A method for inverting ratio-ratio data to estimate end-member compositions in mixing problems

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Abstract. I discuss the general problem of fitting mixing models to ratio-ratio data, and derive formulae for applying non-linear Maximum Likelihood methods for parameter estimation. To estimate mixing model parameters in the under-determined inversion it is necessary to introduce prior constraints, which I implement by penalizing the likelihood function for variations from a starting model. I illustrate practical aspects of the inverse problem by applying the method to synthetic data for a ternary system of putative mantle reservoirs using Sr, Nd, and Pb isotope ratios. I fit the synthetic data using two different starting models to demonstrate the sensitivity of the gradient method used to solve the non-linear inverse to the starting model and the necessity of inspecting the final model to avoid spurious results. I include *Matlab* scripts to facilitate starting model selection and to perform binary and ternary ratio-ratio inversions as an Electronic Appendix.

1. Introduction

Estimation of mixing end-members from compositional data is a common analytical problem in geochemistry. If the compositional parameters are ratios of elements or isotopes (i.e., ratio-ratio data), then the equation for the mixing trend, or surface, contains cross-terms resulting from differences in end-member concentrations of the ratio denominators (*Vollmer*, 1976). These cross-terms generate hyperbolic mixing surfaces in ratio-ratio parameter space, with the deviation from linearity being controlled by the denominator concentration ratios (e.g., *Langmuir et al*., 1978). Except for degenerate (linear) cases, which arise when the concentration ratios are all equal to unity, hyperbolic mixing surfaces have asymptotes that are parallel to the coordinate axes.

The mixing inverse problem for ratio-ratio data therefore requires fitting a hyperbolic surface to data. The dimension of the hyperbolic surface is equal to the dimension of the mixing model less one, such that binary models have 1-d surfaces, ternary models have 2-d surfaces, etc. The inversion is non-unique, or under-determined, because there are an arbitrarily large number of end-member compositions that give rise to the same hyperbolic surface (Figure 1). Least Squares (LS) methods may be used to estimate the asymptotes and scale factors that define the hyperbolic surface (*Albarede*, 1995), but not the mixing model parameters, themselves.

To estimate the mixing model parameters we must select from the range of possible solutions defined by the best-fitting hyperbolic surface. For physically plausible models with mixing proportions defined on the interval [0,1] the end-members must encapsulate the data, but the data are otherwise fit equally well by any set of end-members on the hyperbolic surface (Figure 1). In some cases the solution space can be constrained by chemical or geological arguments, but ultimately there will be a range of potential solutions that fit the data equally well from which to choose.

Non-linear Maximum Likelihood (ML) methods can be used to solve this type of problem (e.g., *Menke*, 1989; *Tarantola and Valette*, 1982) by specifying a starting model and then penalizing the inverse for variations from both the data and the starting model (*Sohn*, 2005). The non-linear inversion requires an iterative solution that converges at maxima in the likelihood function. This approach allows for estimation of the full set of mixing model parameters by incorporating prior information in the form an initial guess for the end-member compositions and then finding a solution that minimizes misfit to both the data and the starting model.

To this point treatments of the ratio-ratio mixing inverse problem have largely been limited to binary models. Many geochemical mixing problems, however, include more than two components, thus motivating extension of the inverse to higher-order systems. This work, for example, is motivated by the desire to use long-lived isotopes to study mixing of mantle reservoirs, and it has been recognized for some time that at least three, and quite possibly more, end-members are required to model the array of oceanic basalt (i.e., MORB and OIB) isotopic compositions (e.g., *Zindler et al*. 1982; *Zindler and Hart*, 1986; *Stracke et al*., 2005). In this paper I review the general problem of fitting mixing hyperbolas to ratio-ratio data, and derive formulae for inverting *n*-dimensional mixture data to obtain ML estimates of end-member compositions. I illustrate practical aspects of the ML method for higher-order models by applying it to synthetic Sr, Nd, and Pb isotope ratio data for a ternary system based on mixing of putative mantle reservoirs. *Matlab* scripts to perform the inversion for ternary ratio-ratio data are provided as an Electronic Appendix.

2. The general, *n*-dimensional, mixing hyperbola

The general, *n*-dimensional, ratio-ratio mixing equation is given by

$$x^j = \frac{\sum_{i=1}^{n} \phi_i a_i^j X_i^j}{\sum_{i=1}^{n} \phi_i a_i^j}, \qquad (1)$$

where X_i^j is the composition of end-member *i* for ratio *j* (e.g., $^{87}Sr/^{86}Sr$, La/Sm, etc.), a_i^j is the concentration of the denominator of ratio *j* in end-member *i*, ϕ_i is the mixing proportion of end-member *i*, and x^j is the sample (mixture) composition for ratio *j*. To conserve mass we also have

