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Exploring for laterally transported copper in gravels using radon detectors

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Preamble

This manuscript is based on an initial idea from the lead author, the late Peter Alan Winterburn as part of the Atacama Gravels Project, started under the MDRU Exploration Geochemistry Initiative in 2015. Thomas Bissig and Peter Winterburn discussed the preparation of this manuscript in May 2019 and Peter indicated that he would read over the first draft once more and provide additional input. His untimely death on June 21st, 2019 in Valparaíso, Chile has prevented this. However, based on his informal comments, we are confident that he would support the publication of this work, and we have therefore decided to continue in Peter´s honour. However, any potential errors and omissions are responsibility of the second and third authors.

Introduction

Copper is readily leached from acid generating sulfide ore during weathering and oxidation (e.g., Chavez 2000, Reich & Vasconcelos 2015). Copper-bearing solutions can be transported laterally for up to 6-8 km until the solutions are neutralized (or reduced) sufficiently to precipitate secondary Cu minerals (carbonates, silicates, oxides, halides, and sulphates). This process can form exotic Cu deposits (e.g., Munchmeyer *et al.* 1996; Mote *et al.* 2001; Sillitoe 2005). Copper-bearing solutions commonly follow the gravel-bedrock contact or permeable layers within the gravel beds (Sillitoe 2005), although Cu transport in surface runoff has recently been proposed for the Tesoro exotic Cu deposit in Chile (Fernandez-Mort *et al.*, 2018). While exotic Cu deposits are attractive exploration targets for oxide Cu in their own right, elevated Cu content within gravel cover may serve to vector towards supergene enriched and primary hypogene porphyry Cu ore bodies up gradient. However, detection of oxidized copper mineral species under gravel cover by traditional geochemical or geophysical means remains challenging.

Radon is an inert radioactive gas produced in the decay chain from the decay of ^{238}U and ^{232}Th to ^{206}Pb and ^{208}Pb respectively. The isotope produced from ^{238}U is ^{222}Rn (half-life 3.84 days) whereas ^{220}Rn (half-life 55.6 seconds) derives from ^{232}Th. The immediate parent of radon gas is the alkali earth element ^{224}Ra (half-life 3.6 days) for the ^{232}Th decay series and ^{226}Ra (half life 1,602 years) for the ^{238}U decay series.

Uranium can, together with Cu, be hydromorphically transported in oxidized and acidic meteoric fluids. Oxidized U^{6+} is easily transported over a wide range of pH conditions as different aqueous complexes (e.g., Krupka & Serne 2002), whereas reduced U^{4+} is generally immobile. Chrysocolla, a common mineral in exotic Cu deposits, has been found to contain significant quantities of U (in some cases more than 0.1 wt%: Barton 1956; Kahou, *et al.* 2020). Radon gas generated from the decay of U via Ra can ascend through permeable gravel deposits for distances up to 200 m or

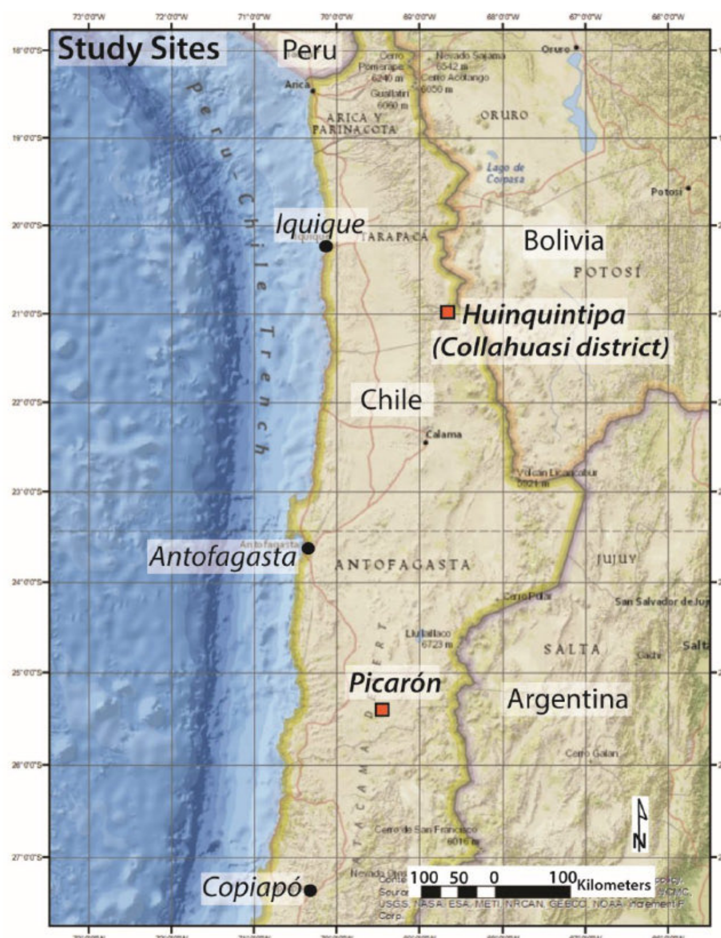

Figure 1. Location of Picarón and Huinquintipa in northern Chile.

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more (Ramola *et al*. 1989) and serve as an indicator for secondary Cu minerals in exotic Cu deposits. Since Ra is much less mobile than U in the supergene environment (Ball *et al*. 1991), it is unlikely that significant quantities of Ra were removed from the areas enriched in U and Cu and elevated Rn counts are assumed to be directly related to elevated U concentrations.

Detection of Rn has long been used for U exploration (e.g., Gingrich 1984). Radon detectors, normally applied to monitor radon emissions in buildings, can be used to detect secondary U and, as a proxy, Cu mineralization under gravel cover. We tested the viability of this approach at the largely mined out Huinquintipa exotic Cu deposit, Collahuasi district and the buried Picarón prospect, Victoria district, both located in northern Chile (Fig. 1).

