting occurs later in the fluvial regime. Due to very low temperatures (~9 month below freezing) and short residence time of the sediment in the basin, no significant weathering profiles are developed (Nesbitt and Young, 1996). Hence, the basin is ideally suited to study the interdependence of sediment grain size and composition in the absence of chemical weathering.

The source rocks of the Guys Bight Basin sediments are mostly high-grade metamorphic rocks with a minor contribution from plutonic rocks. Feldspar, quartz, and biotite are the most abundant minerals in both source rocks and sediments. Grain sizes of the investigated sediments range from coarse sand to mud. Bulk chemical major element composition of the sediments indicates a decrease in SiO_2 and an increase in FeO_{tot}+MgO from coarsest to finest grain size grade. Al_2O_3, CaO, Na_2O, and K_2O do not change
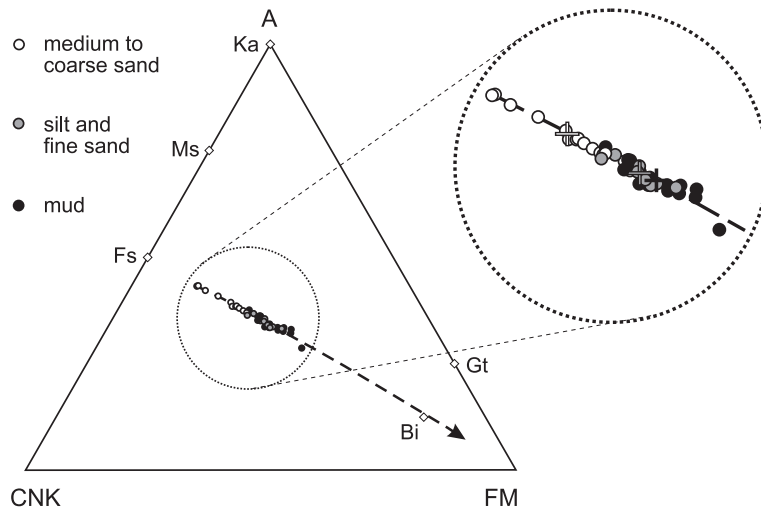
Fig. 6. A–CNK–FM ternary diagram (amalgamated three-part simplex) of the bulk chemical composition of Guys Bight Basin sediments (A=Al$_2$O$_3$, CNK=CaO+Na$_2$O+K$_2$O, FM=FeO$_{tot}$+MgO; all in molar proportions). Data were taken from Nesbitt and Young (1996). The stippled line shows the adjusted compositional linear trend with starting composition given by the mean of the two samples lowest in FM. Cross signatures indicate mean of each grain size group. The trend clearly indicates an increase of FM (i.e., biotite) with decreasing grain size. Note positions of selected mineral compositions (Fs=feldspars, Bi=biotite, Gt=garnet, Ms=muscovite, Ka=kaolinite).

remarkably (Fig. 6). This means, that feldspar content does not vary appreciably with grain size and there is also no marked clay mineral enrichment in the muddy samples as it would be usually the case in environments significantly affected by chemical weathering processes. The increase of FeO$_{tot}$+MgO in fine-grained samples is caused by preferential enrichment of biotite due to hydraulic sorting (Nesbitt and Young, 1996).

The adjusted compositional linear trend in the three-part simplex A–CNK–FM clearly reflects this increase of biotite with decreasing grain size (Fig. 6). The trend almost follows a straight line from feldspar composition (Fs) to the FM-pole of the diagram, implying that there is no Al enrichment at the expense of Ca, Na, and K (CNK); that means, no alteration of feldspar. The compositional linear trend explains over 99% of the total variability. This suggests, in principle, that the interdependence of grain size and chemical composition can be precisely modelled for the case that physical processes alone control the grain size of the sediment. In the present example, however, grain size classification of the samples is not detailed enough to constrain this relation in a sufficiently precise quantitative manner. Additional data on the chemical composition of a range of precisely sepa-

rated grain size fractions from a similar case study are needed to achieve a precise quantitative description of this relation.

4. Discussion and outlook

Trends observed in compositional data of soils, sediments, or sedimentary rocks can be quantified using the proposed approach of adjusting compositional linear trends in the simplex. Although the case studies used in this paper are restricted to three-part compositions for the reason of better visualization of the results, the approach can be applied to compositional data of any dimension. Once a compositional linear trend that is related to a specific process is quantified, the method provides a useful tool for (1) evaluating if this specific process is responsible for an observed compositional trend or not (2) quantifying to what extent this process was active to produce a certain sediment composition, (3) testing the relationship between this quantitative measure (the t-value) and external parameters considered to exert control on the process, and (4) evaluating the combined effects of different processes acting together or consecutively.

