



Supplementary 1

Numerical proof that the equation $R_s = f_1R_1 + f_2R_2 + f_3R_3$ in Gobeil's model is not always established

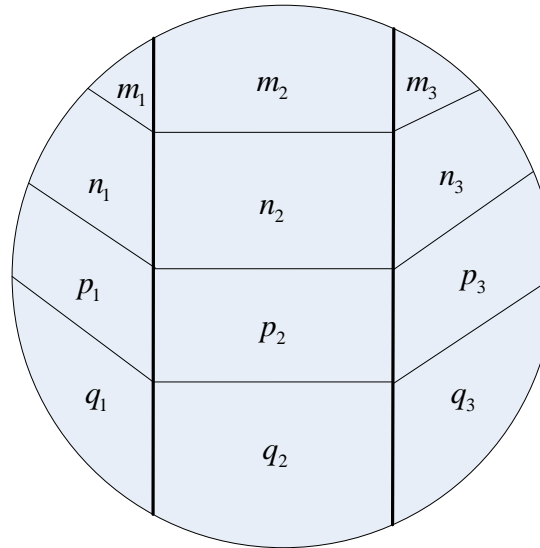


Figure S1. Schematic diagram of lead isotopic quality composition structure of sample.

In Figure S1, m_1 is the quality of ^{206}Pb which comes from pollution source 1, n_1 is the quality of ^{207}Pb which comes from pollution source 1, p_1 is the quality of ^{204}Pb which comes from pollution source 1, q_1 is the quality of ^{208}Pb which comes from pollution source 1. According to this rule, we can mark every isotopic quality which comes from pollution source 2 and pollution source 3. The variable M is labeled as the total quality of the sample. According to the definition of the isotope abundance ratio, pollution contribution rate, we can see:

$$f_1R_1 + f_2R_2 + f_3R_3 = \frac{m_1 + n_1 + p_1 + q_1}{M} \cdot \frac{m_1}{n_1} + \frac{m_2 + n_2 + p_2 + q_2}{M} \cdot \frac{m_2}{n_2} + \frac{m_3 + n_3 + p_3 + q_3}{M} \cdot \frac{m_3}{n_3}, R_s = \frac{m_1 + m_2 + m_3}{n_1 + n_2 + n_3}$$

$$\begin{aligned}
 \text{Proof: } f_1R_1 + f_2R_2 + f_3R_3 - R_S &= \frac{m_1 + n_1 + p_1 + q_1}{M} \cdot \frac{m_1}{n_1} + \frac{m_2 + n_2 + p_2 + q_2}{M} \cdot \frac{m_2}{n_2} + \frac{m_3 + n_3 + p_3 + q_3}{M} \cdot \frac{m_3}{n_3} - \frac{m_1 + m_2 + m_3}{n_1 + n_2 + n_3} \\
 &= \frac{1}{M} \left[\frac{m_1(m_1 + n_1 + p_1 + q_1)}{n_1} + \frac{m_2(m_2 + n_2 + p_2 + q_2)}{n_2} + \frac{m_3(m_3 + n_3 + p_3 + q_3)}{n_3} \right] - \frac{m_1 + m_2 + m_3}{n_1 + n_2 + n_3} \\
 &= \frac{1}{M} \left[\frac{m_1n_2n_3(m_1 + n_1 + p_1 + q_1) + m_2n_1n_3(m_2 + n_2 + p_2 + q_2) + m_3n_1n_2(m_3 + n_3 + p_3 + q_3)}{n_1n_2n_3} \right] - \frac{m_1 + m_2 + m_3}{n_1 + n_2 + n_3} \\
 &= \frac{1}{M \cdot n_1n_2n_3} \left[n_1n_2n_3(m_1 + m_2 + m_3) + m_1n_2n_3(m_1 + p_1 + q_1) + m_2n_1n_3(m_2 + p_2 + q_2) + m_3n_1n_2(m_3 + p_3 + q_3) \right] - \frac{m_1 + m_2 + m_3}{n_1 + n_2 + n_3} \\
 &= \frac{(n_1 + n_2 + n_3) \cdot [n_1n_2n_3(m_1 + m_2 + m_3) + m_1n_2n_3(m_1 + p_1 + q_1) + m_2n_1n_3(m_2 + p_2 + q_2) + m_3n_1n_2(m_3 + p_3 + q_3)] - M \cdot n_1n_2n_3 \cdot (m_1 + m_2 + m_3)}{M \cdot n_1n_2n_3 \cdot (n_1 + n_2 + n_3)}
 \end{aligned}$$

Set $\alpha = (n_1 + n_2 + n_3) \cdot [n_1n_2n_3(m_1 + m_2 + m_3) + m_1n_2n_3(m_1 + p_1 + q_1) + m_2n_1n_3(m_2 + p_2 + q_2) + m_3n_1n_2(m_3 + p_3 + q_3)] - M \cdot n_1n_2n_3 \cdot (m_1 + m_2 + m_3)$

