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Mineralogical and geochemical characterisation of "pouhons" in the Stavelot-Venn Massif

MARTIN DEPRET

Laboratory of Mineralogy, Geology Department, University of Liège, Allée du Six-Août B18, 4000 Liège, Belgium.

Numerous naturally CO_2-rich mineral water springs, locally called *'pouhons'*, occur in southeast Belgium. These water sources are mainly located in the Cambro-Ordovician Stavelot-Venn Massif (SVM). *Pouhon* waters show a particular composition with a high content of iron, manganese, and lithium and are affected by complex subsurface processes that are poorly known. Indeed, hydrogeological interpretations are relatively difficult in the SVM due to its complex geological setting. *Pouhons* are also characterized by a red-orange colour resulting from iron hydroxide precipitation near the land surface. Radon measurements have shown that these ferruginous deposits are weakly radioactive (Vanderschueren, 2011). This study aimed to improve the global understanding of *pouhons* and more specifically determine the origin of radioactivity. Mineralogical and geochemical analyses were therefore performed on various rocks of the SVM.

Radiometric surveys carried out by the Geological Survey of Belgium from 1955 to 1995 showed that the black slates of the La Gleize Formation present radioactive anomalies (Charlet et al., 1977; Charlet et al., 1983). These rocks are enriched in HFSE (Pb, U, Y, Ce, Zr, Ti, Nb) and depleted in transition metals (Co, Ni, Cu, Zn). However, the uranium contents are relatively low and do not exceed 20 ppm. Specific minerals such as florencite-(Ce), monazite-(Ce), xenotime-(Y), and zircon have been detected in these rocks and are probably at the origin of the radioactive anomalies. Uranium was therefore gradually leached from these minerals, transported in solution, and finally concentrated in ferruginous muds. These muds are mainly composed of goethite (most often amorphous), residual quartz, and calcite in some samples. The most probable hypothesis is that uranium is adsorbed in small concentrations on goethite surface.

Schists of the Ottré Formation show more various mineralogy and geochemical composition. They are enriched in Fe_2O_3 and MnO due to the occurrence of hematite, spessartine, and rhodochrosite. These rocks are also rich in lithium compared to other formations of the SVM, thus probably explaining the high lithium contents observed in these groundwaters.

Pouhon waters, therefore, seem to be strongly associated with the Ottré Formation. However, these water springs are mainly located in rocks of the Revin Group and the La Gleize Formation, which appears to be the main source of uranium at the origin of radon activity. Geological, mineralogical, geochemical, and hydrogeological data seem to show that *pouhons* are not associated with a specific formation but that their chemical composition reflects complex interactions with rocks of varied mineralogy and geochemical composition.

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Volatile contents in magmas from ocean islands: Example from Pico, Azores

JOREN CELIS[1], OLIVIER NAMUR[1], NIELS HULSBOSCH[1] & THOMAS VAN GERVE[1]

[1]*KU Leuven, Department Earth and Environmental Sciences, Division of Geology, Celestijnenlaan 200E, B-3001 Leuven, Belgium.*

Mantle melting is one of the most important petrological processes taking place on Earth. The Earth's crust is formed by mantle melting followed by melt differentiation. The Azores region is known to have a very thick oceanic crust, providing an ideal setting for studying mantle processes. The objective of this study exists of two main parts. Firstly, to develop a Raman calibration methodology based on the work of previous authors, which can be used to measure water contents with the Raman microspectrometer at KU Leuven. The second objective is to study the influence of volatiles of basalts of Pico Island on mantle melting underneath the Azores.

The Azores Archipelago is located close to the Azores Triple Junction, east of the Mid-Atlantic Ridge. The islands of the Azores are aligned perpendicular to the Mid-Atlantic Ridge. The geochemical composition varies between islands and even on individual islands. Pico is one of the central Azores islands and consists of three main volcanic complexes: Topo-Lajes Complex, Pico Ridge Complex, and Pico Stratovolcano. A NW-SE-oriented fault intersects the Pico Stratovolcano. Magmas that erupted from the stratovolcano are relatively primitive.

