

Arbitrary scaling in ISOCON method of geochemical mass balance: An evaluation of the graphical approach

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Rock alteration processes can be studied through geochemical Mass Balance (MB) estimation in several ways. Due to the ease of its adaptability, especially its graphical approach, the ISOCON method of mass balance is extensively used for such studies. However the technique suffers from a serious limitation, in particular with reference to the graphical treatment involving arbitrary scaling, which is explained in this paper with the help of illustrative examples. The reference frame for MB obtained by best fit regression is biased to the element scaled upward that plot away from the origin. The scaling factors actually act as weighting factors which bias the slope of the ISOCON line because it is controlled by data points having higher numerical value in the plot. Since scaling of concentration data are unavoidable in ISOCON analysis, the results of MB may potentially be misleading. The slope of the best fit ISOCON line may vary within the range of slopes defined by individual conserved elements. In case, no scaling is done, the ISOCON shall weight in proportion to the abundance of the immobile elements. The co-linearity of the immobile elements with origin is also subjective in nature with the elements near origin apparently seem to satisfy a wide range ISOCON slopes than those that are away from origin. When equal weight is assigned to all elements, regardless of their concentration level, an average of the slopes of the immobile elements (i.e., concentration ratios in altered to unaltered) may provide better approximation for reference frame without recourse to graphical plot and ISOCON solution. The existing weighted least-squares or equal weight least-squares ISOCON method is found to be more appropriate in case of higher uncertainty in the protolith compositions. In any case, a proper identification of the conserved species is an essential prerequisite to successful implementation of mass balance computation minimizing the inherent errors of the ISOCON technique.

Keywords: least squares, regression, volume factor, alteration, mass balance

INTRODUCTION

The foundation of the present form of geochemical mass balance was laid by Gresens (1967) who put forward the inter-relationship of change in volume, composition and density of an altered rock to its protolith. This is universally applicable to all alteration processes when the protolith of the alteration product is known. However, Grant (1986, 2005) proposed a revamped version of Gresens' mass balance equation substituting volume and density by mass, allowing a direct comparison of mass transfer to composition of the protolith and to the altered rock through the ISOCON diagram. In this diagram the elemental abundances of altered and unaltered protolith are plotted on abscissa and ordinate respectively. One of the most crucial prerequisite is to define a reference frame with respect to which the mass-transfer computations are

made. For all practical purpose, it is the immobile species that provide most reasonable reference frame. Grant (1986) proposed that all immobile elements would plot on a straight line through origin in ISOCON plot. Line drawn through these elemental plots is referred to as isochemical line (line of no mass-transfer) or simply as ISOCON line. It provides an illustrative graphical solution to mass balance depicting the elements that are lost, gained or conserved depending upon its plot to the right, left or on the ISOCON line respectively. Due to its simplicity and adaptability, ISOCON method is being extensively used in diverse fields by the researchers (Grant, 2005). Although the formulation and methodology is sound, there are certain ambiguities to its graphical approach resulting in its potential ill-treatment. The ambiguity arises from the arbitrary scaling of the compositional data to suite the ISOCON plot and analysis. The scaling influence the best fit ISOCON and hence the reference frame for mass-balance estimates. This has been first pointed out by Baumgartner and Olsen (1995) in an attempt to modify ISOCON method. Instead of arbitrary numbers, they proposed a rational basis to assign scaling

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factors that gives more weight to elements that show small uncertainty and *vice-versa*, thus minimizing the error arising due to the heterogeneity in protolith composition to some extent. Grant (2005), however, reiterated that scaling do not alter the slope of the ISOCON. In this technical brief we demonstrate the effect and consequences of scaling that introduce biasness and influence the mass-balance results. The limits of expected uncertainties varies from case to case basis and are illustrated through examples from published literature with the aim of cautioning the potential users about the possible pitfalls and limitations of the technique.

ISOCON METHOD OF MASS BALANCE

The ISOCON technique of Grant (1986) is a derivative of the basic mass balance equation of Gresens (1967) where the volume (V) and density (ρ) terms are merged and referred to as mass (M), i.e., mass = volume \times density; $M = V \times \rho$. The mass balance equation of Gresens, relating volume and composition, is given by

$$F_V \left(\frac{\rho^s}{\rho^p} \right) C_n^s - C_n^p = X_n. \quad (1)$$

Here, the superscript p and s refer to protolith (primary) and altered (secondary) rock respectively, C_n^s and C_n^p are the fractional concentration of element n in altered and unaltered rocks respectively, X_n is the amount of mass change (gained or lost) in an element n , and F_V is the overall volume factor (i.e., ratio of volume of the altered rock to that of the unaltered protolith ($F_V = V^s/V^p$)).

