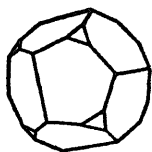


# On 'spurious' correlations in Rb–Sr isochron diagrams

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A general null hypothesis for isotope geochemistry states that the isotopic composition of an element is independent of its concentration or any other geochemical property of the population. 'Spurious' correlations between the ratios  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $^{87}\text{Rb}/^{86}\text{Sr}$  (or  $\text{Rb}/\text{Sr}$ ), due to the common denominator effect, may be ruled out because they contradict this null hypothesis. Mixing processes may be regarded as geochemical counterparts of the common denominator effect. In geochronological systems which satisfy the basic assumptions of the Rb–Sr isochron method observed correlations between Sr isotope abundance and Sr concentration must be secondary to the direct causal dependence of  $^{87}\text{Sr}/^{86}\text{Sr}$  upon  $\text{Rb}/\text{Sr}$  ratios.

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The Rb–Sr isochron method of dating, in which  $^{87}\text{Sr}/^{86}\text{Sr}$  is plotted against  $^{87}\text{Rb}/^{86}\text{Sr}$  for a suite of cogenetic samples, was put forward by H. L. Allsopp (1961) as a simple method of taking account of uncertainty about the isotopic composition of the 'common' strontium present in the samples. As such it is the chief workhorse of many geochronological laboratories, and has been extended by analogy to Sm–Nd dating as well as to the U–Pb and K–Ar methods on occasion. In all cases, provided the samples conform to certain conditions, a colinear array of data points is obtained whose slope yields the age and whose intercept gives the isotopic abundance of the radiogenic Sr (or Nd or Pb or Ar) at that time.

The development of strontium isotope geochemistry as a petrological tool subsequently led to the discovery that in various suites of supposedly cogenetic volcanic rocks the initial  $^{87}\text{Sr}/^{86}\text{Sr}$  was not uniform, as a simple petrogenetic model might have suggested, but varied significantly and was correlated with  $^{87}\text{Rb}/^{86}\text{Sr}$  (Dickinson et al. 1969; Bell & Powell 1969). Such relationships, though not universal (see for example, Bell & Powell 1970), were soon found to be very widespread, and were reviewed in two controversial papers (Brooks et al. 1976a & b). Both papers suggested that the isochron-like relationships found for so many young volcanic rocks reflected the chronology of events in their source regions; indeed, the authors emphasised this point by coining the term 'mantle isochrons'.

Besides arousing a good deal of argument amongst isotope geochemists, these papers pro-

voked a vigorous attack by Chayes (1977) on the statistical approach employed by their authors. The burden of Chayes' argument was that the observed correlations may be partly or wholly caused by 'common element bias': that is, they may be merely a consequence of two ratios sharing a common denominator, and have no geochemical significance. Replying to these criticisms, Brooks et al. (1977) leaned heavily on Monte Carlo numerical simulations which suggested that the effect was very small but failed to deal in detail with the substance of Chayes' criticisms. Butler (1982), in a companion paper to this, has returned to the topic with additional Monte Carlo experiments based on published Rb–Sr geochronological data, and concludes that, for data sets in which the ratios  $^{87}\text{Rb}/^{86}\text{Sr}$  and  $^{87}\text{Sr}/^{86}\text{Sr}$  are measured directly (rather than inferred from independently measured numerators and denominator), the common denominator effect can 'neither be assessed nor dismissed'. He argues that proper analysis, using Pearson's (1896) treatment of spurious correlations between ratios with a common denominator, is only possible with a complete knowledge of the various correlation coefficients and coefficients of variation of the two numerators and the denominator.

Some isotope geologists might be pleased to have available statistical ammunition with which to attack the controversial 'mantle isochrons' of Brooks et al. (1976a, b). Few, however, will understand how the well-tried Rb–Sr isochron method of dating can possibly be so fundamentally invalid. Indeed, the results of the statistical

simulations conducted by Butler (1982), yielding as they do 'ages' up to fifty times greater than the age calculated from the experimental data, suggest that his statistical treatment may contain fundamental flaws.

It is the purpose of this note, written from an isotope geologist's viewpoint, to suggest where those flaws may originate and to outline an alternative approach to the problem posed by Chayes and Butler. Isotope geochemistry and isochron dating are however considered separately, in the belief that the questions to be answered in the two cases are quite distinct. In isochron dating the correlations expected are nearly perfect, and samples are selected to give the best possible accuracy in the regression coefficients (slope and intercept). In isotope geochemistry, on the other hand, one is looking for correlations in situations where they are not necessarily to be expected, and samples are, or should be, randomly chosen.

