

## Fractionation of Nb/Ta during subduction of carbonate-rich sediments

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### Supplementary Information

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### Tyrrhenian Sea Samples Description

Samples from the Tyrrhenian basin were obtained from the MARUM GeoB Core Repository. They were originally collected during the Deep Sea Drilling Project (DSDP) and Ocean Drilling Program (ODP) in expedition 42 and 107. The samples were selected to be representative of the depleted magmatism recorded in the Tyrrhenian basin, which is thought to form in a back arc setting. In terms of composition they resemble that of transitional MORB and they are thought to derive from a depleted mantle slightly re-enriched by subduction-derived fluids (Barberi *et al.*, 1978; Beccaluva *et al.*, 1990; Dietrich *et al.*, 1977, 1978; Gasperini *et al.*, 2002; Hamelin *et al.*, 1979).

### Analytical Methods

Major element concentrations of the seven samples from the Tyrrhenian basin were obtained using a Philips PW 2400 XRF at the Universität zu Köln. The trace element content was determined following the procedure of Garbe-Schönberg (1993) and using an Agilent 7500cs ICP-MS at the Universität zu Kiel. The same samples were analysed for their Sr-Nd-Pb isotopic composition at the Università degli Studi di Firenze, leaching the powders in 1 M HCl and following the procedure described in Avanzinelli *et al.* (2005) for dissolution, chemical separation and TIMS measurements.

Elemental concentrations of HFSE (Nb, Ta, Zr, Hf), Lu, W, Th, and U were obtained along with  $^{176}\text{Hf}/^{177}\text{Hf}$  following the same procedure outlined in Bragagni *et al.* (2022), which is based on the methods described in Luo *et al.* (1997), Kirchenbaur *et al.* (2016), Münker *et al.* (2001), Münker (2010), Weyer *et al.* (2002), Kleine *et al.* (2004). Briefly, 100 mg of powder was weighted along with isotope tracers enriched in  $^{233-236}\text{U}$ – $^{229}\text{Th}$ – $^{183}\text{W}$ – $^{180}\text{Ta}$ – $^{180}\text{Hf}$ – $^{176}\text{Lu}$ – $^{94}\text{Zr}$ . Samples were dissolved and treated as described in Bragagni *et al.* (2022). Specifically, the W fraction was separated using anion resin exchange chemistry (modified from Kleine *et al.*, 2004), the HFSE were purified in three

steps of resin exchange chemistry (LN-spec, anion, LN-spec) using a modified procedure originally developed by Münker *et al.* (2001), U and Th were separated with TRU-spec chemistry (Luo *et al.*, 1997). All measurements were performed at Universität zu Köln with a Thermo-Scientific Neptune MC-ICP-MS equipped with a Cetac Aridus II. Blanks were 36–54 pg for W, 38–163 pg for Ta, 0.7–1.4 ng for Zr, 28–57 pg for Nb, 43–123 pg for Hf, 9 pg for Th and 35 pg for U, and are all negligible with respect to the amounts of sample processed.

## Quantifying the Role of Carbonate-rich Melts in Roman Magmatic Province

Quantifying the effect of silicate and carbonatitic melts in the investigated samples is not straightforward. This is due to the complexity of the involved processes and because melting parameters, such as partition coefficients, are not readily available. Green (2000) provides  $K_d$  for HFSE in rutile in equilibrium with carbonatitic magmas. However, in the case of marls, a silicate melt will also form along the carbonatite melt (*e.g.*, Gülmez *et al.*, 2023; Skora *et al.*, 2015). Therefore, to quantitatively model HFSE and other trace elements, both silicate and carbonatite partition coefficients must be considered. In the literature there are few experimental works reporting the trace element composition of melts in equilibrium with sediments at pressure and temperature relevant for mantle wedge conditions (Herman and Rubatto, 2009; Skora and Blundy, 2010; Skora *et al.*, 2015; Gülmez *et al.*, 2023). Among them, Skora *et al.* (2015) and Gülmez *et al.* (2023) investigated carbonate-rich lithologies but they do not report partition coefficients. The only experimental work with partition coefficients for sediments melt is Skora and Blundy (2010), which used a radiolarian clay lithology. These partition coefficients can be used to model melting of silicic sediments as those expected to influence the Tuscan magmatic province. Although not ideal, the same partition coefficient can be employed to reproduce the composition of a silicic melt in equilibrium with a marl, as needed to explain the Roman magmatism. As such, we assume that HFSE are controlled by rutile in equilibrium with a carbonatitic melt (partition coefficients of Green, 2000), whereas other elements are controlled by silicates in equilibrium with the silicic melt (partition coefficients of Skora and Blundy, 2010). It should be stressed that this is a simplification because also other elements can be influenced by carbonatite melts (especially REE). Moreover, also the partitioning of HFSE between rutile (and possibly other phases) and the silicate melt is not considered.

An alternative proxy of the metasomatic melt affecting the mantle wedge under the Roman magmatic province is represented by the carbonate-rich inclusions observed by Korsakov and Herman (2006) in orogenic massifs. Therefore, we model the effect of adding three different melts to a DMM, two inferred from melting silica- and carbonate-rich sediments, and one simply using the composition of a carbonate-rich inclusions of Korsakov and Herman (2006). Modelling was performed only with Nb/Ta and radiogenic isotope ratios to avoid variations of concentrations due to partial melting in the metasomatised mantle wedge. As shown in Figure S-1, mixing the DMM with silicic melt can reproduce the signature of the Tuscan magmatic province in Nb/Ta vs. Sr or Nd isotopes. The Roman magmas lie below the two mixing lines obtained for carbonate-rich metasomatic melts. This is readily explained by the concurrent occurrence of carbonate-rich metasomatic melts along with silicic melts (*i.e.* with canonical Nb/Ta) or fluids (*i.e.* negligible HFSE content but significant Sr and Nd).