The volume strain defined by any immobile element k (F_V^0) can be obtained by substituting $X_n = 0$ in the above equation, as

$$F_V^0 = \frac{C_k^p}{C_k^s} \times \frac{\rho^p}{\rho^s}, \quad (2)$$

F_V^0 is the isochemical volume factor (ICVF) of element k .

Equation (1) can be rewritten as

$$\left(\frac{V^s}{V^p} \right) \left(\frac{\rho^s}{\rho^p} \right) C_n^s - C_n^p = X_n,$$

$$\left(\frac{M^s}{M^p} \right) C_n^s - C_n^p = X_n.$$

In case of immobile element k , the $X_n = 0$, yielding

$$C_k^s = \frac{M^p}{M^s} C_k^p. \quad (3)$$

This equation can be recast in standard notation as

$$Y = mX + C.$$

This is an equation of a straight line with slope $m (=M^p/M^s)$ passing through the origin ($C = 0$) in a plot of protolith ($X = C_k^p$) versus altered ($Y = C_k^s$) composition of immobile elements. This line is called the ISOCON (Grant, 1986). The slope of this line provides an estimate of the inverse of mass factor or enrichment factor (EF; C_k^s/C_k^p) as a function of the observed concentration change in conserved elements due to loss or gain in other mobile elements in any alteration process.

A reference frame with respect to which the mass transfer estimates are made need be defined. This reference frame could be a known mobility (X_n) of any element, immobility ($X_n = 0$) of one or more elements or a known volume factor (F_V) for the alteration process. Conventionally, the immobility of one or more elements is assumed as reference frame. The net mass transfer of the process is thus quantified with respect to the immobility of these assumed element(s). If we assume only a single element to be immobile, then the solution is straight forward and unique. However, when a number of elements are immobile, the resulting solution is non-unique because each of these elements defines a different reference frame. Under ideal conditions, all the solutions should converge to give a unique solution but in practice, it does not happen. In order to find an acceptable solution, an average value is taken as in case of Gresens (1967) or a best fit line is obtained as in case of Grant's (1986) ISOCON method or weighted least squares regression as in case of Baumgartner and Olsen (1995).

AMBIGUITIES IN ISOCON RESULTS

In any natural example, the identified or assumed concentrations of immobile elements when plotted in the ISOCON diagram do not fall strictly on a straight line and show some scatter. This scatter may result from (1) geochemical heterogeneity in the assumed protolith, (2) analytical error, and (3) departure from ideal immobility of the assumed elements. In view of this, Grant proposed that the ISOCON line through these elements and origin is drawn apparently using visual estimates or one can use mathematical least squares regression. Since, the magnitude of elemental concentrations in rocks varies widely; the concentrations are scaled to highlight their dispersion on the diagram. It is argued by Grant (1986) that such scaling is not only mathematically valid but it also gives

Table 1. Variation of ISOCON slopes as a function of scaling factor. The ISOCON element data are from Barnes *et al.* (2004) corresponding to unaltered host Zentralgneis (ST01-F) and altered shear zone sample (ST01-1B). The authors' factors are given in Scale factor-1.

	Unaltered	Altered	Not scaled	Scale factor-1	Scale factor-2	Slope (C^s/C^p)
TiO ₂ (%)	0.64	1.03	1.0	3.5	20	1.61
Al ₂ O ₃ (%)	14.97	19.33	1.0	2.5	1.6	1.29
Y (ppm)	20	19	1.0	1	2	0.95
Ni (ppm)	24	19	1.0	0.1	2	0.79
Cr (ppm)	45	55	1.0	0.125	0.4	1.22
ISOCON slope			1.22	1.16*	0.95	
r^2			0.97	0.93	0.22	
F_V			0.84	0.98	1.08	
Volume strain			-16%	-2%	+8%	

*Authors' reported this as 1.36; cf., table A.1, p. 299, Barnes *et al.* (2004).

