

APPENDIX

The basalt glass standard ALV-519-4-1 is used to track and correct for instrumental drift and bias over the course of a session. For simplicity, the following steps are described for water, but fluorine and phosphorus count ratios can be substituted for OH/Si for reducing these elements.

The first correction accounts for instrumental drift over the course of a day, or occasionally multiple days. The count ratio $^{16}\text{O}^{1}\text{H}/^{30}\text{Si}$ for ALV-519-4-1 is plotted against analysis number and fit with a linear regression of the form, $y(x) = mx + b$, where y is the count ratio $^{16}\text{O}^{1}\text{H}/^{30}\text{Si}$ and x is analysis number (Figure A1a). This regression is then used to calculate percent change as a function of analysis number, $P(x) = y(x) / y(1)$.

While $P(x)$ is not linear, it is very close to linear over the number of analyses one can gather in a single day. For simplicity, we approximated $P(x)$ as linear, thus forming the equation $P(x) = m_p x + b_p$. This allows P to be calculated based purely on analysis number, independent of the exact values of $y(x)$. P can then be applied to all unknown data (a_{raw}), including the ALV-519-4-1 data, as

$$\alpha'(x) = a_{\text{raw}}(x) * P(x) = a_{\text{raw}}(x) * (m_p x + b_p). \quad (1)$$

After these calculations, corrected $^{16}\text{O}^{1}\text{H}/^{30}\text{Si}$ values for ALV-519-4-1 should have a standard deviation of $<10\%$ (Figure A1b).

This drift correction accounts for differences of the course of a day, but it does not account for any differences between days nor any differences between mounts since each linear correction is referenced to the start of that day. In order to account for the differences between days and between mounts, a K-factor correction (K) is calculated. The K-factor is equal to the average $^{16}\text{O}^{1}\text{H}/^{30}\text{Si}$ count ratio of ALV-519-4-1 on the standard mount divided by the average $^{16}\text{O}^{1}\text{H}/^{30}\text{Si}$ count ratio of ALV-519-4-1 on the unknown mount. The K-factor correction is then applied to all α' data from the Equation 1 as

$$\alpha''(x) = \alpha'(x) * K \quad (2)$$

where α'' represents the data after both corrections. The average count ratio of ALV-519-4-1 in α'' should now have a standard deviation of $<10\%$ across data from every day and on every mount from the session (Figure A1c).

► **FIGURE A1:** Example set of analyses of ALV-519-4-1 from one day of the January 2016 session, shown at each step of the instrumental drift/bias corrections described. For each plot, the blue dots are individual analyses of ALV-519-4-1 on the unknown mount, the solid red line is the linear least-squares solution to those data as a function of analysis number, and the dashed red line is the average value for ALV-519-4-1 on the standard mount. Raw data before any correction are shown in (a), drift-corrected data are shown in (b), and data after all corrections are shown in (c). The x- and y-scales are the same for all plots.

