A mass fractionation law for high-transmission MC-ICP-MS

FRANCIS ALBARÈDE, PHILIPPE TELOUK AND EMMANNUELLE ALBALAT[1]

[1]Ecole Normale Supérieure de Lyon, 69007 Lyon, France, (albarede@ens-lyon.fr)

Russell *et al*'s [1] exponential law provides the standard form used for the correction of the mass bias on isotopic ratios measured by mass spectrometry and more specifically since 1992 for MC-ICP-MS. It is nevertheless expected to fail as soon as the ion/atom efficiency exceeds the percent level and preliminary observations on new-generation MC-ICP-MS confirms this prediction. At 100% transmission, the mass bias must by unity. The physics of mass fractionation in the torch involves collisions with abundant neutral argon atoms, the formation of a shock wave in the interface, and the presence of a boundary layer in the nozzle, and is therefore particularly complicated. At low transmission, the ratio between two isotopes 1 and 2 is affected by a factor $(M_1/M_2)^{\beta}$, where M is the atomic mass. The mass fractionation factor β remains essentially constant and equal to ~2 across the mass range, which suggests that the velocity of radial expansion is proportional to mass [2], but this law has not received a universally accepted.physical justification.

At high transmission, the exponential law breaks down. What is the relationship between mass fractionation and transmission? A simple conservation principle applied to the fraction lost suggests an example of a transmission-compliant mass fractionation law:

$$R_{\mathrm{meas}} = R_{\mathrm{true}} \times \frac{(M_1/M_2)^{\beta}}{(1 - n/n_0) + (n/n_0)\,(M_1/M_2)^{\beta}}$$

in which n/n_0 is the transmission. At the limit, the exponential law is obtained when $n/n_0 \rightarrow 0$, while the mass bias disappears when $n/n_0 \rightarrow 1$. Other forms involving different functional relationships are also possible and need to be tested. The instrument transmission for a particular element is an essential parameter of mass fractionation laws and should therefore be recorded with the best possible precision. We will present data on the isotope compositions of various elements across the mass range that will allow the present model to be tested.

[1] Russell WA *et al* (1978) *GCA* **42**, 1075-1090 [2] Maréchal *et al* (1999) *Chem. Geol.* **156**, 251-273.

The Y-3 tephra: New insights

P. G. ALBERT[1,2]*, M. HARDIMAN[3], J. KELLER[4], E.L. TOMLINSON[5], U.C. MÜLLER[6], V.C. SMITH[2] AND M. MENZIES[1]

[1]Earth Sciences, Royal Holloway University of London, UK
[2]RLAHA, University Oxford, UK (paul.albert@rlaha.ox.ac.uk)
[3]Geography, Royal Holloway University of London, UK
[4]Albert-Ludwigs-University Freiburg, Germany
[5]Geology, Trinity College Dublin, Ireland.
[6]Institute of Geosciences, Frankfurt, Germany

The 'Y-3' tephra is a crucial stratigraphic marker within the central Mediterranean region, reported occurrences within palaeoenvironmental archives occur at or close to the Marine Isotope Stage 3/2 transition, which in turn is linked to the onset of Heinrich Stadial 3 (HS3) [1]. Consequently this tephra offers enormous potential to assess potential leads and lags between Mediterranean archives. Until now the type locality Ionian Sea Y-3 tephra has remained poorly characterised. Consequently, numerous other distal tephras occurring in broadly similar stratigraphic positions and displaying a similar trachyte composition have been correlated to the Y-3 tephra without the appropriate geochemical validation.

Here we present the first grain-specific major, minor (EPMA) and trace element (LA-ICP-MS) characterisation of the Y-3 tephra recorded in its type locality of the Ionian Sea (M25/4-12). The High-K phono-trachyte geochemistry of the Ionian Sea Y-3 supports a provenance from within the Campi Flegrei caldera consistent with previous interpretations. Geochemical glass data reveals problems with a correlation between the Ionian Sea Y-3 and the proposed proximal equivalent, the VRa Tufi Biancastri unit. As a result the VRa age must not be imported to the Ionian Sea Y-3 tephra.

Using the new diagnostic major, minor and trace element chemistry of the type locality Y-3 tephra we are able to assess and establish precise distal-distal tephra correlations. Robust identification of the Y-3 in the Tenaghi Philippon (Greece) sedimentary archive both offers an opportunity to determine a precise age for this distal marker and assess its stratigraphic position in the context of a high resolution palaeoenvironmental archive. A Bayesian-based ^{14}C age model provides an age of 29, 350-30, 160 cal yrs BP for the Y-3 tephra. The Y-3 tephra resides within the latter part of a period of reduced total tree pollen percentages, which is related to drier stadial conditions. The independent age of the Y-3 tephra and it environmental context suggests that this marker tephra post dates the onset of both HS3 in the Mediterranean region and GS-5 in the INTIMATE event stratigraphy [2].

[1] Zanchetta *et al*, (2008) *JVGR* **177**, 145-154; [2] Blockley *et al*, (2012), *Quat. Sci. Revs.* **36,** 2-10.

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