Then $\alpha = m_1n_1^2n_2n_3 + m_1n_1n_2^2n_3 + m_1n_1n_2n_3^2 + m_1^2n_1n_2n_3 + m_1^2n_2^2n_3 + m_1^2n_2n_3^2 + m_1(p_1 + q_1)n_1n_2n_3 + m_1(p_1 + q_1)n_2^2n_3 + m_1(p_1 + q_1)n_2n_3^2 - Mm_1n_1n_2n_3$ +
 $m_2n_1^2n_2n_3 + m_2n_1n_2^2n_3 + m_2n_1n_2n_3^2 + m_2^2n_1^2n_3 + m_2^2n_1n_2n_3 + m_2^2n_1n_3^2 + m_2(p_2 + q_2)n_1^2n_3 + m_2(p_2 + q_2)n_1n_2n_3 + m_2(p_2 + q_2)n_1n_3^2 - Mm_2n_1n_2n_3$ +
 $m_3n_1^2n_2n_3 + m_3n_1n_2^2n_3 + m_3n_1n_2n_3^2 + m_3^2n_1^2n_2 + m_3^2n_1n_2^2 + m_3^2n_1n_2n_3 + m_3(p_3 + q_3)n_1^2n_2 + m_3(p_3 + q_3)n_1n_2^2 + m_3(p_3 + q_3)n_1n_2n_3 - Mm_3n_1n_2n_3$

$= \beta + \chi + \delta$

$$\beta = m_1n_1n_2n_3 \left[(n_1 + n_2 + n_3) \cdot \left(1 + \frac{m_1}{n_1} + \frac{p_1 + q_1}{n_1} \right) - M \right] \tag{1}$$

$$\chi = m_2 n_1 n_2 n_3 [(n_1 + n_2 + n_3) \cdot (1 + \frac{m_2}{n_2} + \frac{p_2 + q_2}{n_2}) - M] \tag{2}$$

$$\delta = m_3 n_1 n_2 n_3 [(n_1 + n_2 + n_3) \cdot (1 + \frac{m_3}{n_3} + \frac{p_3 + q_3}{n_3}) - M] \tag{3}$$

The following is obtained by $\alpha = \beta + \chi + \delta$:

$$\alpha = n_1 n_2 n_3 \cdot [(n_1 + n_2 + n_3) \cdot m_1 \cdot (1 + \frac{m_1}{n_1} + \frac{p_1 + q_1}{n_1}) - m_1 M + (n_1 + n_2 + n_3) \cdot m_2 \cdot (1 + \frac{m_2}{n_2} + \frac{p_2 + q_2}{n_2}) - m_2 M + (n_1 + n_2 + n_3) \cdot m_3 \cdot (1 + \frac{m_3}{n_3} + \frac{p_3 + q_3}{n_3}) - m_3 M]$$

$$= n_1 n_2 n_3 \cdot \left\{ (n_1 + n_2 + n_3) \cdot \left[m_1 + \frac{m_1^2}{n_1} + \frac{m_1(p_1 + q_1)}{n_1} + m_2 + \frac{m_2^2}{n_2} + \frac{m_2(p_2 + q_2)}{n_2} + m_3 + \frac{m_3^2}{n_3} + \frac{m_3(p_3 + q_3)}{n_3} \right] - M(m_1 + m_2 + m_3) \right\}$$

$$= n_1 n_2 n_3 \cdot \left\{ (n_1 + n_2 + n_3) \cdot \left[(m_1 + m_2 + m_3) + (\frac{m_1^2}{n_1} + \frac{m_2^2}{n_2} + \frac{m_3^2}{n_3}) + (\frac{m_1(p_1 + q_1)}{n_1} + \frac{m_2(p_2 + q_2)}{n_2} + \frac{m_3(p_3 + q_3)}{n_3}) \right] - M(m_1 + m_2 + m_3) \right\}$$

$$\Rightarrow f_1 R_1 + f_2 R_2 + f_3 R_3 - R_S = \frac{\alpha}{M \cdot n_1 n_2 n_3 \cdot (n_1 + n_2 + n_3)}$$

$$= \frac{\left\{ (n_1 + n_2 + n_3) \cdot \left[(m_1 + m_2 + m_3) + (\frac{m_1^2}{n_1} + \frac{m_2^2}{n_2} + \frac{m_3^2}{n_3}) + (\frac{m_1(p_1 + q_1)}{n_1} + \frac{m_2(p_2 + q_2)}{n_2} + \frac{m_3(p_3 + q_3)}{n_3}) \right] - M(m_1 + m_2 + m_3) \right\}}{M \cdot (n_1 + n_2 + n_3)} \tag{4}$$