$$\sum_{i=1}^{n} \phi_i = 1. \qquad (2)$$

By rewriting Eq. (1) we obtain

$$\sum_{i=1}^{n} \phi_i a_i^j \left(x^j - X_i^j \right) = 0. \qquad (3)$$

For non-trivial solutions for the mixing proportions ϕ_i we must have

$$\det\left(a_i^j \left(x^j - X_i^j \right) \right) = 0, \qquad (4)$$

yielding the general expression for the *n*-dimensional, ratio-ratio, mixing surface with $2n^2$ parameters (n^2 for a_i^j and n^2 for X_i^j). We can reduce the number of parameters by expressing the equation in terms of the concentration ratios

$$K_{ij}^{kl} = \frac{\left(a_i^k / a_i^l \right)}{\left(a_j^k / a_j^l \right)} = \frac{a_i^k a_j^l}{a_j^k a_i^l}, \qquad (5)$$

which are related by two properties:

$$K_{ij}^{kl} = 1/K_{ji}^{kl} = 1/K_{ij}^{lk}, \qquad (6)$$

$$K_{ij}^{km} K_{ij}^{ml} = K_{ij}^{kl}. \qquad (7)$$

Note also that $K_{ii}^{kl} = K_{ij}^{kk} = 1$. There are thus $(n-1)^2$ independent concentration ratios, and the mixing equation can be rewritten by exploiting the fact that all of the concentration ratios can be defined by knowing K_{i1}^{k1} (a *n* by *n* matrix whose first row and column have entries equal to 1). From Equation (5) we have

$$a_i^j = K_{i1}^{j1} \frac{a_1^j a_i^1}{a_1^1}, \qquad (8)$$

and when this is substituted into Equation (4) we obtain

$$\det\left(K_{i1}^{j1}\left(x^j - X_i^j\right)\right) = 0 \qquad (9)$$

by using the matrix property that we can scale any row or column of $M_i^j = a_i^j\left(x^j - X_i^j\right)$ by any factor (i.e., $a_1^j a_i^1 / a_1^1$) and the determinant of the resulting matrix must still be zero.

Equation (9) represents the simplest and most general version of the *n*-dimensional hyperbolic mixing equation, and it has $2n^2 - 2n + 1$ free parameters (as opposed to $2n^2$ for Eq. 4): $(n-1)^2$ for K_{i1}^{j1} and n^2 for X_i^j. When $n = 2$ Eq. (9) yields the mixing hyperbola of *Vollmer* (1976), which can be expressed as

$$Ax^1x^2 + Bx^1 + Cx^2 + D = 0, \qquad (10)$$

where

$$
\begin{aligned}
A &= K - 1 \\
B &= X_2^2 - X_1^2 K \\
C &= X_1^1 - X_2^1 K \\
D &= X_1^2 X_2^1 K - X_2^2 X_1^1 .
\end{aligned}
\qquad (11)
$$

Note that $K = K_{21}^{21}$ is the only independent concentration ratio in the binary system.

The coefficient for the cross-term, *A*, goes to zero when *K* = 1 (i.e., the end-member concentrations of the elements in the denominator are equal), yielding a degenerate hyperbola (straight line). For $K \neq 1$ the mixing equation yields an equilateral hyperbola (*Albarede*, 1995), which can be seen by rearranging Eq (10) as follows. Assuming that $A \neq 0$ we can divide Eq (7) by *A* to obtain

$$x^1 x^2 + b x^1 + c x^2 + d = 0 \qquad (12)$$

where *b* = *B/A*, *c* = *C/A* , and *d* = *D/A*. This can be rewritten to yield

$$\left(x^1 + c\right)\left(x^2 + b\right) = bc - d, \qquad (13)$$

and relabeled to obtain the general form of an equilateral hyperbola

$$\left(x^1 - x_{\mathrm{inf}}^1\right)\left(x^2 - x_{\mathrm{inf}}^2\right) = h, \qquad (14)$$

where x_{inf}^1 and x_{inf}^2 are the asymptotes of the hyperbola, and *h* is a scale factor.

The equation for the equilateral hyperbola is of special interest as it highlights two important facts about the inverse problem of fitting mixing hyperbolas to ratio-ratio data. First, the mixing inversion is formally under-determined because there are $2n^2 - 2n + 1$ degrees of freedom (DOFs) in the mixing model (Eq. 9) but only $n^2 - n + 1$ DOFs (the number of independent minors of an n x n matrix) in the mixing surface. In the binary (n = 2) case, for example, there are 5 DOFs in the model ($X_1^1, X_1^2, X_2^1, X_2^2, K$), but only 3 DOFs in the hyperbolic equation ($x_{\mathrm{inf}}^1, x_{\mathrm{inf}}^2, h$). Second, the shape of a hyperbola is defined by its eccentricity, and the eccentricity of all equilateral hyperbolas is $\sqrt{2}$. Thus all mixing hyperbolas (except the degenerate straight line case) have the same shape, regardless of the model parameter values. Thus, while the position of the asymptotes and the scale factor may vary, the shape is fixed. The curvature of a binary mixing hyperbola varies continuously and approaches zero near the asymptotes. If the end-member ratios are fixed then the curvature at a given point is controlled by the concentration ratio (e.g., *Langmuir et al*., 1978), but the concentration ratio does not affect the shape of the hyperbola.