Methods

Radon emissions were measured using alpha decay testing devices (Accustar AT-100). The AT-100 device consists of a plastic cup which contains nitro cellulose film that records alpha decays of Rn as fission tracks. No distinction can be made between Rn derived from Th or U by this device but, based on typical concentrations in the rock types present at the study sites and the much longer half-life of ^{222}Rn, the detected Rn is assumed to be largely derived from ^{238}U via ^{226}Ra. The AT-100 devices were attached to the bottom of regular plastic cups and placed in Tyvek bags (Fig. 2). These bags then were buried in the soil, with the opening of the plastic cups downward, at about 30 cm depth (Fig. 2). The devices were left in the soil for 10-12 days. After removal, the Rn detection devices were sealed in Ziploc bags and sent for analysis to Accustar Labs in Ward Hill, Massachusetts, United States. The results are reported in bequerel per cubic meter (Bq/m^3) which is a measure of alpha decays per second and volume. According to the specifications given by Accustar, counting uncertainty is 12%. Field duplicates collected in the program are within this 12%. Blanks were used to establish background emission values of 30 to 81 Bq/m^3. For further specifications see http://www.accustarlabs.com/ or http://www.accustarcanada.com/.

A limited number of rock samples of exotic mineralization from Huiquintipa, as well as samples from reverse circulation (RC) drilling chip trays from Picarón were analyzed by the 4-acid digestion ultra-trace method (MA250) at Bureau Veritas Mineral Laboratory, Vancouver, Canada. The aim of these analyses was to measure the concentration of U present and its relationship to Cu, Mn and Fe concentrations. Sequential leach methods were not applied and therefore no inference on mineralogical association of U can be made. At Picarón, rock-fragments were collected from drillhole RC-11-17 every other 2 m interval from surface to 24 m depth and at every 2 m interval throughout the mineralized zone (24—46 m). Additional samples at approximately 10 m spacing were collected below the mineralized zone. From RC-11-12, samples were only collected from the mineralized zone (26-36 m) at every 2 m interval. Note that coarse rejects from RC drilling are no longer available. Thus, the type and amount of sample material is not fully representative for the drillhole intercepts due to the small amount of material, sample bias and repeated sample wetting for logging. The original aqua regia digestion multielement ICP-AES analyses from the drill campaign generally yielded U values below detection limit for that method (i.e. < 10 ppm), which is too high to resolve variations of U content associated with oxide Cu minerals in gravels.

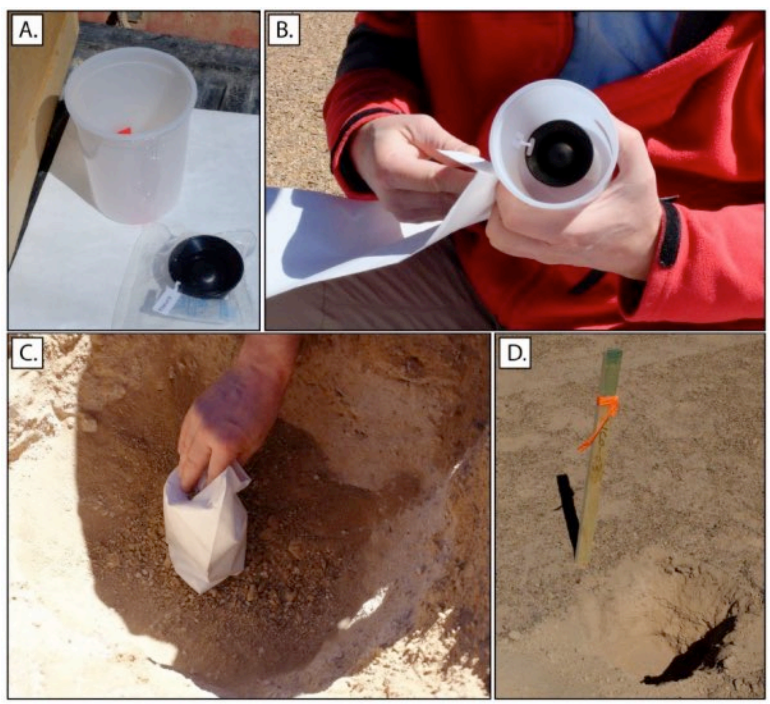

Figure 2. Radon alpha track test devices and method for Rn in soil testing. A) Accustar AT-100 device (black), plastic cup and Tyvek bag. B) AT-100 device attached to the bottom of the plastic cup to be inserted in a Tyvek bag. C) The Tyvek bag containing the plastic cup with the AT-100 in a test hole at ca. 30 cm depth, before being buried for 10 days. Note that the opening of the plastic cup is downward. D) Typical test site.

At Picarón, soil samples were collected at 5 and 30 cm depth from 26 sites from two transects across the exotic mineralization. Samples (n=52) were analyzed by ICP-MS following deionized water leach and aqua regia digestion at Bureau Veritas Mineral Laboratory, Vancouver, Canada. The aim of these analyses was to test whether traditional soil geochemical methods are capable of detecting oxide copper mineralization under cover and whether high Rn counts can be explained by near-surface accumulation of U.

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Huinquintipa study site and Rn detector results:

Huinquintipa is an exotic Cu deposit from which approximately 29 Mt at 1.07% Cu has been mined (Nelson *et al.* 2007). It is located to the west of the Rosario Porphyry (Fig. 3) and hosted within gravel deposits which have been partly eroded after the formation of exotic Cu mineralization. Copper is mostly hosted in chrysocolla and a variety of Mn-bearing oxides (Fig. 4). Most of the Cu has been mined out and the Cu-bearing manto is only locally preserved (Fig. 4).