One possible source of confusion touched upon by both Chayes and Butler should be clarified. In isochron dating  $^{87}\text{Rb}/^{86}\text{Sr}$  ratios are used for the independent variable, without exception. Isotope geochemists sometimes use instead the element ratio  $\text{Rb}/\text{Sr}$ . Although this quantity is not strictly proportional to  $^{87}\text{Rb}/^{86}\text{Sr}$ , because of variations in the isotopic composition of strontium, the relevant variations in common Sr are small enough that deviations from proportionality may be ignored, being insignificant in comparison with analytical uncertainties.

## The origin of the common denominator effect

Pearson (1896) first recognised the possibility of spurious correlations in the context of simple anatomical measurements. A pair of ratios used as indices of the shape of organs (e.g. length/breadth and depth/breadth of skulls) could be strongly correlated even if the three variates were completely independent. Considering three variates  $x_1$ ,  $x_2$  and  $x_3$ , the effect is qualitatively easily understood if we remember that ratios  $x_1/x_2$  and  $x_3/x_2$  will vary together with variations in  $x_2$ : if, then, the coefficients of variation of  $x_1$  and  $x_3$  are much less than that of the common denominator  $x_2$ , the main source of variation in the two ratios will be variation in  $x_2$ , so they will be strongly correlated even though the original three variates are completely independent; cer-

tainly the correlation between two ratios tells us nothing directly about the relationship between their numerators  $x_1$  and  $x_3$ . Similarly, according to Chayes and Butler, variations in  $^{86}\text{Sr}$  may be responsible for correlations between  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $^{87}\text{Rb}/^{86}\text{Sr}$  which isotope geologists wrongly attribute to the association of radiogenic strontium-87 with its parent rubidium-87.

I believe that this statistical argument is at best a half-truth, and misses one fundamental point about isotopic measurements: Isotopic ratios such as  $^{87}\text{Sr}/^{86}\text{Sr}$  are never calculated from independent measurements of the concentration of each of two isotopes in a rock or mineral sample: they are directly measured properties of the element under consideration, unrelated to the amount sampled, and can only be altered in a strictly limited number of ways. Any chemical process which alters the concentration of an element such as strontium affects all isotopes equally, except for small mass fractionation effects which in the case of Sr are eliminated by the standard normalisation procedure.

Statisticians may therefore like to consider the following null hypothesis for isotope geochemistry: *variations in the measured isotopic composition of an element are unrelated to its concentration, or to any other petrochemical property of the materials sampled.* In effect this statement denies the possibility of spurious, geochemically meaningless correlations between an isotope ratio and an element ratio with the same denominator: formal analysis, given below, appears to confirm that the expected value of the ratio correlation coefficient, if the null hypothesis is true, is zero. Significant correlations must therefore be related to one of the three types of process which can alter isotopic composition: mass fractionation, nuclear reactions (including radioactive decay) and mixing of two or more samples with different isotopic compositions. The third of these processes can perhaps be linked directly to the common denominator effect.

## Formal analysis

Using the notation of Butler (1982) we may write  $V_i = ^{87}\text{Sr}/^{86}\text{Sr} = x_1/x_2$ , and  $V_j = ^{87}\text{Rb}/^{86}\text{Sr} = x_3/x_2$ . Let  $c_i$  and  $c_j$  be the coefficients of variation of  $V_i$  and  $V_j$  and  $c_1$ ,  $c_2$ ,  $c_3$  those of  $x_1$ ,  $x_2$ ,  $x_3$ . From the null hypothesis of isotope geochemistry  $V_i$  is independent of  $x_2$ , so since  $x_1 = V_i x_2$  we have,

for small enough values of the coefficients of variation,

$$c_1^2 = c_2^2 + c_i^2 \quad (1)$$

We can now find the correlation coefficient  $r_{12}$  between  $^{87}\text{Sr}$  and  $^{86}\text{Sr}$  from Pearson's equation iii (1896), which becomes in the present notation:

$$c_i^2 = c_1^2 + c_2^2 - 2r_{12}c_1c_2 \quad (2)$$

so that, from (1) and (2)

$$r_{12} = c_2/c_1 \quad (3)$$

With a shared denominator,  $x_2$ , Pearson's equation for  $r_{ij}$ , the coefficient of correlation between  $V_i$  and  $V_j$  is:

$$r_{ij} = \frac{r_{13}c_1c_3 - r_{12}c_1c_2 - r_{23}c_2c_3 + c_2^2}{(c_1^2 + c_2^2 - 2c_1c_2r_{12})^{1/2}(c_3^2 + c_2^2 - 2c_3c_2r_{32})^{1/2}} \quad (4)$$