4. Formulation of the hyperbolic mixing inversion

As described above, there are $2n^2-2n+1$ DOFs in the mixing model (Eq. 9) but only n^2-n+1 DOFs in the mixing surface. The inverse problem is thus formally under-determined, and we cannot solve for the model parameters (i.e., end-member compositions) without the introduction of additional information. The only parameters that may be estimated from the data alone are the n^2-n+1 asymptotes and scale factor(s) of the n-dimensional hyperbolic mixing surface. The Least Squares method presented by *Albarede* (1995) can be extended to general, n-dimensional systems to yield estimates for these parameters.

In order to estimate mixing model parameters, as opposed to hyperbolic asymptotes and scale factors, it is necessary to introduce additional constraints to the inversion. ML methods are well-suited to this type of inverse problem (e.g., *Menke*, 1989; *Tarantola and Valette*, 1982), and *Sohn*

(2005) used this approach to derive an inversion for binary (i.e., n = 2) ratio-ratio mixing models. The ML approach can be extended to general, n-dimensional models as follows.

We begin by defining a data vector,

$$\mathbf{d} = \left[x_1^1, x_2^1, \ldots, x_N^1, x_1^2, x_2^2, \ldots, x_N^2, \ldots, x_1^n, x_2^n, \ldots, x_N^n \right]^{\mathrm{T}}, \qquad (15)$$

representing the n independent ratio observations from N samples ($N \geq n$). We then define the model vector,

$$\mathbf{m} = \left[X_1^1, X_2^1, \ldots, X_n^1, X_1^2, X_2^2, \ldots, X_n^2, \ldots, X_1^n, X_2^n, \ldots, X_n^n, K_{21}^{21}, \ldots, K_{21}^{n1}, \ldots, K_{n1}^{21}, \ldots, K_{n1}^{n1} \right]^{\mathrm{T}}, \qquad (16)$$

representing the $2n^2 - 2n + 1$ parameters in the mixing model. The data and model vectors are grouped into a single vector,

$$\mathbf{z} = \left[\mathbf{d}, \mathbf{m} \right], \qquad (17)$$

which has $n*N$ (data) + $2n^2 - 2n + 1$ (model parameter) rows.

To begin the iterative inversion we make initial guesses for the model parameters, $\mathbf{m}_0$, which are used to form the initial vector, $\mathbf{z}_0 = \left[\mathbf{d}, \mathbf{m}_0 \right]$. Assuming Gaussian distributions for the data and model, the prior distribution of $\mathbf{z}$ is

$$P(\mathbf{z}) = \exp\left[-1/2 \left(\mathbf{z} - \mathbf{z}_0 \right)^T \left[\operatorname{cov} \mathbf{z} \right]^{-1} \left(\mathbf{z} - \mathbf{z}_0 \right) \right]. \qquad (18)$$

The inversion is then carried out by maximizing Eq. (18) subject to the constraints of the mixing model (Eq. 9) $\mathbf{f}(\mathbf{z}) = \sum_{j=1}^{N} \det\left(K_{i1}^{k1}\left(x_j^k - X_i^k\right)\right) = 0$. This set of equations can be solved iteratively using Lagrange multipliers, yielding

$$\hat{\mathbf{z}}_{j+1} = \mathbf{z}_0 + [\mathrm{cov}\,\mathbf{z}]\mathbf{F}_j^{\mathrm{T}}\left(\mathbf{F}_j[\mathrm{cov}\,\mathbf{z}]\mathbf{F}_j^{\mathrm{T}}\right)^{-1}\left(\mathbf{F}_j\left[\hat{\mathbf{z}}_j - \mathbf{z}_0\right] - \mathbf{f}\left(\hat{\mathbf{z}}_j\right)\right) \quad (19)$$

where $\mathbf{F} = \nabla\mathbf{f}(\mathbf{z})$ is a gradient matrix. **F** has one row for each sample in the dataset and one column for each element of **z**, and is therefore an N x $(N-2)n + 2n^2 + 1$ matrix. The elements of **F** are defined by $\mathbf{F}_i^j = \partial f_i / \partial z_j$, which can be calculated using the formula for the differentiation of determinants

$$\frac{\partial}{\partial\alpha}\det\mathbf{A} = \mathrm{tr}\left(\mathrm{adj}(\mathbf{A})\frac{\partial\mathbf{A}}{\partial\alpha}\right). \quad (20)$$