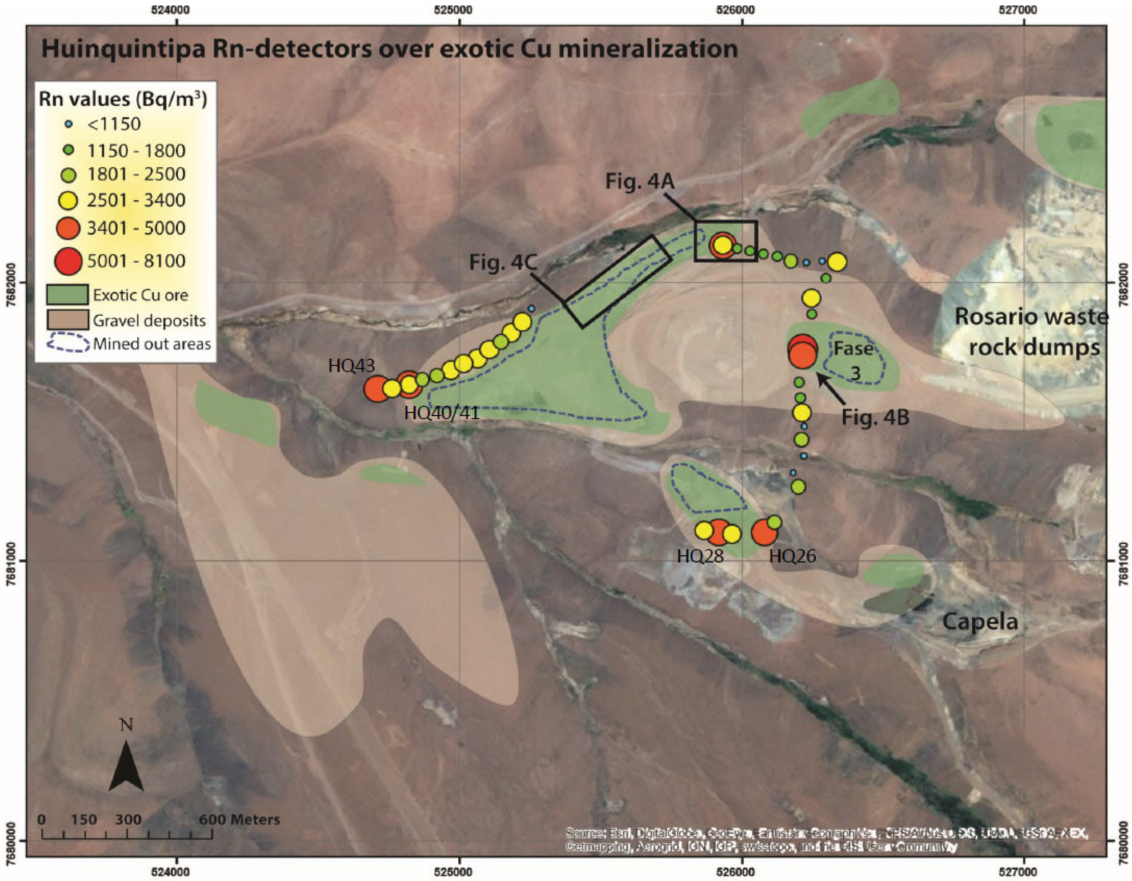

Figure 3. Radon detector results over Huinquintipa, Collahuasi. Locations illustrated on Figure 4 are indicated; additional sample locations mentioned in the text are labelled. Background imagery is from ESRI digital globe.

Figure 4. (below) Green dashed lines outline exotic Cu mineralization. Approximate locations of Rn detector sites and corresponding radon values (Bq/m^3) relative to exotic Cu mineralization at Huinquintipa. A) Looking east at northern extent of main manto. B) Looking east at the exposed relict exotic Cu mantos of Fase 3. C) Main exotic Cu manto as it appears along strike to the southwest of area shown in A.

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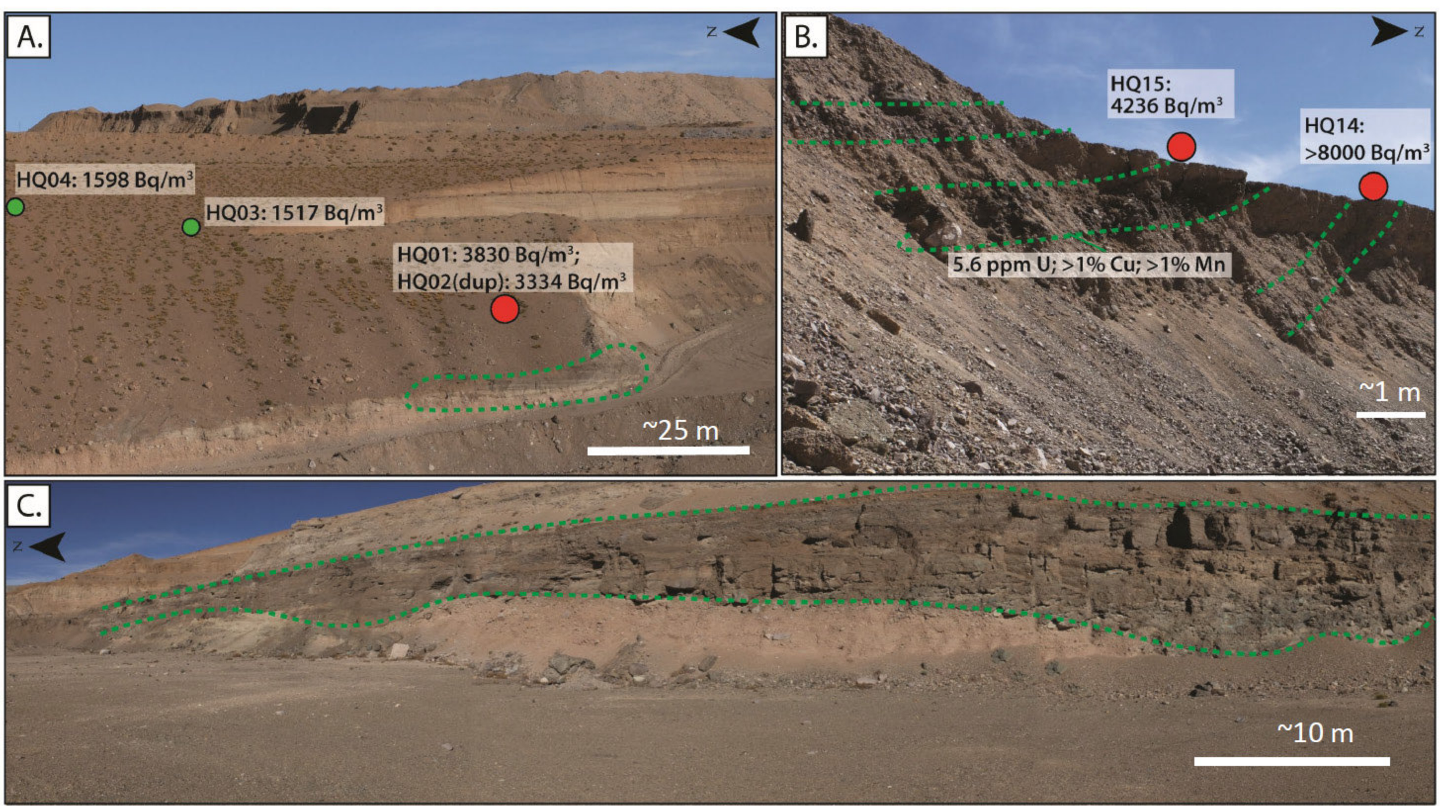

