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Linking thallium isotope cycling to seafloor hydrothermal sulfides forming by active diffusers at the Kolumbo submarine volcano, Hellenic volcanic arc

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We report the first $\varepsilon^{205}Tl$ composition of hydrothermal sulfides from active rhyolitic pumice-hosted, CO_2-degassing, SMS diffusers ("KCO_2Ds")[1], from the Aegean Kolumbo arc-volcano. Nineteen samples of Tl-rich (≤8.2 wt%) SMS came from actively boiling "KCO_2Ds" reflecting different mineralogic zones, i.e., *inner* (chalcopyrite-pyrite-galena-marcasite), *intermediate* (pyrite-sphalerite-semseyite) and *outer* (orpiment-realgar-stibnite, semseyite-meneghinite-pyrite); also, fifteen pumice samples were analyzed. The $\varepsilon^{205}Tl$ values of our samples range from -2.38 to +6.96, and -3.43 to -1.27 ε-units, for SMS and pumice, respectively. The measured range, and average $\varepsilon^{205}Tl_{pumice}$ values, of -3.43 to -1.27 ε-units, -2.21 ε-units, respectively, are consistent with magmatic Tl being transported via volcanism to the (sub)seafloor, and incorporated into rhyolitic pumice[2], preserving its primary depleted-mantle signature[3]. This is because, despite its extreme volatility, Tl remains in the sulfide melt[2] during magmatic degassing, and its isotopic composition is only marginally affected during magmatic differentiation[4]. An isotopic zoning towards heavier compositions exists, between the inner (range: +0.02—+2.6 $\varepsilon^{205}Tl_{inner}$; average: +1.42 $\varepsilon^{205}Tl_{inner}$) and intermediate sections (range: +2.23—+6.96 $\varepsilon^{205}Tl_{intrm}$; average: $\varepsilon^{205}Tl_{intrm}$=+4.29) whereas the outer sections exhibit significantly lighter values (range: -2.38—+1.86 $\varepsilon^{205}Tl_{outer}$; average: -1.4 $\varepsilon^{205}Tl_{outer}$). Combined with textural and geochemical characteristics for fluid-boiling[1], the measured $\varepsilon^{205}Tl_{outer}$ values are consistent with transport of Tl by liquid, preferentially enriched in ^{205}Tl values governed by Rayleigh-type distillation. The broad isotopic fractionation from the inner to intermediate diffuser sections ($\leq\varepsilon^{205}Tl$=6.94) are interpreted by sulphide precipitation during crystallization of a sulphide melt[6]. The latter is manifested by sulfide-hosted (sub)micron polyphase sulphide/sulphosalt inclusions enriched in Tl and low-melting point chalcophile elements (LMCEs)[5], generated by ≥270°C LMCE-rich fluid-induced CDR reactions as further reflected in $\varepsilon^{205}Tl$ vs. molar Tl/As-ratios diagram[6]. Rayleigh distillation modelling and sulphide precipitation during crystallization of a sulphide melt explain the substantial Tl isotope fractionation up to 9.34 ε values.

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