If we set $A_i^k = K_{i1}^{k1}\left(x^k - X_i^k\right)$ and **B** = adj **A**, we then have

$$F_i^j = \frac{\partial f_i}{\partial x^p} = \left(\sum_{k=1}^{n} B_p^k K_{k1}^{p1}\right)\delta(i \bmod j, 0) \quad \text{for } 1 \le j \le \boldsymbol{n} \cdot N \quad (21)$$

$$F_i^j = \frac{\partial f_i}{\partial X_k^p} = -B_p^k K_{k1}^{p1} \quad \text{for } \boldsymbol{n} \cdot N + 1 \le j \le n(N+n) \quad (22)$$

$$F_i^j = \frac{\partial f_i}{\partial K_{k1}^{p1}} = B_p^k\left(x^p - X_k^p\right) \quad \text{for } n(N+n)+1 \le j \le n(N-2) + 2n^2 + 1 \quad (23)$$

where $\delta(i, j)$ is the Kronecker delta function with the property that $\delta(i, j) = 1$ for $i = j$, and $\delta(i, j) = 0$ for $i \neq j$.

The estimation procedure of Eq. 19 requires specification of the prior covariance matrix, $\mathrm{cov}\,\mathbf{z}_0$, which contains data and model parameter uncertainties, and has the effect of weighting the inversion. The misfit penalty for each element of $\mathbf{z}$ is inversely proportional to the prior variance (uncertainty) of the individual parameters. Note that the covariance matrix determines the degree to which the solution is penalized for variations from the starting model, and that each starting model parameter must explicitly be assigned an uncertainty. If we assume that the data and model are independent then the prior covariance matrix will be diagonal, but if prior knowledge regarding covariations in the data and model is available it can also be incorporated.

5. Solution of the hyperbolic mixing inversion

We obtain the posterior vector, $\hat{\mathbf{z}} = \left[\hat{\mathbf{d}}, \hat{\mathbf{m}}\right]$, when the iterative solution converges, with the elements of $\hat{\mathbf{m}}$ representing the model parameter estimates. Because of the non-linear nature of the inverse problem, it is possible for the method to converge on a local, as opposed to global, minimum in the solution space, and it is also possible that the solution will not converge at all. For these reasons it is necessary to carefully inspect the inverse results, and some amount of trial-and-error using different sets of initial guesses will usually be necessary to obtain the best results.

The goodness of fit of the model is expressed in terms of the likelihood function (Eq. 18), but the misfit of the posterior mixing model to the data can also be expressed as a Residual Sum of Squares (*RSS*) by summing the data residuals from the best-fitting hyperbolic surface,

$$RSS = \sum_{i=1}^{n} \left(r_{xi}^{\ 2} + r_{yi}^{\ 2} + r_{zi}^{\ 2} \right)^{1/2}, \qquad (24)$$

where r_{xi} is the residual of the i^{th} data point in the *x*-direction, etc. The residuals are determined by finding the minimum distance between each data point and the best-fitting hyperbolic surface, which is accomplished by finding the point on the hyperbolic surface with a normal vector passing through the data and calculating the Euclidean distance between the two points. This is also a non-linear problem that requires solving an iterative system of equations. If the *RSS* is normalized by the number of samples and the data uncertainty then we can obtain the Mean Square of Weighted Deviates (*MSWD*) (e.g., *Brooks et al*., 1972; *McIntyre et al*., 1966), which quantifies the misfit relative to the data error.

Estimation of model parameter uncertainties is problematic owing to the non-linear nature of the inverse and the fact that prior information must be introduced to solve the inversion. Non-parametric methods can be applied to address the first issue but the second issue is more problematic. There will always be a range of possible solutions for the end-member compositions on the (infinite) hyperbolic surface (e.g., Figure 1), and in this sense the parameter uncertainties are arbitrarily large. Nevertheless, parameter estimates require uncertainties, and to address this issue I use non-parametric methods to estimate uncertainties by exploring the likelihood function (Eq. 18) in the vicinity of the final solution. I use a modified version of the bootstrap method (e.g., *Efron and Tibshirani*, 1986) wherein the bootstrap replicates used to estimate error include random perturbations to the starting model as well as permutations of the data. The bootstrapped parameter estimates will thus incorporate the sensitivity of the solution to the starting model, albeit only within the neighborhood of the starting model. Uncertainty estimates derived in this way are conditional on the starting model, and are thus only valid within a small region of the solution space.

6. Application to test cases and discussion

I apply the method to a synthetic ternary dataset to illustrate practical aspects of higher-order mixing analyses. The synthetic dataset is based on the Sr, Nd, and Pb isotopes of oceanic basalts, which have been used to study mixing of long-lived mantle reservoirs (e.g., *Hart et al*., 1992; *Zindler and Hart*, 1986). I used the putative DMM, EM1, and EM2 mantle reservoirs as the three end-member components, with isotopic compositions drawn from *Zindler and Hart* (1986) (Table 1). I generated 100 synthetic mixtures with random end-member mass fractions and then added Gaussian noise to mimic geological variability and/or analytical uncertainty (see Electronic Appendix).