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A total of 42 Rn detectors were placed in a north to south transect across the Fase 3 zone with two short east to west oriented transects at either end (Fig. 3). An additional northeast oriented transect was sampled at the western limit of the deposit. The Rn counts range from 340 Bq/m^3 to 4850 Bq/m^3 with one detector registering alpha decays above the upper detection limit of 8000 Bq/m^3. A few of the detectors (HQ01, HQ14, and HQ15) were placed near and above exposed portions of Cu mantos (Figs. 3, 4) and recorded among the highest Rn counts (> 3400 Bq/m^3), in one case exceeding the upper limit of detection (HQ14). Additional high values were obtained from detectors placed above gravel-hosted exotic mineralization in the southern E-W transect (HQ26 and HQ28) and from those placed west of the known extent of exotic mineralization. The latter detectors were not placed above gravel deposits but over bedrock (HQ43) and at the bedrock-gravel contact (HQ40 and duplicate HQ41). Conversely, the lowest Rn counts came from several detectors placed where gravels were eroded south of Fase 3 zone (< 1150 Bq/m^3). Low values were also obtained from areas where mineralization has been mined out, and where detectors were located over bedrock except for those west of the known exotic mineralzation mentioned above.

The overall dataset demonstrates that Rn counts are high in detectors placed above known exotic mineralization and low in areas where gravels and possible exotic mineralization were eroded. Thus, high Rn counts are a good proxy for the presence of exotic Cu mineralization. However, the detectors placed at the far west of Huinquintipa are at odds with this interpretation because high Rn counts were detected over bedrock, albeit close to the gravel contact. Exotic Cu mineralization may not only be hosted in gravels but can also be hosted in fractures in the bedrock below gravel, such as described from e.g., Damiana in the El Salvador district (Mote *et al.* 2001). Whether bedrock-hosted exotic Cu mineralization is present in fractures at Huinquintipa remains to be confirmed.

Picarón study site and Rn detector results:

Picarón is an exotic Cu prospect, perhaps better described as a hydromorphically transported Cu anomaly, located some 4.5 km west of the small, historically mined Vaquillas epithermal deposit (Fig. 5). Modest Cu grades (generally <0.5 wt.%) are hosted in Mn-bearing oxides observed in RC drill chips near the contact between gravel and bedrock at approximately 32-38 m depth below surface. The bedrock is dominantly Jurassic limestone whereas the gravel