Taking into account eq. (3), and setting  $r_{13} = r_{23} = 0$  (no correlation between  $^{87}\text{Rb}$  and either strontium isotope) we find immediately

$$r_{ij} = 0 \quad (5)$$

The treatment is slightly more complicated if the other ratio,  $^{87}\text{Rb}/^{86}\text{Sr}$ , is also independent of its denominator. We then have by analogy with eq. (3)

$$r_{32} = c_2/c_3 \quad (6)$$

so that  $r_{ij}$  becomes

$$r_{ij} = \frac{r_{13}c_1c_3 - c_2^2}{(c_1^2 + c_2^2)^{1/2}(c_3^2 + c_2^2)^{1/2}} \quad (7)$$

The question immediately arises, what is the value of  $r_{13}$ ? Both  $x_1$  and  $x_3$  are correlated with  $x_2$ , so one would not expect it to be zero. Fisher (1958:188) gives an equation for partial correlation which states that the independent correlation of  $x_1$  and  $x_3$  in these circumstances, after excluding the effect of  $x_2$ , is

$$r_{13.2} = \frac{r_{13} - r_{12}r_{32}}{(1 - r_{12}^2)^{1/2}(1 - r_{32}^2)^{1/2}} \quad (8)$$

Hence if there is no independent source of correlation between  $x_1$  and  $x_3$  we have

$$r_{13} - r_{12}r_{32} = 0 \quad (9)$$

so from eqs. (3) and (5)

$$r_{13} = c_2^2/c_1c_3 \quad (10)$$

and  $r_{ij}$  is again zero.

These results support the intuitive notion that 'common element bias' will not occur if either or both of the ratios is independent of its denominator, and reinforce the proposition that the 'null hypothesis of isotope geochemistry' implicitly excludes spurious correlation due to the common denominator effect.

If  $^{87}\text{Sr}/^{86}\text{Sr}$  is correlated with  $^{86}\text{Sr}$  concentration, we can immediately see two possible geochemical causes. The first is mixing. A number of authors (e.g. Langmuir et al. 1978) have shown that a perfect linear correlation between  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $1/^{86}\text{Sr}$  will be found for a suite of samples formed by random mixing of two uniform sources of strontium with different isotopic composition. A plausible generalisation is that significant correlation will be found for random mixing amongst a population of strontium sources with a bimodal distribution of  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios. Faulty analytical techniques might create such a distribution, thus leading to spurious correlations due to blank problems. Some early Rb-Sr work on samples with low Sr concentrations may suffer from this problem, and quite a number of argon isochrons are probably spurious for similar reasons (Roddick 1978). However, in most Sr-isotope work on volcanic rocks analytical problems are not serious, and petrogenetic explanations must be sought for observed correlations. Mixing between different sources is an obvious candidate, and in fact is considered by Brooks et al. in connection with their mantle isochrons for continental volcanic rocks. They argue that, although the strong correlation between  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $1/^{86}\text{Sr}$  supports mixing, it is precluded by the lack of correlation between Rb and Sr. However, this conclusion would only be correct for two sources each uniform in Rb and Sr concentrations as well as in  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios. If the source(s) with high  $^{87}\text{Sr}/^{86}\text{Sr}$  have high  $^{87}\text{Rb}/^{86}\text{Sr}$ , irrespective of the concentrations of Rb and Sr, then a mixing process can certainly produce the observed correlations.

Finally, we must consider the significance of correlations between  $^{87}\text{Rb}$  and either of the strontium isotopes. According to equation (4) one or both could produce a ratio correlation. However, because of the strong correlations between  $^{87}\text{Sr}$  and  $^{86}\text{Sr}$  (see equation (3)) the two relevant coefficients  $r_{12}$  and  $r_{32}$  will be almost equal, as will the coefficients of variation, for the related terms in the numerator, so that the terms in  $r_{12}$  and  $r_{32}$  will almost exactly cancel out. Rigorous analysis has not yet been carried out,

but if  $^{87}\text{Sr}$  is thought of as consisting of a large constant 'common' component to which has been added a small, variable radiogenic component, it seems likely that the difference between  $r_{13}c_1c_3$  and  $r_{32}c_3c_2$  will be found to correspond to an association between  $^{87}\text{Rb}$  and radiogenic  $^{87}\text{Sr}$ .