Raman analysis was conducted on three basaltic glass standards with fixed water content. Different acquisition parameters were evaluated to obtain Raman spectra with a distinct topology and high signal-to-noise ratio. A Python script was coded which handles the calibration in four main steps. Raman spectra were treated with the Long correction, their baselines were subtracted with cubic spline interpolation, the internal standard $R_{w,s}$ is directly linked to the water content and external basaltic glass standards with a fixed water content were used to validate the calibration. Based on the semantic definition of the baseline and the results of the calibration, it is concluded that the calibration method of Le Losq et al. (2012) is preferred over the method of Di Genova et al. (2017).

Olivine crystals from lavas from Pico were handpicked and polished one by one to check for melt inclusions. They were mounted in epoxy for Electron Probe Micro Analyser analysis to obtain their major element compositions. This data showed chemical disequilibrium between the olivine-liquid pair, indicating post-entrapment crystallisation. The data was corrected until equilibrium was reached. The corrected major element compositions were compared to those of Métrich et al. (2014), which showed that the samples in this study are more primitive.

The olivine crystals were remounted in indium for Secondary Ion Mass Spectrometry to study their volatile contents. The samples are highly enriched in water (0.20–2.55 wt%). The spread in volatiles might indicate a geochemically heterogeneous mantle below Pico or re-equilibration of melt inclusions, which should be investigated in further studies using highly incompatible and slowly diffusing trace elements. Both the major element and volatile compositions were used to obtain crystallisation and mantle potential temperatures below Pico. The anhydrous crystallisation temperature is calculated to be ~1221 °C and the hydrous ~1196 °C. The primary magma composition is derived for the melt inclusions to calculate a dry mantle potential temperature of ~1394 °C and wet mantle potential temperature of ~1367 °C. The resulting mantle excess temperatures of 44 °C and 17 °C are too low to be caused by a hot spot, where excess temperatures of 100 - 300 °C are often estimated. Previous authors derived excess temperatures of ~130 °C below Pico, which might indicate a combination of a hot spot and a so-called "wet spot".

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Applicability of uncertainty analysis to groundwater environmental risks through Fault Tree Analysis and Monte Carlo simulations

ROBIN GLAUDE

Università di Bologna, Italy - ULiège, Belgium.

The Anthropocene epoch initiated by human influence on its Earth System (Atmosphere, Biosphere, Hydrosphere) leads to an irreversible change: Global Warming. Since Climate Change would increase the occurrence's probability of undesired events (sea-level rise, floods, extreme droughts, and so on), it leads to the concept of environmental risk which refers in its most basic definition as the combination of the consequences of an undesired event (Vulnerability) and its probability of occurrence (Hazard). As a consequence, it is necessary to study this alteration of existing natural processes, including the ones related to hydrogeology, in a probabilistic manner.

The present thesis aims to study the occurrence's probability of two particular groundwater risks: the generation of thermokarst lakes in permafrost environment and its subsequent thermal consequences in the surroundings as well as seawater intrusion inducing saltwater contamination in abstraction pumping wells. These processes are dependent on physical parameters to which is attached uncertainty. Consequently, two uncertainty analysis methods have been applied to determine the failure's probability of these undesired events: Fault Tree Analysis and Monte Carlo Simulation.

Besides the rough approximation performed to evaluate the probability of thermokarst lake generation (48%) and talik development under this latter (73%) by means of Fault Tree Analysis, these high failure probabilities translate the urge to restrain Global Warming due to its irreversible effects on permafrost environment. These include the thawing of the permafrost and the consequent releasing of its trapped methane into the atmosphere.

On the other hand, Monte Carlo simulations have been performed to compare different scenarios related to seawater intrusion in the Akrotiri aquifer in Cyprus. The results once again translate the disastrous effect of climate change regarding the probability of occurrence of these unwanted events. Indeed, a failure probability around six times greater (43%) is observed in the climate change scenario with respect to the reference scenario (7%).

Uncertainty analysis is a good methodology to apply to environmental concerns to quantify the occurrence's probability of these undesired events. This would urge public authorities to perform decision making to avoid or reduce the failure's probability of these groundwater issues that have irreversible consequences on its surrounding environment.