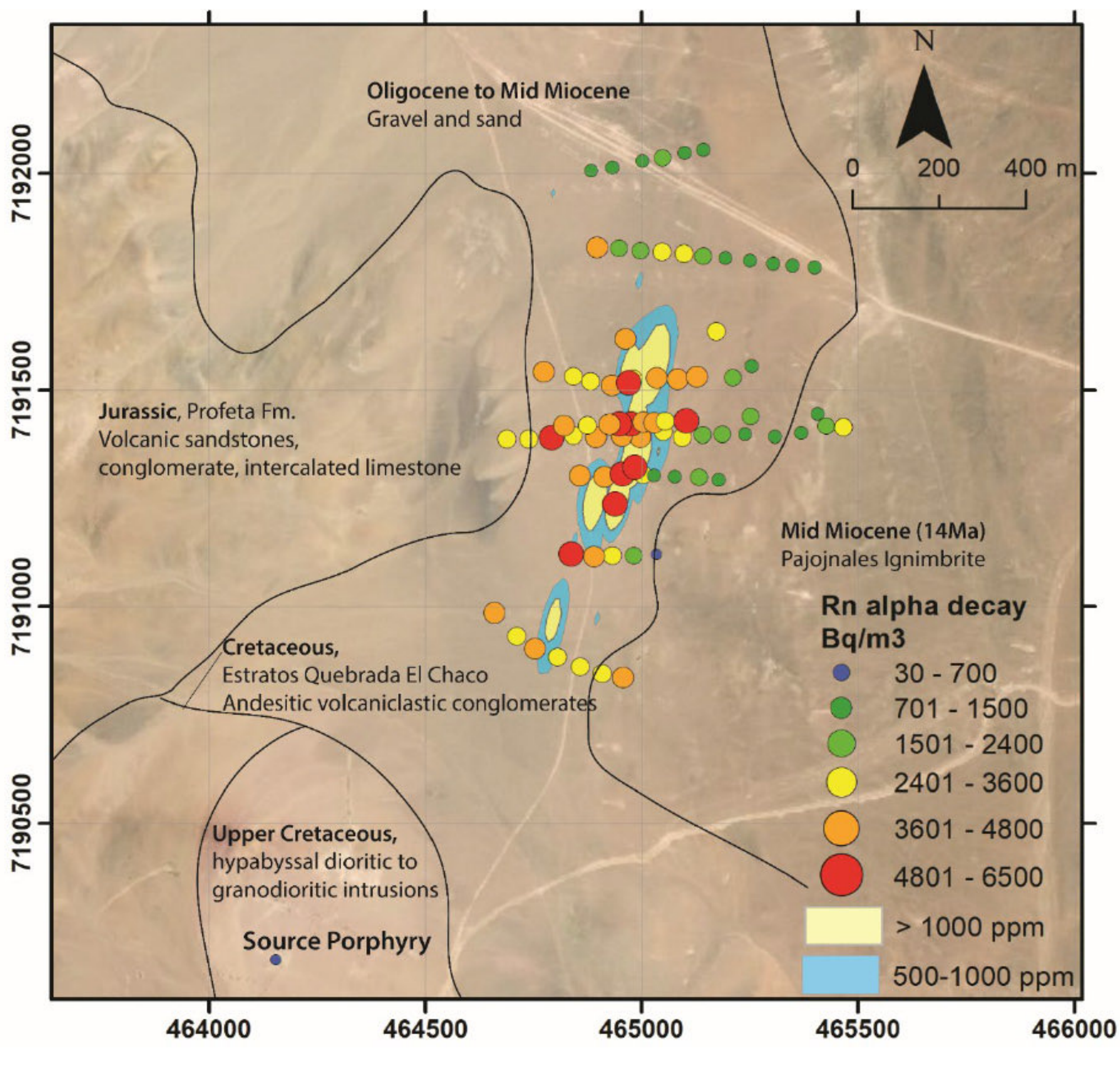

Figure 5. Radon detector results from Picarón. Copper grade contours are projected to surface from approximately 35 m depth. Geology from Venegas et al. (2017). Background imagery is from ESRI digital globe.

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sequence includes increasing amounts of intermediate to felsic volcanic and intrusive clasts from the bedrock upward. The mineralization is fully concealed by gravel deposits and has been defined by 6 RC-drillhole fences with 3 to 6 drill holes each. The likely bedrock source of Cu is a small porphyry prospect exposed about 1 km to the SW of the exotic mineralization.

A total of 86 Rn detectors were placed over the zone of oxide Cu and over nearby adjacent background (Fig. 5). An additional 12 detectors were deployed near Quebrada El Chaco (Fig. 6), some 7 km to the SE from Picarón, to test an area without exotic Cu mineralization in the gravel. The detectors of the background control survey all showed Rn values of less than 2312 Bq/m^3 (Fig. 6).

The Rn decay counts over Picarón range from about 300 Bq/m^3 to about 6500 Bq/m^3. Detectors placed over Cu mineralization in the gravels yielded Rn values of > 3500 Bq/m^3 (Fig. 5). The lowest Rn value pertains to a sample placed over bedrock where oxidized primary mineralization is exposed (PICO-18: 333 Bq/m^3), and areas to the east of the mineralized zone (Fig. 5).

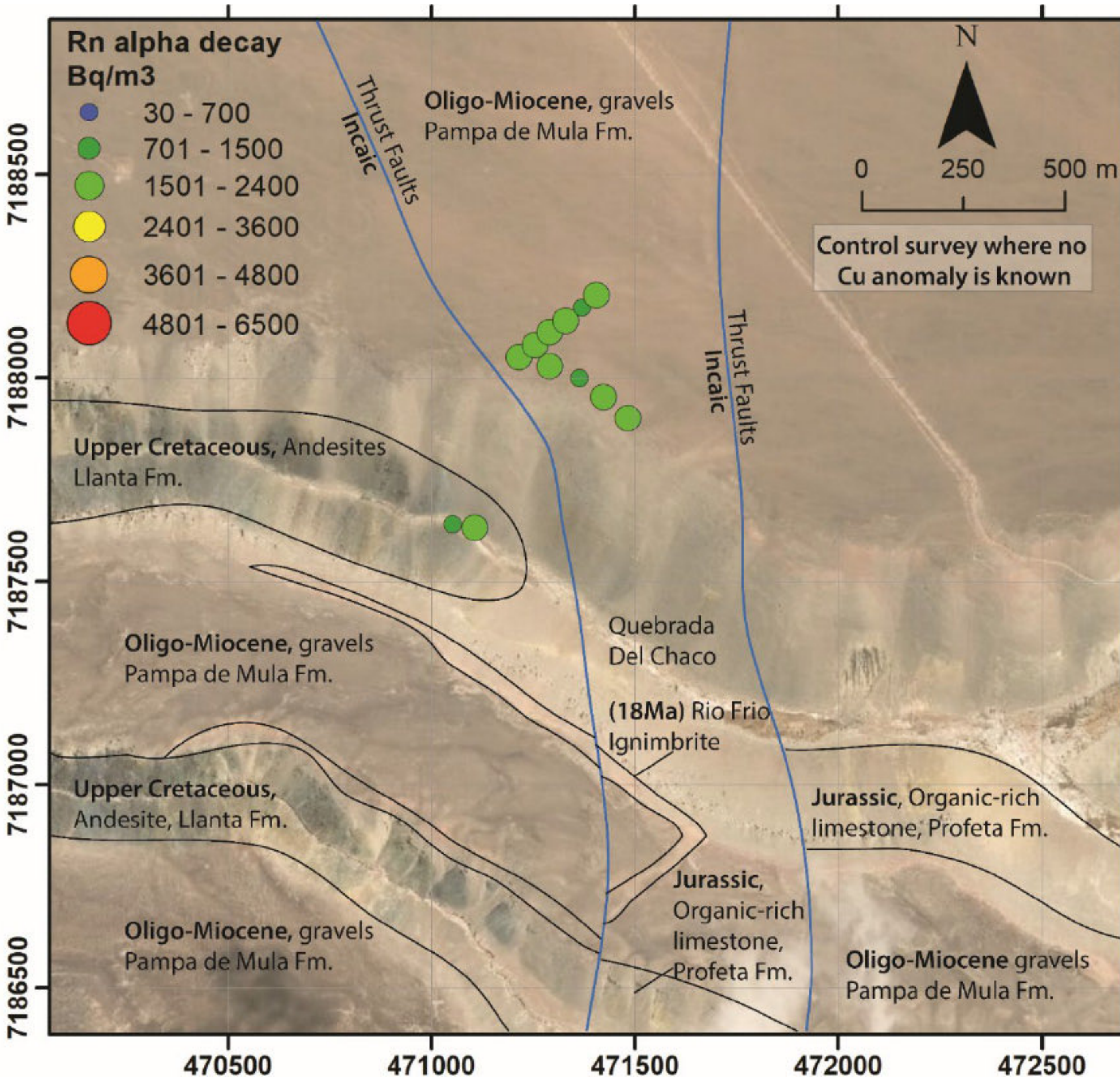

Figure 6. Radon detector results from the control survey at Quebrada del Chaco, located approx. 7 km to the SE from Picarón. Geology from Venegas et al. (2017). Location of thrust faults under gravel cover is inferred. Background imagery is from ESRI digital globe.