due weight to the elements whose concentration magnitude is low. However, Baumgartner and Olsen (1995) argued and illustrated using a hypothetical example that how scaling can bias the slope of the ISOCON. In spite of this, Grant (2005) in his recent review article out-right rejected this mathematically proved fact giving an untenable argument that scaling is irrelevant to ISOCON slope because it is (C_i^s/C_i^p) a ratio and the scale factors would cancel out. This is far from convincing as the argument is true only in case the slope is defined by an individual element with respect to origin, but in the case of scatterplot of several immobile elements on an X-Y diagram, each immobile element defines a slope of its own with the origin. Therefore, the best fit optimum ISOCON thus obtained is no more a simple function of the ratios of their concentrations alone but is also functions of the magnitude too. Since in simple least squares regression analysis, the squared sum of differences (residue) of the estimated and observed Y values for given X values, is minimized, the slope of the best fit ISOCON line is bound to be influenced by the scaling factors. Due to the requirement of fitting a line through the origin, the scaling emphasizes the data points with higher numerical values than those with smaller numerical values. When a data point of an element is scaled forward by multiplying with an integer, the contribution of the element increases because the squared residue also becomes large. This was termed by Baumgartner and Olsen (1995) as torque exerted by data points on the visually or mathematically determined best fit ISOCON. This would mean that the best fit regression line shall become biased towards the elements that plot away from the origin compared to those near the origin. Thus, a *simple* least squares fit turns in to a weighted least squares regression. Even if the best fit is obtained by visual estimates, the eyes also act as least squares integrator. Since scaling is inevitable for obtaining graphical solution by ISOCON analysis, Baumgartner

and Olsen (1995) proposed a rational basis for scaling that would minimize the error due to uncertainties. They proposed to do this by scaling down (towards origin) the abundance of an element with larger uncertainties and scaling up (away from origin) the elements that are associated with smaller uncertainties such that the torque on the best fit ISOCON by least squares minimization becomes appropriately weighted and statistically more robust. It is reasonably a better option than arbitrary scaling, however, the scaling factor in this case is constrained by the reciprocal of the standard deviation and hence it may or may not yield the desired spread of the data points for ISOCON analysis. Therefore, the rigorous statistical and weighted least squares technique of Baumgartner and Olsen (1995) is more appropriate where large compositional heterogeneity in the samples is apparent.

ILLUSTRATIVE EXAMPLES

Baumgartner and Olsen (1995) demonstrated the influence of scaling on the ISOCON using a hypothetical example. In addition to the effect of scaling on the slope of ISOCON and hence on volume strain, the inadvertent misuse of the technique is illustrated here through examples from published literature. The first example consists of a shear zone sample and its protolith (ST01-F → ST01-1B; Barnes *et al.*, 2004) and the results are summarized in Table 1. The slope of the ISOCON through the selected elements is 1.22 without any scaling. When the same data are scaled (scale factor-1, author's choice) to move Al and Ti further away and to bring Ni and Cr towards the origin, the slope obtained is 1.16 and in the case of scale factor-2, slope is further reduced to 0.95. Thus, for the same set of conserved elements as reference frame, the ISOCONs predict different volume strains, i.e., a volume loss of 16% (unscaled) to a volume gain of 8% (scale factors-2; Table 1, Fig. 1). The corresponding effect on

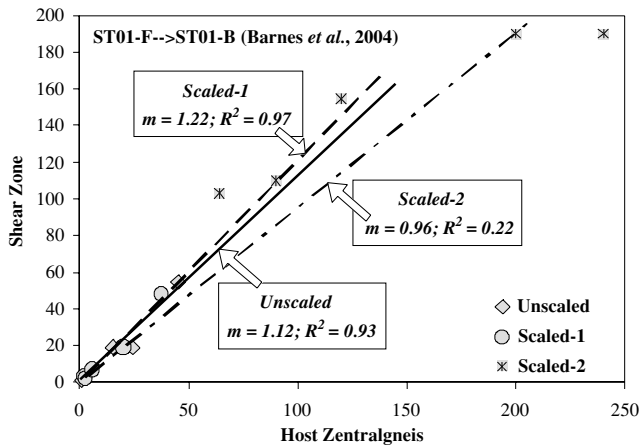


Fig. 1. ISOCON diagram showing influence of scaling factor on the slope of ISOCON during least squares regression fit. The example data are from Barnes *et al.* (2004). The least squares fit is obtained for original and two different scaling factors (Table 1) in case of samples ST01-F → ST01-B.