## References for Literature Data in Figures 2 and 3

Reference values for PM values are taken from Münker *et al.* (2003) and Palme and O'Neill (2014). Literature isotope dilution data and Sr-Nd-Hf isotopes reported in Figure 3 are from: Tonga (Beier *et al.*, 2017), Cyprus (König *et al.*, 2008), Solomon (König *et al.*, 2008; Schuth *et al.*, 2009), Papua New Guinea (PNG) (König *et al.*, 2010), Kamchatka and Aleutian arc (Churikova *et al.*, 2001; Münker *et al.*, 2004; Yogodzinski *et al.*, 1995), Santorini and Bulgaria (Kirchenbaur *et al.*, 2012; Kirchenbaur and Münker, 2015), Sunda (Kirchenbaur *et al.*, 2022), Stromboli (Bragagni *et al.*, 2022).

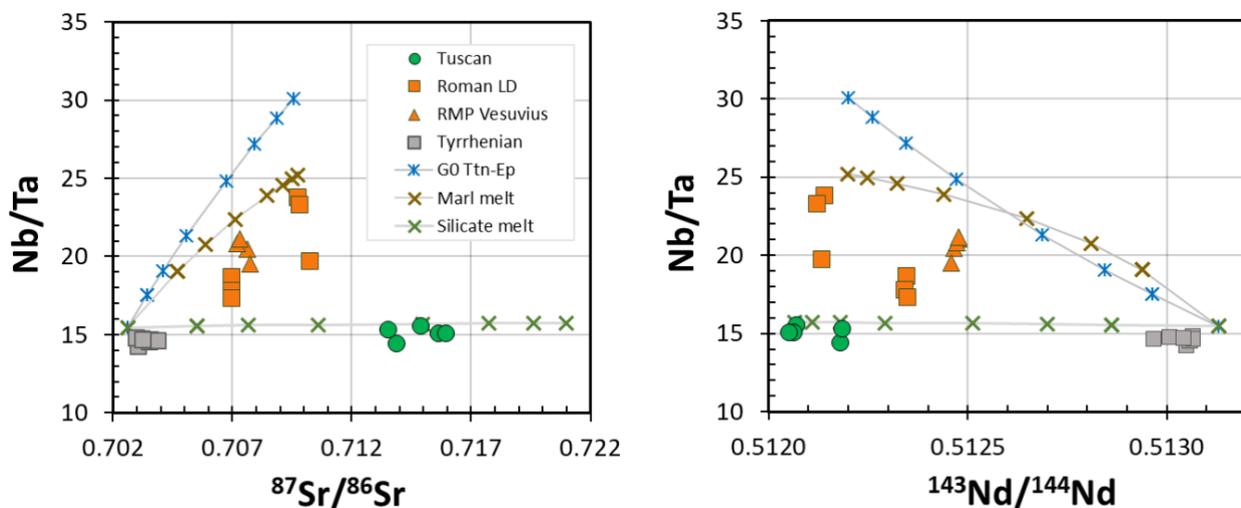


## Supplementary Table

**Table S-1** Compiled dataset with HFSE concentrations measured by isotope dilution (this work and Bragagni *et al.*, 2022) along with major-trace elements and Hf-Sr-Nd-Pb isotope data (this work and indicated literature).

Table S-1 is available for download (.xlsx) from the online version of this article at <https://doi.org/10.7185/geochemlet.2410>.

## Supplementary Figure



**Figure S-1** Mixing models to reproduce the Nb/Ta signature of the Roman and Tuscan magmatic provinces. The blue symbols represent a mixing curve between the DMM and the carbonate-rich melt (inclusion G0 Ttn-Ep) reported by Korsakov and Herman (2006) and assuming Sr and Nd isotope composition from marl SD53 (Casalini *et al.*, 2019). The brown and green crosses display a mixing between the DMM and melts from a carbonate-rich marl (SD53 of Conticelli, 1998; Conticelli *et al.*, 2015; Casalini *et al.*, 2019) or a carbonate poor lithology (SD75 of Conticelli, 1998; Conticelli *et al.*, 2015; Casalini *et al.*, 2019), respectively. For the marl melting, we assume that HFSE are controlled only by rutile, using the partition coefficients of Green (2020) for a carbonatitic melt and imposing a 1 % modal fraction of rutile. Partition coefficients for Sr and Nd are from Skora and Blundy (2010) (900 °C). We assume an arbitrary 50 % of partial melting of the marl. Such a melt was then mixed at variable proportions with the DMM. For melting the carbonate-poor lithology, we only used the partition coefficients of Skora and Blundy (2010) (900 °C) and assume a melting degree of 50 % before mixing it with the DMM. Symbols for the mixing models are reported adding 0, 0.5, 1, 2, 5, 10, 20, 50 % of sediment melt to the DMM. In a possible scenario, the Roman magmas are explained by the double contribution of melts from carbonate-rich (*i.e.* marl melting or G0 Ttn-Ep melt) and carbonate-poor lithologies (to lesser extent). Instead, the Tuscan magmas are explained by melting only the carbonate-poor lithology. DMM values are taken from Workman and Hart (2005).



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