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Association of Cu with U

The use of Rn α-decay to detect anomalous U and by inference Cu is based on the assumption that exotic Cu mineralization is characterized by elevated U concentrations. This has been demonstrated as early as the 1950s (Barton 1956) and recently in the Atacama Desert (Kahou *et al.,* 2020). An attempt was made to investigate the presence of elevated U in Cu mineralization in this study as well. The ore samples from Huinquintipa all yielded more than 1% Cu (above upper LOD) and U concentrations between 5 and 21 ppm, whereas all samples analyzed from Picarón contained <5 ppm U and <0.35% Cu. From limited macroscopic observations, it is evident that the highest U contents pertain to those samples with chrysocolla as the dominant Cu mineral and not black Cu-bearing Mn-oxides. In a downhole diagram of Picarón drillhole RC-11-17, U, Cu and Mn show a co-variance through the mineralized zone (Fig. 7). However, the highest Cu values were obtained from a sample representing a depth of 50-52 m and this sample does not have elevated U. Conversely, the highest U value of 4.8 ppm was obtained from a sample at 82-84 m. This depth corresponds to a transition between dark grey and beige-grey limestone, which is interpreted as the redox boundary and therefore may explain the slightly elevated U.

Overall, the geochemical data indicate that exotic

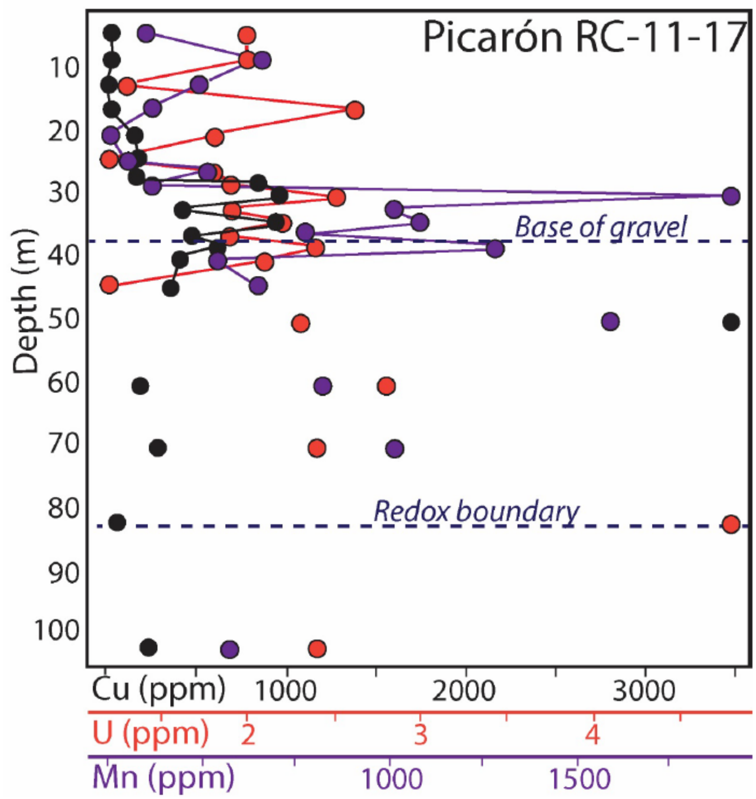

Figure 7. Total digestion data from rock chips taken from RC trays. Downhole plot of Cu, Mn and U concentrations from Picarón RC hole 11-17. Symbols are only connected where consecutive samples were analyzed. Note the co-variance of Cu-U and Mn through the mineralized zone immediately overlying the base of gravel/bedrock contact.

Cu mineralization at Huinquintipa contains U concentrations significantly above crustal abundance. However, additional sampling and sequential leach analyses would be required to elucidate the mineralogical associations of Cu and U and establish background U and Cu concentrations. Limitations of sample material accessible at Picarón does not allow establishing an association of Cu with U with confidence beyond the suggestion of a co-variance through the mineralized zone.

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Selective leach geochemistry of surface materials above exotic Cu mineralization at Picaron

Soil samples were taken at the gravel surface, some 30-40 m above bedrock contact. The surface materials at Picarón are slightly alkaline with a soil-slurry pH range of 7.3 to 9.5, and a similar range of values from the samples taken at 5 cm and 30 cm depth. Soil conductivity at 5 cm depth ranges between 30 and 835 µS/cm^3 (15-440 ppm total dissolved solids (TDS)). At 30 cm depth, soils are substantially more conductive with values ranging from 76 to 2820 µS/cm^3 (38-1570 ppm TDS) with one outlier for which 5620 µS/cm^3 (3190 ppm TDS) recorded. There is no systematic difference between samples collected above exotic mineralization and those over background (Fig.8).