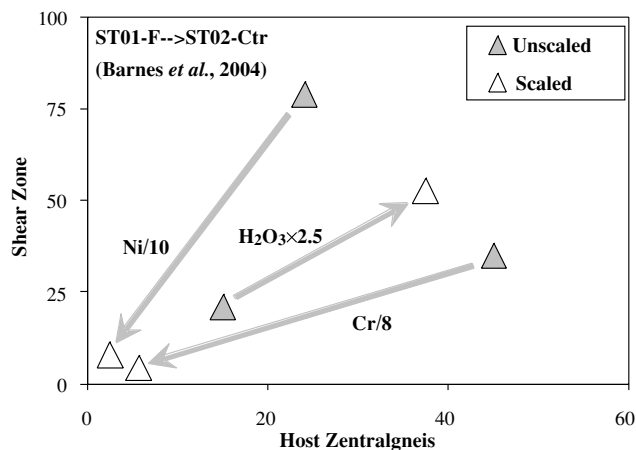


Fig. 2. Illustrative example (Barnes *et al.*, 2004) exhibiting how scaling can influence correlation coefficient of the least squares trend. The filled triangles are original data that too scattered to an ISOCON but the scaled data points (empty triangles) purportedly define a reasonable ISOCON good correlation coefficient.

mass-transfer estimates are summarized in Table 2. The amount enrichment or depletion of major components alters as much as by 4 times between the two extreme values. This example clearly demonstrates how different scaling factors influence the slope of the reference ISOCON.

The ambiguity resulting from different scaling factors and incorrect choice of immobile element is illustrated in another example of protomylonite (sample #A) to mylonite (sample #B) alteration case studied by Hippert

Table 2. Results of mass-balance computation showing effect of different scaling factors applied to the assumed immobile elements which results in varying ISOCON slope value (Fig. 1) and hence volume strain. Negative (and positive) sign refers to loss (and gain) in the respective component. The results are in grams with respect to per 100 gm of protolith for major elements and ppm in case of trace elements.

F_v	MB computations at different F_v values		
	$F_v = 0.84$	$F_v = 0.98$	$F_v = 1.08$
ISOCON slope	1.22	1.16	0.95
Volume strain	-16% loss	-2% loss (~isovolumetric)	+8% gain
SiO ₂	-26.51	-19.75	-14.92
TiO ₂	0.24	0.39	0.49
Al ₂ O ₃	1.57	4.33	6.30
FeOt	3.97	5.44	6.50
MnO	0.03	0.04	0.05
MgO	3.06	4.06	4.77
CaO	-0.12	0.34	0.66
Na ₂ O	-1.79	-1.48	-1.27
K ₂ O	1.52	2.03	2.40
P ₂ O ₅	0.17	0.21	0.24
Nb	-13.46	-10.04	-7.59
Zr	1.14	16.83	28.04
Y	-3.74	-1.03	0.91
Sr	-130.20	-108.23	-92.54
Ba	9.24	77.28	125.88
Rb	40.98	58.81	71.55
Co	46.77	55.90	62.42
Ni	-7.74	-5.03	-3.09
Cr	2.07	9.92	15.52
Cu	-57.86	-57.01	-56.40
Zn	86.25	121.63	146.89

(1998). Ti (in ppm) is scaled down by 1/30 and Zr by 1/4 allowing the author to draw visibly an attractive ISOCON (Fig. 3b). Note here that the isochemical volume factor for Zr is 1.76 and that of Ti is 2.15, corresponding to a broad range of volume gain of 76% and 115% respectively. The slope of the regression line through these two elements and origin is estimated at 0.44 for original data set while it is 0.48 and 1.19 for scaled factors 1 and 2 respectively (Fig. 3).

It is also possible to improve the correlation coefficient by applying suitable scaling factors. This is illustrated in another sample pair (ST01-F → ST02-Ctr) from Barnes *et al.* (2004). Actually the assumed elements (Al, Cr and Ni) are not co-linear with origin at all and hence do not qualify to define any ISOCON. Any attempt to draw a best fit line through these elements would result in a poor correlation coefficient value (slope = 1.34; $r^2 = -0.5491$; Fig. 2, filled triangles). Applying scaling factor of 2.5 for Al and 0.1 for Ni and 0.125 for Cr, the Al is shifted away from the origin and Ni and Cr are brought closer to the origin (Fig. 2, open triangles) leading to an apparently good looking ISOCON with slope = 1.40 and $r^2 = 0.98$. This is an example of inappropriate application