Set $\varepsilon = n_1 + n_2 + n_3$, $\phi = m_1 + m_2 + m_3$, $\varphi = \frac{m_1^2}{n_1} + \frac{m_2^2}{n_2} + \frac{m_3^2}{n_3}$, $\gamma = \frac{m_1(p_1 + q_1)}{n_1} + \frac{m_2(p_2 + q_2)}{n_2} + \frac{m_3(p_3 + q_3)}{n_3}$

$$\text{so, } f_1R_1 + f_2R_2 + f_3R_3 - R_S = \frac{\varepsilon \cdot (\phi + \varphi + \gamma) - M\phi}{M \cdot \varepsilon} \quad (5)$$

hypothesis : $f_1R_1 + f_2R_2 + f_3R_3 - R_S = 0$, If the original formula was established ,the following conditions should be met:

$$\begin{aligned} \varepsilon \cdot (\phi + \varphi + \gamma) - M\phi &= 0 \\ (M - \varepsilon) \cdot \phi &= \varepsilon \cdot (\varphi + \gamma) \end{aligned}$$

$$\frac{\phi}{\varepsilon} = \frac{\varphi + \gamma}{M - \varepsilon} \quad (6)$$

From Equation 6: $Left = \frac{\phi}{\varepsilon} = \frac{m_1 + m_2 + m_3}{n_1 + n_2 + n_3} = R_S$; $right = \frac{\varphi + \gamma}{M - \varepsilon}$;

However,

$$\varphi + \gamma = \frac{m_1^2}{n_1} + \frac{m_2^2}{n_2} + \frac{m_3^2}{n_3} + \frac{m_1(p_1 + q_1)}{n_1} + \frac{m_2(p_2 + q_2)}{n_2} + \frac{m_3(p_3 + q_3)}{n_3}$$

$$= \frac{m_1}{n_1} (m_1 + p_1 + q_1) + \frac{m_2}{n_2} (m_2 + p_2 + q_2) + \frac{m_3}{n_3} (m_3 + p_3 + q_3)$$

$$= R_1 \cdot (m_1 + p_1 + q_1) + R_2 \cdot (m_2 + p_2 + q_2) + R_3 \cdot (m_3 + p_3 + q_3)$$

So:

$$\begin{aligned} right &= \frac{\varphi + \gamma}{M - \varepsilon} = \frac{R_1 \cdot (m_1 + p_1 + q_1) + R_2 \cdot (m_2 + p_2 + q_2) + R_3 \cdot (m_3 + p_3 + q_3)}{M - \varepsilon} \\ &= R_1 \cdot \frac{(m_1 + p_1 + q_1)}{M - \varepsilon} + R_2 \cdot \frac{(m_2 + p_2 + q_2)}{M - \varepsilon} + R_3 \cdot \frac{(m_3 + p_3 + q_3)}{M - \varepsilon} \end{aligned} \quad (7)$$

From $\left\{ \begin{aligned} \frac{\varphi + \gamma}{M - \varepsilon} &= R_S \\ f_1R_1 + f_2R_2 + f_3R_3 &= R_S \end{aligned} \right.$

It shows: To set up the original hypothesis, $\frac{\varphi + \gamma}{M - \varepsilon} = f_1R_1 + f_2R_2 + f_3R_3$ should be correct.

That is $R_1 \cdot \frac{(m_1 + p_1 + q_1)}{M - \varepsilon} + R_2 \cdot \frac{(m_2 + p_2 + q_2)}{M - \varepsilon} + R_3 \cdot \frac{(m_3 + p_3 + q_3)}{M - \varepsilon} = f_1R_1 + f_2R_2 + f_3R_3$ (8)

Must meet the conditions: $\frac{(m_1 + p_1 + q_1)}{M - \varepsilon} = f_1, \frac{(m_2 + p_2 + q_2)}{M - \varepsilon} = f_2, \frac{(m_3 + p_3 + q_3)}{M - \varepsilon} = f_3;$

Because the form of the three equations above are similar, we only need to discuss $\frac{(m_1 + p_1 + q_1)}{M - \varepsilon} = f_1$ as follows:

$$\frac{(m_1 + p_1 + q_1)}{M - \varepsilon} = f_1$$

$$\frac{m_1 + p_1 + q_1}{M - \varepsilon} = \frac{m_1 + n_1 + p_1 + q_1}{M}$$

Set $\eta = m_1 + p_1 + q_1$, transform to $\frac{\eta}{M - \varepsilon} = \frac{\eta + n_1}{M}$

That is $Mn_1 = \varepsilon\eta + \varepsilon n_1, \frac{\varepsilon}{M} = \frac{n_1}{\eta + n_1}$

Let ε, η back, that is $\frac{n_1 + n_2 + n_3}{M} = \frac{n_1}{m_1 + n_1 + p_1 + q_1}$ (9)

We can see $\frac{n_1 + n_2 + n_3}{M}$ is exactly ²⁰⁷Pb abundance ratio of the sample, $\frac{n_1}{m_1 + n_1 + p_1 + q_1}$ is exactly ²⁰⁷Pb abundance ratio of the source 1, From a realistic point of view, the probability when the two abundance ratios are identical is very small.

So, $\frac{n_1 + n_2 + n_3}{M} = \frac{n_1}{m_1 + n_1 + p_1 + q_1}$ is not necessarily always true, and the probability of establishment is very small.

As a result, original hypothesis $f_1R_1 + f_2R_2 + f_3R_3 - R_s = 0$ is not necessarily always true, and the probability of establishment is very small. The Gobeil’s model actually is not a perfect method to resolve the pollution sources.