To demonstrate the sensitivity of the method to the starting model I begin the inverse with two different parameter sets. Both models have the same end-member ratio values, but in the first one all the concentration ratios are set to unity (such that the mixing surface is a plane), whereas in the second the concentration ratios have been adjusted (using the graphical interface included in the *Matlab* scripts) to better fit the data prior to the inversion (Table 1). In both cases the inverse is successful in fitting a hyperbolic surface to the data, but the inverse only finds the 'true' model when the starting model has been pre-warped to fit the data (Figures 2, 3). From the perspective views shown in Figure 2 we can see that when the starting model does not fit the data very well the solution converges to a hyperbolic surface that fits the data, but that there are some samples (mixtures with low mass fractions of end-member 2) that are outside the sample space defined by the model. This illustrates the fact that the inverse fits a hyperbolic surface to the data by adjusting the model parameters, but it does not have any way of knowing whether the resulting model parameters violate the requirement that $0 \le \phi_i \le 1$ for all *i*. Thus the inverse may generate parameter estimates that produce a hyperbolic surface that fits the data, but which are nevertheless in violation of mass fraction constraints. The only way to prevent this is to carefully inspect the results, which is trivial for binary cases but requires somewhat more effort for ternary (and higher-order) cases.

When the starting model is warped to better match the data before starting the inversion, however, the method performs well (Figure 3). In the software included with this publication I facilitate this process by including a graphical interface that allows the user to manually perturb the model parameters until a satisfactory starting model is obtained. Comparison of the final model with the true values for the synthetic data (Table 1) reveals that the parameters have been perturbed towards the true values, but that they have not quite reached them. This is a consequence of the penalty paid for variations from the starting model, and it can be addressed by simply running the inversion again using the final model as a starting model. When this is done the solution converges to the true model within error. Careful selection of the starting model is thus essential for obtaining useful results, and some amount of forward modeling will always be needed prior to implementing the inverse method.

Inspection of the initial vs. final parameter estimates in Table 1 reveals that the inverse method perturbs the concentration ratios while leaving the end-member ratio values essentially unchanged. This is because the gradients in the likelihood function with respect to the end-member ratios are very weak, reflecting the aforementioned fact that the end-members can be anywhere on the best-fitting hyperbolic surface and produce the same data misfit. Thus, from a practical point of view, the inverse finds the concentration ratios that provide the best-fitting hyperbolic surface for a given starting model of end-member compositions, and may be viewed as a way of ‘tuning’ an initial model to the data by adjusting the concentration ratios.

The non-linear ML method presented herein is completely general and can be applied to mixing problems with arbitrarily large numbers of end-members. There are, however, practical considerations that render the method ill-suited for mixing problems with large numbers of end-members. Firstly, the disparity between the DOFs in the mixing model and the best-fitting hyperbolic surface increases with model order as $n(n-1)$, such that the inverse problem becomes increasingly under-determined as the model order grows. Secondly, inspection of the model fit to the data, which, as illustrated above is essential to avoid spurious results, becomes problematic for $n > 3$ because there is no way to generate a synoptic view of the model.

In summary, I describe a method for inverting ratio-ratio data to obtain estimates of mixing model parameters. The derivation allows for treatment of the general n-dimensional problem, but in practice it is best suited to binary and ternary mixing models. Care must be taken when implementing the method to find a starting model that produces a reasonable fit to the data, and to inspect the final model to ensure that it does not violate mass fraction positivity constraints. The method effectively tunes a starting model to mixture data, reinforcing the fact that fitting a mixing model to mixture data requires prior knowledge, however it may be derived, regarding the end-member compositions.

Figure Captions

1. The non-uniqueness of end-member components in the ternary mixing inverse problem. A suite of mixtures in a ternary system (red dots) defines a hyperbolic surface. For simplicity a planar surface is used in this figure, representing the special case where all concentration ratios are equal to one. In order to satisfy the positivity requirements for the mass fractions (e.g., $f_i \geq 0$) the end-member ratios must lie outside the data field, but their position on the hyperbolic surface defined by the data is otherwise formally unconstrained. For example, the data are equally well fit by the co-planar surfaces defined by end-member components [*1*,*2*,*3*] (gray/black surface) and [*A*,*B*,*C*] (yellow/blue surface).

2. Inverse results for Case I – initial guess is planar surface. Three different perspective views of the synthetic data (black dots) are shown with the mixing surface defined by the initial model (A) – (C) and the final model (D) – (F). Residuals for the initial and final models are shown in panel (G). Note that the mixing surfaces are slightly transparent to allow for visibility of all data points.