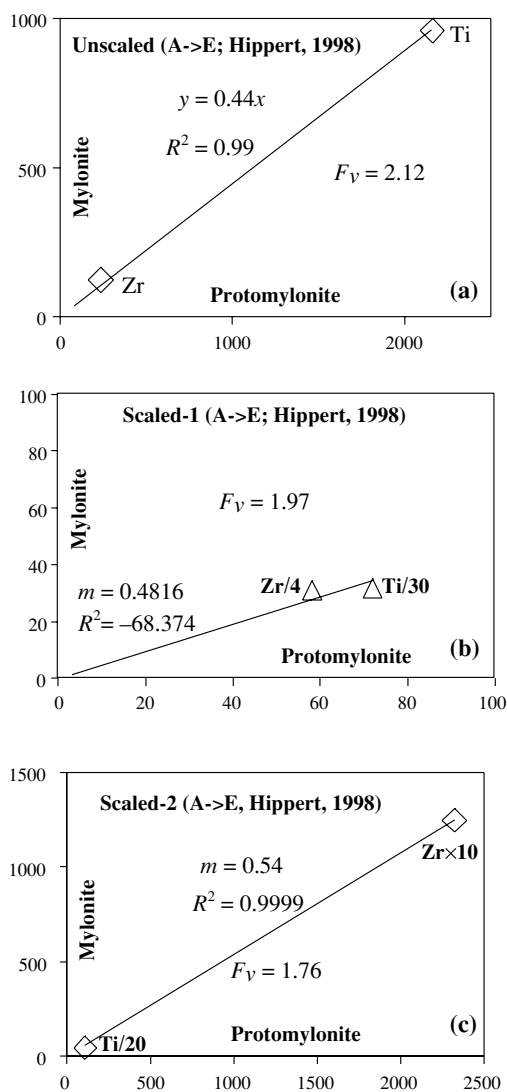


Fig. 3. Comparison of ISOCON slopes for (a) original, (b) scaled-1 and (c) scaled-2 data for sample #A (protomylonite) and E (mylonite) taken from Hippert (1998). The scaling factors are given in the figure. Scaled-1 is the author's choice of scaling factors. Note that the slopes are biased to the point further from the origin.

of ISOCON technique with many more such unintentional maltreatment that have probably gone unnoticed in literature wherein misleading interpretation of volume strain resulting from the arbitrary influence of scaling and wrong selection of immobile element.

DISCUSSION

The examples discussed above and that of Baumgartner and Olsen (1995) sufficiently reveal that the graphical ISOCON technique is likely to yield mislead-

ing results in certain cases. The scaling factors can change the slope of the ISOCON obtained by regression within the range of ratios (slope) of the immobile elements in unaltered to altered rock. The best fit line is intrinsically related to the magnitude of the plotted X-Y values and the discrete data points on the ISOCON diagram are not ratios but the scaled concentration values. Thus Grant's (2005) assertion that—"... if the slope of isocon is based on C_i^A/C_i^O values, scaling can not affect the results", is not practically tenable. It is true for an individual element, but when taken in totality where the reference frame is to be drawn using several immobile species, the slope of the ISOCON is not a ratio of any of these elements. In principle, this ratio as well as Isochemical volume factor (ICVF) should be identical and practically similar for all truly immobile elements and is concomitant with the mass/volume change. This ratio is also referred to as enrichment factor (EF; Ague, 1994; Brimhall and Dietrich, 1987). If the assumed elements were nearly conserved in an alteration process, and the assumption of the protolith is reasonably good, then the range of enrichment factors (slope) or ICVF defined individually by these elements should not vary substantially leaving little scope for error while obtaining an acceptable value of the slope. For example, the application of ISOCON by Spandler and Hermann (2006) judiciously constrained LREEs to be immobile resulting in a very narrow range of ICVF values (1.07 to 1.13) equivalent to range of volume dilation of only 7–13%. In this case even extreme scaling would not alter the results substantially. It follows that the choice of immobile elements is very crucial for successful implementation of ISOCON technique. The assumed sets of conserved elements by Barnes *et al.* (2004) and Hippert (1998) do not have coherent slope or ICVF values and is probably incorrect selection. Truly immobile elements are expected to have identical slopes, however in practice; they invariably show some degree of variation owing to analytical uncertainty, heterogeneity in protolith and lack of definitive evidence for immobility of an element. Srivastava *et al.* (2004) observed that the assumed immobile elements (Sc, Nb, Y and Ti) define a wide range of volume strain (18–64%). In order to avoid any misinterpretation, they preferred to report these two limiting values of volume strain and corresponding mass balance as range, rather than forcing a single best fit ISOCON reference frame through such scattered immobile element plot.