Chlorine and Na as well as Ca and S values from the

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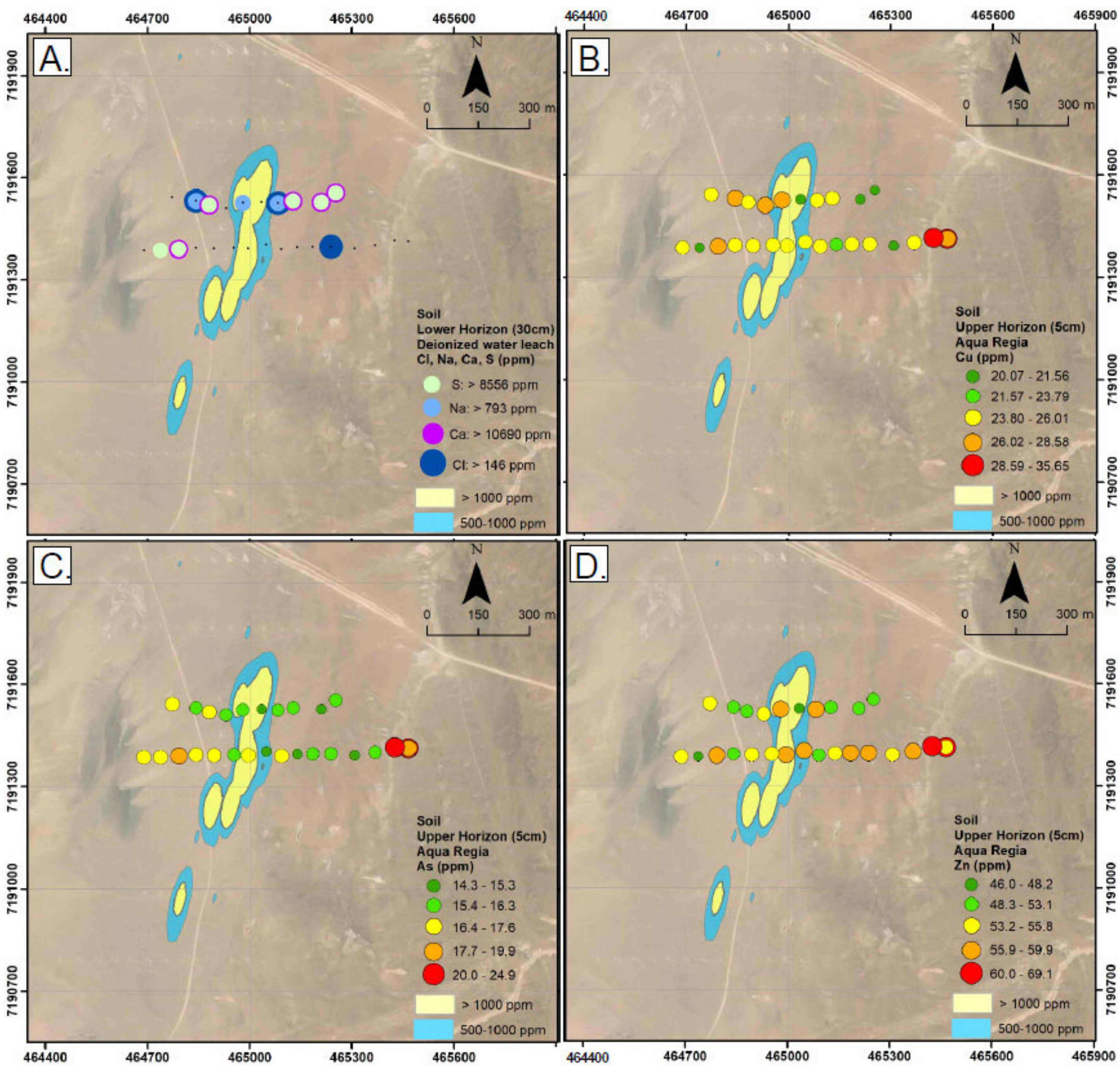

Figure 8. Soil analysis results for: A) Cl, Na, Ca, and S by deionized water leach ICP-MS from samples taken at 30 cm depth. Soil analyses results by aqua regia digestion ICP-MS of B) Cu, C) As and D) Zn from samples taken at 5 cm depth at Picarón. Copper grade contours are projected to surface from approximately 35 m depth. Background imagery is from ESRI digital globe.

deionized water leach of lower soil horizon samples spatially correlate with the conductivity results, indicating that much of the TDS can be explained by the presence of gypsum and salts in the soil (Fig. 8A).

Soil analyses by aqua regia digestion indicate that elements commonly used as pathfinder elements in Cu exploration (e.g., Cu, Mn, Zn, As, Mo, Ag, and U) have only a narrow range of values and are generally not elevated in the soil above the exotic mineralization (Fig. 8). Uranium ranges between 0.7 and 1 ppm, Cu between 20 and 36 ppm (Fig. 8B). Copper and As concentrations are highest in samples collected over an older gravel surface above ignimbrite to the east (Fig. 8). This pattern is interpreted as an effect of the age of the gravel plain and/or dominant rock units in the gravel, and not as an indication of underlying mineralization.

Soil analyses by deionized water leach reveal the presence of weakly bound or soluble ions. Elevated values of chalcophile elements in association with highly conductive material may indicate accumulation in surface material due

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to capillary transport or seismic pumping of metal-bearing fluids along structures above oxidizing sulfide ore bodies (Brown *et al.* 2019). At Picarón, there is no discernible systematic pattern of Cu anomalies over concealed oxide Cu mineralization, or a relationship between elevated Cu and high TDS values in association with structural trends. Copper values are < 40 ppb (Fig. 9). The upper soil horizon, however, has slightly higher Cu values than the lower horizon, which is consistent with published results from Spence, where Cu is elevated near the top of the soil and lower concentrations occur at depth (30 cm; Cameron & Leybourne 2005).