3. Model results for Case II – initial guess with curvature. As for Figure 2, three different perspective views of the synthetic data are shown with the mixing surface defined by the initial model (A) – (C) and the final model (D) – (F). Residuals for the initial and final models are shown in panel (G). Note that the mixing surfaces are slightly transparent to allow for visibility of all data points.

Acknowledgments. I wish to thank the geochemists in the Geology & Geophysics department at the Woods Hole Oceanographic Institution, including former members Ken Sims and Matt Jackson, for motivating this research and providing helpful discussions. I especially wish to thank John Rudge for his mathematical insight and helpful advice deriving the linear algebra formulation for the general equation for *n*-dimensional mixing surfaces. Finally, I thank the Woods Hole Oceanographic Institution for supporting this research through the Independent Study program.

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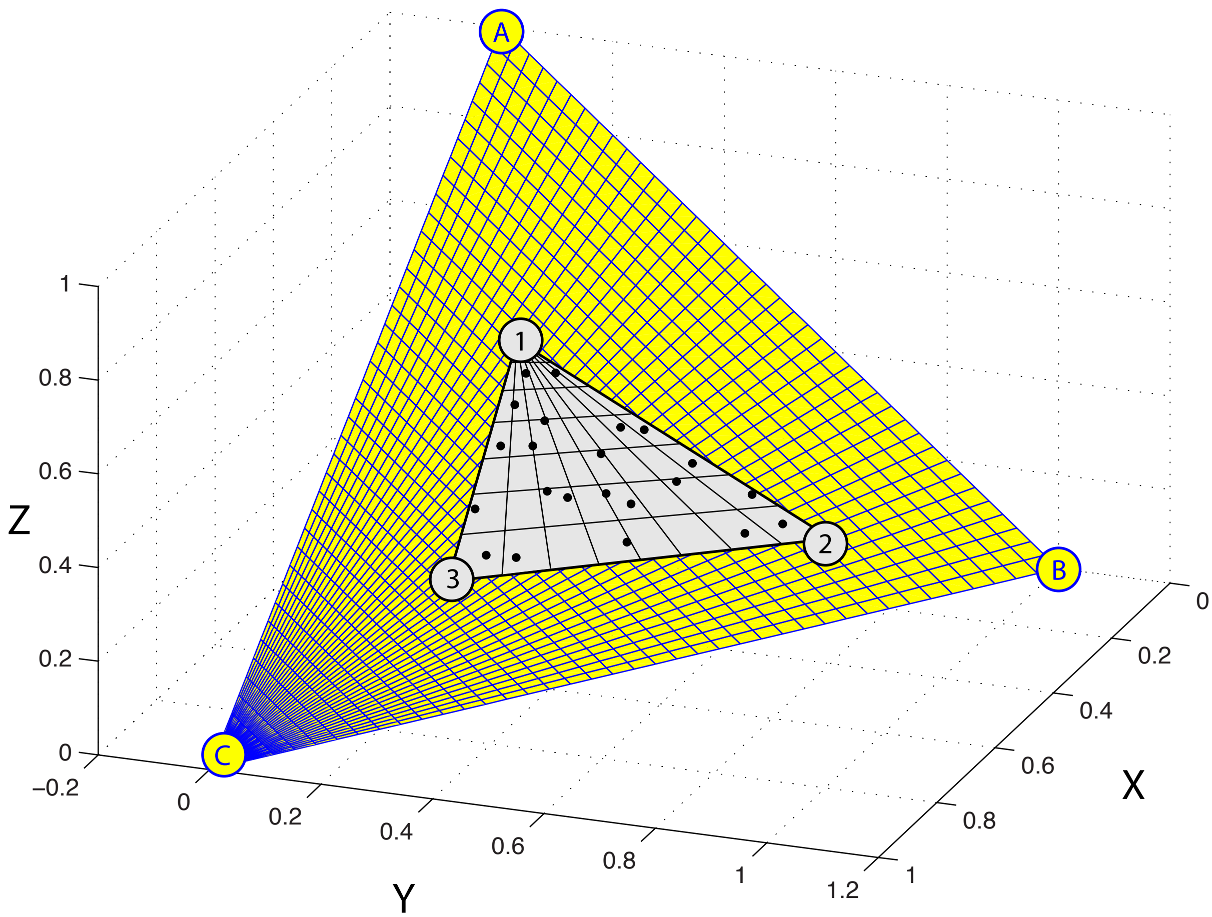
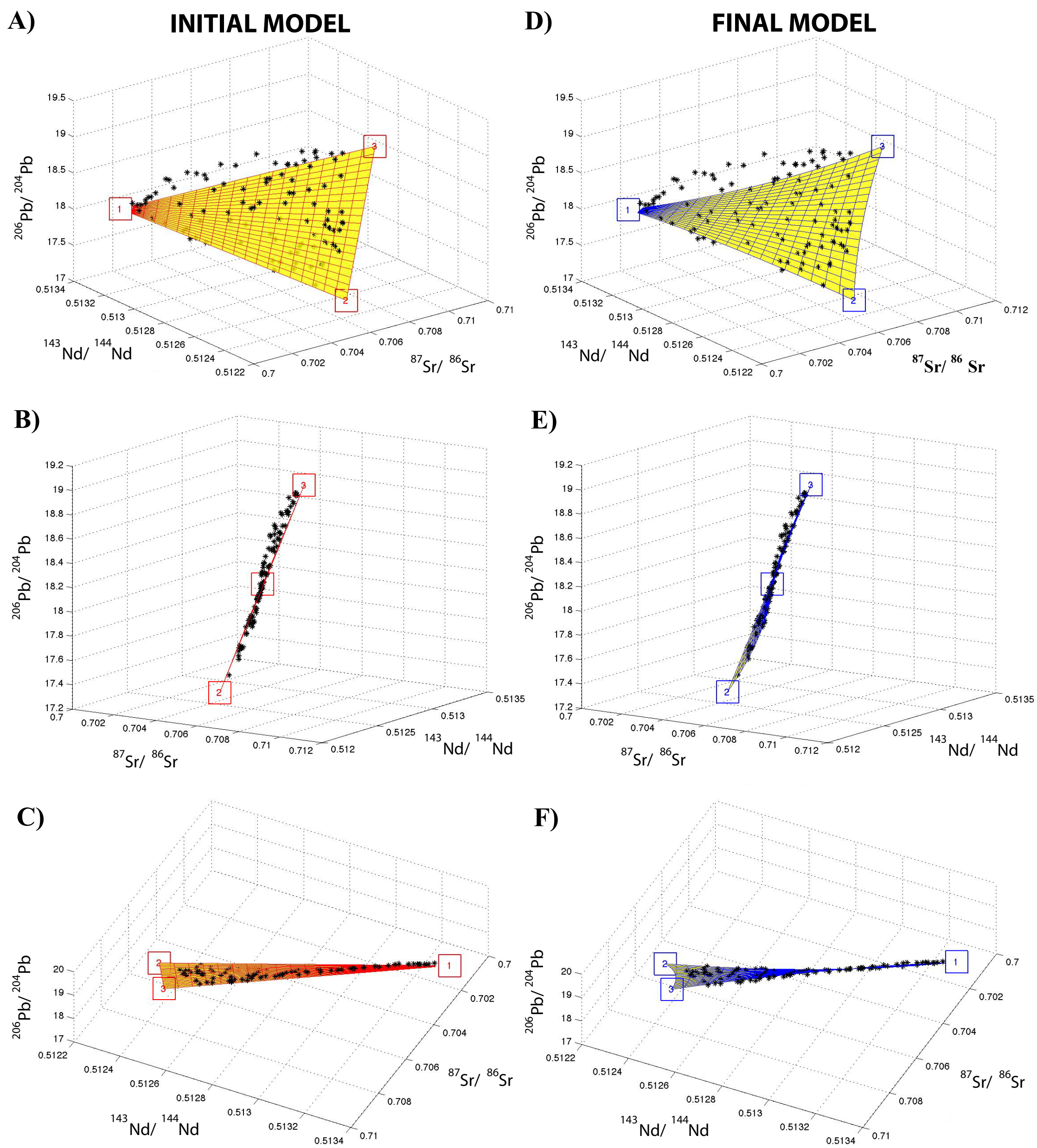