The latter point still needs some discussion. Combining numerically calibrated compositional trends that are each related to a specific process within the sedimentary cycle potentially allows for the description of more complex systems generating certain sediment compositions. For example, case study no. 3 (Section 3.3: grain size vs. sediment composition) still lacks sufficient quantification of the interdependence between chemical composition and physically controlled grain size, but it can be anticipated that, in principle, after solving the problem of the poor grain size resolution, the combination of the results from case study no. 1 (Section 3.1: chemical weathering) and no. 3 will allow for modelling bulk chemical compositions of sediments affected by both chemical and physical processes. If so, the relative contribution of each of the processes to the resulting sediment composition can also be quantified. Of course, some additional factors may complicate this simple scenario, for example, physical comminution and sorting may vary with changing source-rock characteristics and depositional regimes. Further complications arise from the fact that statistical mixing of two single-process compositional trends does not necessarily give the same results as the concurrent operation of these two processes. For example, chemical and physical weathering that operates simultaneously may produce feedback effects that are not covered by the simple mixing or additive accumulation of the individual end-member processes. However, such feedback effects should not change the direction of the (cumulative) trend, but may accelerate the degree of compositional change induced by the processes and, therefore, will affect only the t-value of the resulting composition linear trend and not the perturbation vector $\mathbf{p}$. If the two processes act consecutively, like the example of chemical weathering followed by later K-metasomatism (Fedo et al., 1995; von Eynatten et al., 2003) the final compositions should not be affected by such feedback mechanisms and, hence, the statistical mixing of the two single processes should allow for calculating reasonable results.

Although numerical or statistical modelling has become a well-known tool in several fields of geosciences, for example, stratigraphic modelling and basin modelling in the field of sedimentology, little has been done so far concerning a comprehensive numerical model for describing the composition of clastic sediments. Such a model would allow for (1) predicting sediment compositions under known conditions, i.e., topography, climate, depositional environment, diagenesis, anthropogenic factors, etc., and (2) reconstructing provenance characteristics of a given sediment or sedimentary rock in a much more quantitative manner than we can do these days. The statistical approach proposed in this paper may prove helpful on the way towards a quantitative description of sediment-forming processes.

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References

Aitchison, J., 1986. The Statistical Analysis of Compositional Data. Chapman and Hall, London. 416 pp.

Aitchison, J., 1992. On criteria for measures of compositional difference. Mathematical Geology 24, 365–379.

Aitchison, J., Thomas, C.W., 1998. Differential perturbation processes: a tool for the study of compositional processes. In: Buccianti, A., Nardi, G., Potenza, R. (Eds.), Proceedings of IAMG'98—The Fourth Annual Conference of the International Association for Mathematical Geology, De Frede, Napoli, pp. 499–504.

Butler, J.C., 1979. Trends in ternary petrologic variation diagrams— fact or fantasy? American Mineralogist 64, 1115–1121.

Chayes, F., 1960. On correlation between variables of constant sum. Journal of Geophysical Research 65, 4185–4193.

Cox, R., Lowe, D.R., 1995. A conceptual review of regional-scale controls on the composition of clastic sediment and the co-evolution of continental blocks and their sedimentary. Journal of Sedimentary Research. Section A, Sedimentary Petrology and Processes 65, 1–12.

Dorsey, R.J., 1988. Provenance evolution and unroofing history of a modern arc-continent collision: evidence from petrography of Plio-Pleistocene sandstones, eastern Taiwan. Journal of Sedimentary Petrology 58, 208–218.

Fedo, C.M., Nesbitt, H.W., Young, G.M., 1995. Unravelling the effects of potassium metasomatism in sedimentary rocks and paleosoils, with implications for paleoweathering and provenance. Geology 23, 921–924.

Grantham, J.H., Velbel, M.A., 1988. The influence of climate and topography on rock-fragment abundance in modern fluvial sands of the southern Blue Ridge Mountains, North Carolina. Journal of Sedimentary Research 58, 219–227.

Heins, W.A., 1993. Source rock texture versus climate and topography as controls on the composition of modern, plutoniclastic sand. Special Paper-Geological Society of America 284, 135–146.

Johnsson, M.J., 1993. The system controlling the composition of clastic sediments. Special Paper-Geological Society of America 284, 1–19.

McLennan, S.M., 1993. Weathering and global denudation. Journal of Geology 101, 295–303.

Nesbitt, H.W., Young, G.M., 1982. Early Proterozoic climates and plate motions inferred from major element chemistry of lutites. Nature 299, 715–717.

Nesbitt, H.W., Young, G.M., 1996. Petrogenesis of sediments in the absence of chemical weathering: effects of abrasion and sorting on bulk composition and mineralogy. Sedimentology 43, 341–358.

Nesbitt, H.W., Fedo, C.M., Young, G.M., 1997. Quartz and feldspar stability, steady and non-steady-state weathering, and petrogenesis of siliciclastic sands and muds. Journal of Geology 105, 173–191.

Pawlowsky-Glahn, V., Egozcue, J.J., 2002. BLU estimators and compositional data. Mathematical Geology 34, 259–274.

Pettijohn, F.J., Potter, P.E., Siever, R., 1972. Sand and Sandstone. Springer, New York. 618 pp.

Taylor, S.R., McLennan, S.M., 1985. The Continental Crust: Its Composition and Evolution. Blackwell Science, Oxford. 315 pp.

von Eynatten, H., Pawlowsky-Glahn, V., Egozcue, J.J., 2002. Understanding perturbation on the simplex: a simple method to better visualise and interpret compositional data in ternary diagrams. Mathematical Geology 34, 249–257.

von Eynatten, H., Barceló-Vidal, C., Pawlowsky-Glahn, V., 2003. Modelling compositional change: the example of chemical weathering of granitoid rocks. Mathematical Geology 35, 231–251.