The application of ISOCON diagram for identification of immobile elements is based on co-linear plot of the elements along a line through the origin. However, as discussed above, manipulability of the co-linear relationship of elemental plot by appropriate scaling seriously limits the purpose. As shown in the example and illustrated in Fig. 2, a well-scattered array of data points can

apparently be forced to lie on linear array allowing a reasonably good-looking ISOCON to be drawn. These errors may frequently occur because it is an artefact of arbitrary scaling and an inherent limitation of ISOCON method itself especially where the immobile elements are not well constrained though independent criteria. In the worked out example (using data from Mori *et al.*, 2003), Grant (2005) proposed an ISOCON through Al_2O_3 , SiO_2 , TiO_2 , Cu and Sc. However, judicious scrutiny would reveal surprise selection of Sc in preference to V, in spite of the fact that the slope of V is more close to the chosen cluster of immobile elements in comparison to Sc. Sc being plotted relatively nearer to the origin (compared to V), apparently looks more close to the proposed ISOCON. Though the error involved in this specific case may not be significant, nevertheless, it may become a potential source of unforced error in mass-balance estimation.

By scaling the concentrations, weighted least squares regression is invoked without any rational basis for employing different weighting factors for different conserved elements. In case no scaling is done and same concentration unit is followed, the ISOCON diagram would yield a mass balance that would again be weighted according to relative abundance of the elements. More abundant element would have proportionately higher influence. This is perhaps more acceptable proposition, because the major elements control the ultimate volume strain, provided the data plots for immobile elements are sufficiently spread over allowing ISOCON analysis. Even the weighted least squares ISOCON technique (Baumgartner and Olsen, 1995) would logically perform much better than such arbitrary scaling. An innovative way of data presentation in ISOCON diagram was proposed by Humphris *et al.* (1998) in which the elements were scaled down in such a way that the squared sum of values for altered and unaltered rock become unity, allowing the elements to plot along an arc of a circle centered on the origin at a uniform distance of 1 unit. This not only removes the visual effects of arbitrary scaling but it gives equal weight to all elements. It also helps in the identification of immobile reference frame through the point groups of elements (clusters) behaving similarly having similar ISOCON slopes. In such modified ISOCON diagram, no best fit linear regression would be required and a central value such as average of the slopes of suite of elements may be used as representative of the reference frame for mass balance computations. Thus graphical approach of Grant (1986, 2005) is unneeded as slope of the ISOCON can easily be calculated as the average of the concentration ratios once the cluster of elements is identified. In the method of Grant, the key reference parameter for mass balance estimation is the slope of ISOCON (or EF; enrichment ratio) while in the method of Gresens, it is the isochemical volume factor (ICVF;

$F_V^0 = (C_n^p/C_n^s) (\rho^p/\rho^s)$). As a first approximation, following Gresens (1967), one can simply use average, median or the central point of the EF or ICVF values of the clustered elements as a rational reference frame without taking recourse to graphically manoeuvring a central value by least squares best fit ISOCON.

CONCLUSIONS

The ISOCON technique offers an alternative representation of Gresens' mass balance equation in terms of mass ratio of alteration process instead of volume strain. However, mass balance solution obtained through graphical method of least squares fitting applying indiscriminate scaling factors is inappropriate. The co-linearity of the immobile elements with origin gets exaggerated by undue scaling leading to wrong choice of conserved elements. Contrary to the Grant's (2005) claim that scaling does not affect the slope of the ISOCON, it is convincingly established and illustrated that the claim is theoretically not true and practically bias the mass-balance results appreciably. Scaling factors are in fact weighting factors assigned to the elements that influence the slope of the best fit ISOCON line. Thus identification of immobile elements through the co-linearity of the element plot in ISOCON diagram is debatable. Since it is not possible to visualize the ISOCON without appropriate dispersion of the element plots (i.e., through scaling), the only alternative is that the ISOCON must either be drawn prior to scaling such that an element with higher abundance is proportionately weighted or the equal-weighted least squares ISOCON method (Humphris *et al.*, 1998) or that of simple weighted least squares method (Baumgartner and Olsen, 1995) of scaling may be adopted that has a rational basis for weighting the elements and are statistically more robust in case where significant heterogeneity in protolith and altered rock is expected. In case all the identified or assumed immobile elements are to be given equal weight, a more sensible way is to take the average of the slopes (EF) or ICVF of these elements to represent the reference frame for mass balance assessment without recourse to the uncertain graphical treatment. The immobile elements in this case may be identified by the close cluster of calculated FE or ICVF values that can be rearranged in ascending order in a table for ease. Thus a revamped version of the Gresens' (1967) original method may provide a better approximation of the reference frame for mass-balance computation.

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