Figure 1. The non-uniqueness of end-member components in the ternary mixing inverse problem. A suite of mixtures in a ternary system (red dots) defines a hyperbolic surface. For simplicity a planar surface is used in this figure, representing the special case where all concentration ratios are equal to one. In order to satisfy the positivity requirements for the mass fractions (e.g., $\phi_i \geq 0$) the end-member ratios must lie outside the data field, but their position on the hyperbolic surface defined by the data is otherwise formally unconstrained. For example, the data are equally well fit by the co-planar surfaces defined by end-member components [1,2,3] (gray/black surface) and [A,B,C] (yellow/blue surface).



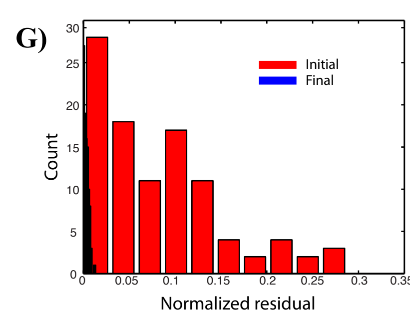
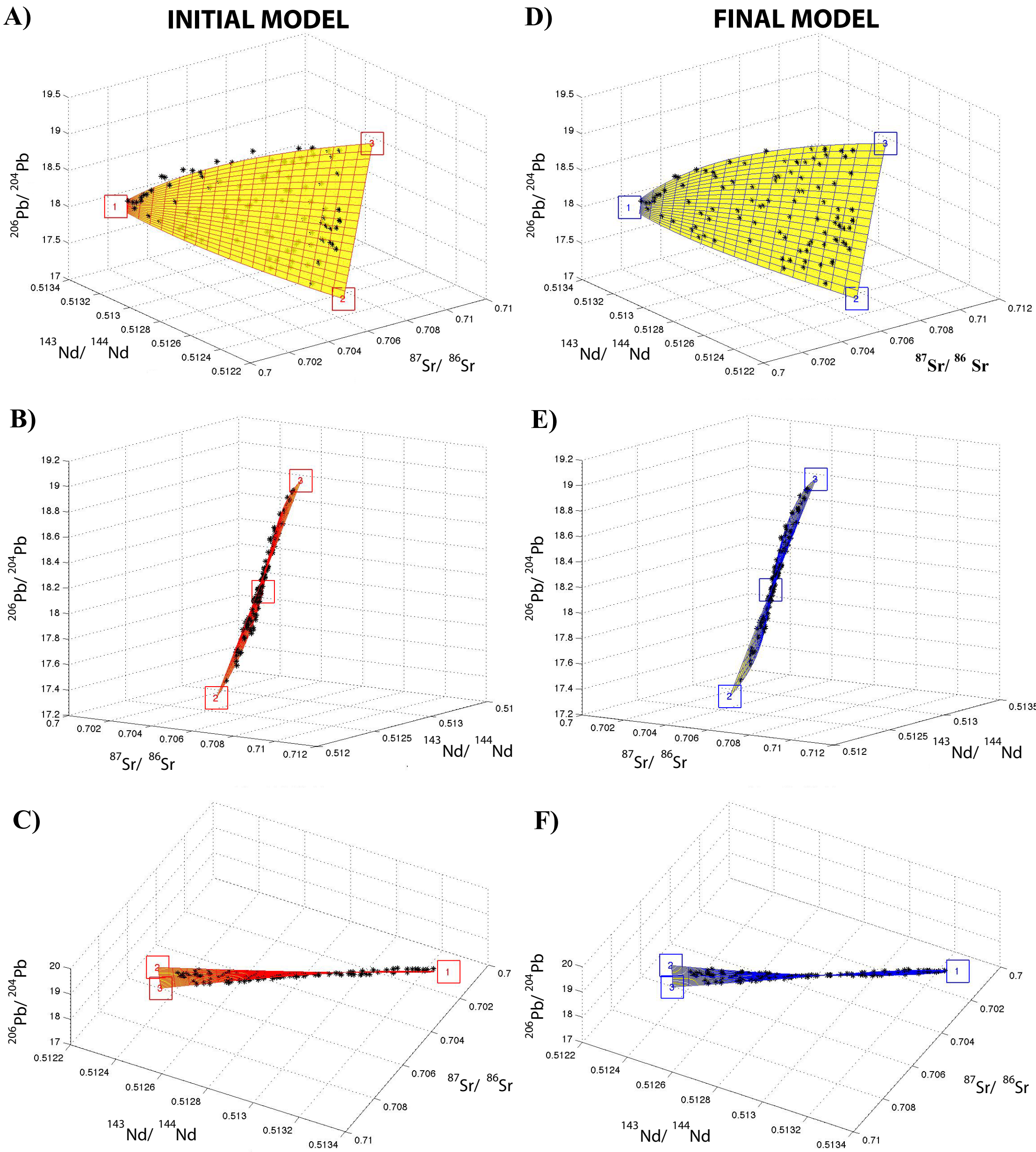


Figure 2. Inverse results for Case I – initial guess is planar surface. Three different perspective views of the synthetic data (black dots) are shown with the mixing surface defined by the initial model (A) – (C) and the final model (D) – (F). Residuals for the initial and final models are shown in panel (G). Note that the mixing surfaces are slightly transparent to allow for visibility of all data points.



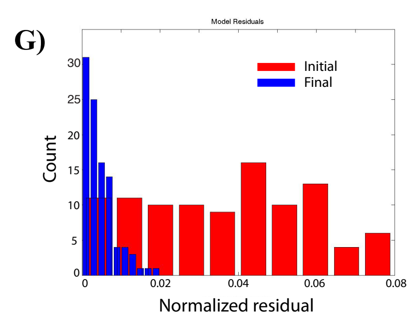


Figure 3. Model results for Case II – initial guess with curvature. As for Figure 2, three different perspective views of the synthetic data are shown with the mixing surface defined by the initial model (A) – (C) and the final model (D) – (F). Residuals for the initial and final models are shown in panel (G). Note that the mixing surfaces are slightly transparent to allow for visibility of all data points.