## Artificial isochrons?

Application of the standard isochron equation of Rb–Sr geochronology,

$$\frac{{}^{87}\text{Sr}}{{}^{86}\text{Sr}} = \frac{{}^{87}\text{Sr}}{{}^{86}\text{Sr}_i} + \frac{{}^{87}\text{Rb}}{{}^{86}\text{Sr}} (e^{\lambda t} - 1)$$

(subscript  $i$  denotes initial ratio), depends on two assumptions: that all samples were cogenetic, having initially the same value of  $^{87}\text{Sr}/^{86}\text{Sr}$ ; and that no subsequent changes have occurred other than the decay of  $^{87}\text{Rb}$  to  $^{87}\text{Sr}$ . Together these assumptions imply perfect correlation between  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $^{87}\text{Rb}/^{86}\text{Sr}$ .

In the initial state of the system the value of  $^{87}\text{Sr}/^{86}\text{Sr}$ , being uniform, is obviously independent of the concentration of Sr. If the second assumption is also valid, and no movement of Sr in or out of the system has occurred, there is clearly no way in which 'local fluctuations in total Sr may tend to obscure the relationship between  $^{87}\text{Sr}$  and  $^{87}\text{Rb}$ ' (Chayes, quoted by Butler 1982). Of course, bad experimental techniques could add variable amounts of common Sr to the samples, resulting in a mixing line, but this is not a common problem. When the normal kinds of analytical error are taken into account, the correct statistical approach to Rb–Sr isochron dating is undoubtedly that pioneered by McIntyre et al. (1966) and York (1969), reviewed by Brooks et al. (1972): their 'null hypothesis' is that imperfections in correlation are due entirely to analytical errors; significant failure of that hypothesis denotes a failure in one or both of the basic assumptions.

There remains the possibility of a *secondary* correlation between  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $^{86}\text{Sr}$ . (Butler gives correlation coefficients of  $-0.928$  and  $-0.906$  for two sets of published data.) This need not be surprising, since high Rb/Sr ratios, leading to high  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios by the production of radiogenic Sr, are commonly associated with low Sr concentrations.

Finally, it should be noted that suites of sam-

ples for geochronological work normally are far from being randomly selected: given a population believed to satisfy the two assumptions stated above, the geochronologist always chooses his samples to include those with extreme values of Rb/Sr in order to maximise the precision of his results.

## Conclusion

It is proper that statisticians should draw the attention of isotope geologists to possible defects in their procedures. However, Butler's and Chayes' criticisms of isotope geology appear to be founded upon an inadequate assessment of the nature of isotopic measurements. In particular, Butler's approach to Rb–Sr geochronology is unlikely to help directly with the problems of Rb–Sr dating. In the field of isotope geochemistry, however, where sampling is supposed to be random, and observed correlations are used to test hypotheses concerning the source history, there is a need for further analysis of the statistical procedures. The arguments presented above suggest that it is wrong to think of the 'common denominator effect' in isotope geochemistry as an artefact of the method of calculation, as was the case in Pearson's anatomical examples. One should preferably test for its geochemical counterpart, a possible mixing process, by studying the correlations between each ratio and its denominator. Confusion will remain, however, until there is a clearer understanding of the relationships amongst the ten possible interdependent correlation coefficients amongst  $^{87}\text{Sr}$ ,  $^{86}\text{Sr}$ ,  $^{87}\text{Rb}$  and the two ratios. In the case of a 'good' Rb–Sr isochron it is plausible that the near perfect correlation between the ratios should be regarded as 'primary' and the anti-correlation between strontium isotopic composition and strontium abundance as 'secondary', but in isotope geochemistry the question is likely to be less clearcut, and it would be of value to have statistical procedures to enable us to decide which correlations are the most significant. If, for example, they were the correlations between each ratio and its denominator, it would seem plausible to suggest a mixing model; such may be the appropriate interpretation of many of the Brooks et al. 'mantle isochrons'. If, on the other hand, the correlation between the two ratios was more

significant, an interpretation involving association between  $^{87}\text{Rb}$  and associated radiogenic Sr would perhaps be more acceptable.

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