Supplementary Material

Figure S1. Lithogenic–carbonate interaction (Ca–Sr)

The Ca–Sr signal is interpreted as a lithogenic–pedogenic signature linked to carbonate minerals and secondary soil phases, rather than to hydrothermal processes.

Score and anomaly maps of Calcium (Ca) and Strontium (Sr) showing their intermediate geochemical behaviour between mafic (Fe–Mg–Cr–Ni–Co) and felsic volcanic sources (Ba–Pb–Ag). Elevated Ca–Sr values in the Maquedade–Maka sector reflect lithological mixing between volcanic and Quaternary carbonate units, consistent with PCA–CLR loadings where Ca, Sr, and Ba occupy transitional positions between lithogenic and hydrothermal fields.

Spatial distribution maps for Ca and Sr derived from CLR-transformed datasets further illustrate this behaviour. High anomaly classes within the central–western sector delineate a felsic–carbonate transitional zone, reinforcing the mixed lithogenic and secondary carbonate influence described in the main PCA interpretation.

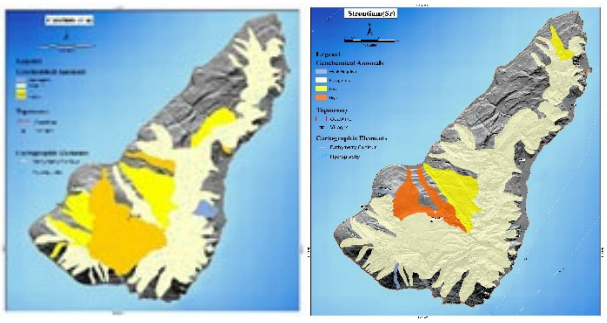
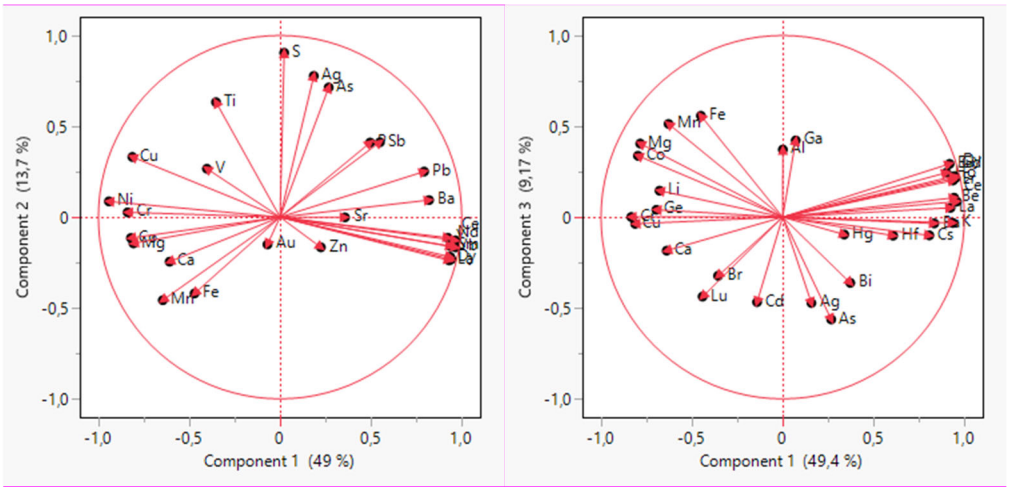


Figure S2. PCA–CLR biplots and loading coefficients for the extended dataset (including Ca, Sr, P, and REE).

(a) PC1 vs. PC2 (49%+13.7%) distinguishing the lithogenic and hydrothermal components.

(b) PC1 vs. PC3 (49%+9.9%) highlighting supergene–oxidative processes.



PC1 (49%) — Lithogenic component: Separates mafic (Ni–Co–Cr–Cu–Mg) from felsic–carbonate sources (REE + Ba–Pb–Ca–Sr), reflecting primary magmatic differentiation and volcanic–carbonate mixing.

PC2 (13.7%) — Hydrothermal vector: Dominated by As–Sb–Ag–S (± minor Au), characteristic of acid-sulfate to intermediate-sulfidation epithermal systems.

PC3 (9.9%) — Supergene–oxidative component: Characterized by Fe–V–Zn enrichment linked to incipient oxidative weathering under humid tropical conditions.

Transitional elements (Ti, P, Y, Yb, Sr) show mixed loadings between lithogenic and supergene domains, indicating partial oxidation or carbonate interaction.

Figure S3. Loading coefficients of all elements.

Bar chart displaying PC1 (red), PC2 (green), and PC3 (blue) loadings for all major, trace, and REE elements.