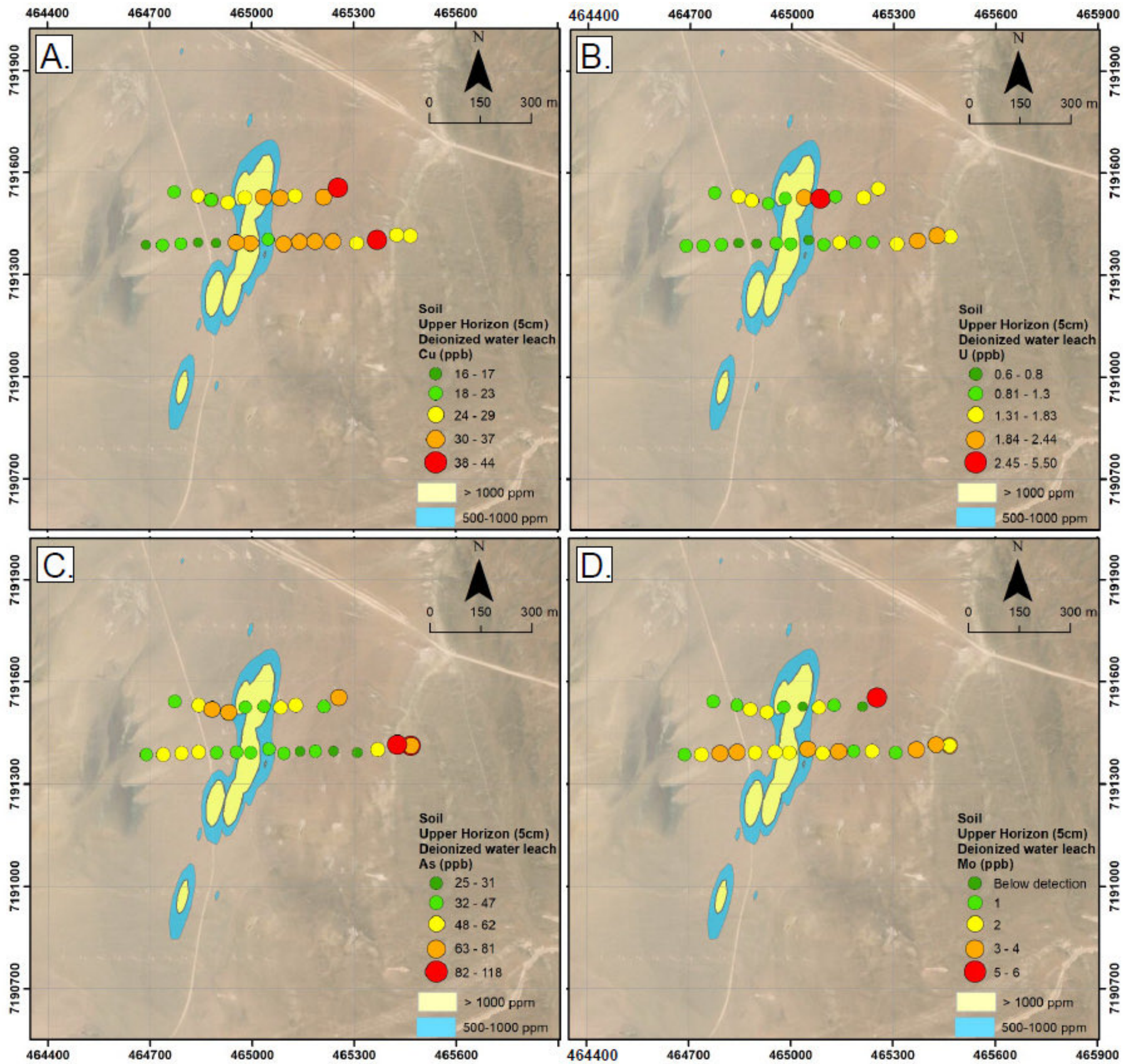

Figure 9. Soil analyses results by deionized water leach ICP-MS of A) Cu, B) U, C) As, and D) Mo from samples taken at 5 cm depth at Picarón. Copper grade contours (blue and yellow) are projected to surface from approximately 35 m depth. Background imagery is from ESRI digital globe.

Discussion

The results demonstrate an empiric spatial relationship of Rn counts above or near exotic Cu mineralization. However, additional research is required to better understand where elevated U is located within the gravels relative to exotic Cu mineralization. Likewise, the influence of Ra mobility and concentration on the Rn counts requires additional work. The

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soil data from Picraón indicate that elevated Rn counts cannot be explained by U transported along faults through gravel cover and precipitated or adsorbed to soil particles near surface. However, high Rn counts could theoretically also be explained by decay from Ra transported along permeable structures, adsorbed to soil matrices and concentrated near surface. Radium 226 is the immediate parent isotope of ^{222}Rn in the U-series decay chain and has a half-life of 1602 years which is long enough for such transport and accumulation to occur. Radium was not analyzed, but it is more easily transported in acidic fluids than under the mildly alkaline conditions observed currently at Picarón (Rachkova *et al.* 2010). Radon derived from Ra transported along faults and concentrated near surface is therefore an unlikely explanation for the Rn patterns at Picarón. Moreover, no evidence for faults intersecting the gravel surface has been observed. Conversely, it is possible that low Rn counts directly over bedrock, including over primary mineralization at Picarón, can be explained, in part, by poorly developed soil, limiting the possibility for Ra to accumulate.

Radon gas, on the other hand, can diffuse over 200 m or more in unconsolidated, dry gravels (Ramola 1989). The water table at Picaron is currently 60-80 m below surface and therefore below the exotic mineralization. Water saturation therefore did not limit the movement of Rn gas from the exotic Cu mineralization. What remains to be shown is to what extent atmospheric conditions (pressure and temperature) can influence the results.

The primary source of Cu is chalcopyrite from porphyry Cu mineralization, likely hosted in quartz-sericite altered rocks which contained enough pyrite to generate the acid required for Cu transport during supergene oxidation (cf. Chavez 2000). The acidic fluids generated during supergene oxidation were probably also capable of dissolving and mobilizing U (Tieh & Ledger 1981). In addition, hypogene sericitic alteration may have also played a key role in destabilizing primary U-bearing phases such as apatite, feldspars and zircon and making U available for aqueous transport (Bouzari *et al.* 2016; Geisler et al. 2007), enhancing the availability of U during supergene oxidation of porphyry systems compared to unaltered granitoids.

Conclusions

The results from Huinquintipa and Picarón suggest that Rn detecting devices are a potential low-cost exploration tool for buried oxide Cu mineralization. Anomalous Rn is detected near concealed oxide Cu mineralization transported laterally from its primary source at both Huinquintipa and Picarón. Radon detecting devices are useful not only for the exploration of exotic Cu ore bodies but can, as demonstrated at Picarón, detect Cu anomalies below ore grade level under the gravels, which then can be used as a vector towards primary Cu targets upstream. In these areas, traditional soil geochemistry fails to detect oxide Cu mineralization buried under cover.

Acknowledgements

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