PC1: lithogenic framework (mafic vs. felsic–carbonate), PC2: hydrothermal association, (As–Sb–Ag–S–Au), PC3: oxidative–supergene trend (Fe–V–Zn–P–REE).

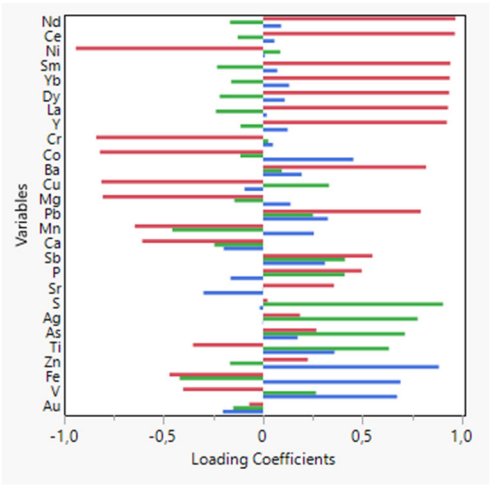


Table S1. PCA–CLR loadings of all analyzed elements for the first three principal components (PC1–PC3).

Element	PC1	PC2	PC3
Nd	1.0	-0.2	0.1
Ce	1.0	-0.1	0.1
Ni	-0.9	0.1	0.0
Sm	0.9	-0.2	0.1
Yb	0.9	-0.2	0.1
Dy	0.9	-0.2	0.1
La	0.9	-0.2	0.0
Y	0.9	-0.1	0.1
Cr	-0.8	0.0	0.0
Co	-0.8	-0.1	0.5
Ba	0.8	0.1	0.2
Cu	-0.8	0.3	-0.1
Mg	-0.8	-0.1	0.1
Pb	0.8	0.3	0.3
Mn	-0.6	-0.5	0.3
Ca	-0.6	-0.2	-0.2
Sb	0.5	0.4	0.3
P	0.5	0.4	-0.2
S	0.0	0.9	0.0
Ag	0.2	0.8	0.0
As	0.3	0.7	0.2
Ti	-0.4	0.6	0.4
Zn	0.2	-0.2	0.9
Fe	-0.5	-0.4	0.7
V	-0.4	0.3	0.7

Sr	0.4	0	-0.3
Au	-0.1	-0.2	-0.2

Table S2. PCA–CLR loadings and interpretation of the main geochemical processes identified in the Ataúro Island dataset.

This table summarizes the compositional structure of the dataset derived from PCA–CLR analysis. PC1 (49%) represents the lithogenic component separating mafic–ultramafic elements (Ni–Co–Cr–Cu–Mg) from felsic–carbonate associations (REE + Ba–Pb–Ca–Sr). PC2 (13.7%) defines the hydrothermal vector dominated by As–Sb–Ag–S (±Au), whereas PC3 (9.9%) reflects supergene–oxidative processes expressed by Fe–V–Zn enrichment. Transitional elements (Ti, P, Y, Yb, Sr) show mixed loadings, indicating partial oxidation and/or carbonate interaction.

Element group	PC1	PC2	PC3	Dominant process	Interpretation summary
Nd, Ce, Sm, Yb, Dy, La, Y	+0.9 to +1.0	–0.2	+0.1	Felsic–carbonate (lithogenic)	REE cluster defining felsic end-member; enriched in evolved volcanic + carbonate units
Ba, Pb	+0.8	+0.2–0.3	+0.2–0.3	Felsic / hydrothermal transitional	Associated with felsic magmatism and minor hydrothermal input
Ni, Cr, Co, Cu, Mg	–0.9 to –0.8	≈ 0	0–0.5	Mafic lithogenic	Magmatic signature of mafic basement; Co shows slight PC3 affinity
Mn, Fe, V, Zn	–0.6 to –0.4	–0.5 to +0.3	+0.7–0.9	Supergene–oxidative	Secondary Fe–V–Zn enrichment; redox mobility under humid tropical weathering
S, Ag, As, Sb	0 to +0.5	+0.7–0.9	0 to +0.3	Hydrothermal (epithermal)	Diagnostic As–Sb–Ag–S association; intermediate- to high-sulfidation fluids
Au	–0.1	–0.2	–0.2	Hydrothermal minor	Weak but coherent association with Ag–As–Sb trend; late-stage remobilization
Ti	–0.4	+0.6	+0.4	Transitional	Partly magmatic, partly oxidized; bridges Fe–V with hydrothermal influence
Ca, Sr, P	–0.6 to +0.5	–0.2 to +0.4	–0.3 to –0.2	Felsic–carbonate transitional	Reflect carbonate–volcanic mixing; Sr–P trending toward Fe-oxide affinity