Application of Laser-Induced Breakdown Spectroscopy (LIBS) to gold-bearing quartz veins from Abitibi greenstone belt (Val d'Or, Canada)

LAURA LEMINEUR[1*], JEAN-MARC BAELE[1], BERTRAND ROTTIER[2] & GEORGES BEAUDOIN[2]

[1] *Université de Mons, Géologie Fondamentale et Appliquée, 20, place du Parc, 7000 Mons, Belgique.*
[2] *Université Laval, 1065 Avenue de la Médecine, G1V 0A6 Québec, Canada.*
[*] *corresponding author: laura.lemineur@student.umons.ac.be*

The Val d'Or district, located in the Archean Abitibi greenstone belt, Canada, hosts important gold and base metal resources. In this district, gold is sourced from orogenic gold deposits formed by quartz-carbonate and quartz-carbonate-tourmaline veins accompanied by pyrite and/or chalcopyrite (Desrochers et al., 1996). The auriferous veins formed in two mineralization phases of hydrothermal origin dated before 2690 Ma and after 2680 Ma (Robert, 1990; Couture et al., 1994). They are actively mined and exploration efforts are needed to sustain mining activities.

This work consisted of assessing an in-house developed Laser-Induced Breakdown Spectroscopy (LIBS) as a new exploration tool for discriminating auriferous from barren veins. LIBS is an emerging geochemical technique capable of fast analysis of un- or minimally prepared samples. It employs high-energy laser pulses that are focused on the surface of the sample to create a plasma. The plasma light is then analyzed with a spectrometer and the detected atomic emission lines can provide qualitative geochemical information in seconds, including major, minor, and trace elements.

LIBS analyses were performed in both spot and raster (mapping) modes on a selection of quartz and sulfides from auriferous and barren veins. Both were analyzed separately as

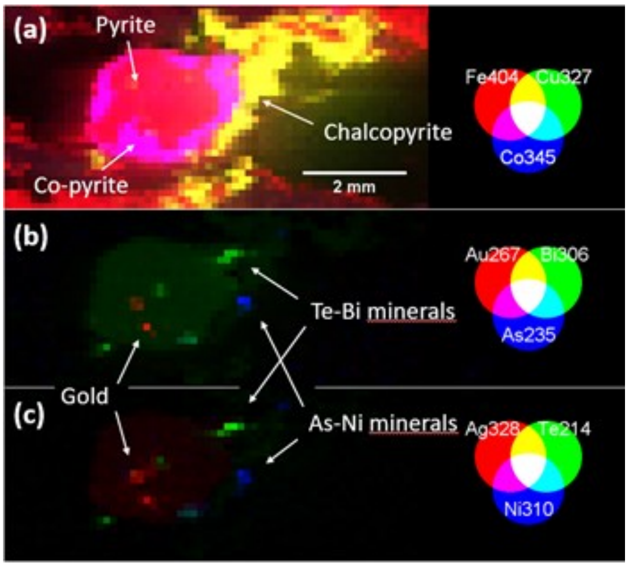

Figure 1. Red-green-blue composite images of the LIBS intensities of selected elements in sulfides in auriferous quartz veins from Val d'Or, Canada. (a) Image of Fe, Cu, and Co. (b) Image of Au, Bi, and As. (c) Image of Ag, Te, and Ni.

LIBS spectra of sulfides are very complex due to the high density of emission lines from iron. This necessitated in-depth analysis of the spectra to extract accurate line intensities of the chemical elements. The dataset was further subjected to bi- and multivariate statistical analysis such as Principal Component Analysis (PCA).

Our results suggest that boron and, to a lesser extent, silver could be used as indicators of gold as these elements are preferentially detected in auriferous quartz veins. The correlation of boron with both magnesium and aluminum points to Mg-tourmaline (dravite) inclusions, which are indeed known to precipitate during the second gold mineralization phase in the Val d'Or district. However, silver is unrelated to boron but to copper and iron, suggesting that this element is associated with pyrite and chalcopyrite inclusions, which are common in both mineralization phases.

In sulfides, the detection of gold by LIBS correlates well with the abundance of gold inclusions that were observed under the microscope. Furthermore, LIBS analyses confirmed that sulfides concentrate most of the gold and show that Ag, Cu, Co, Te, and Bi are associated with higher gold contents. These elements could then be used as indicators of gold in sulfides. The distribution of these elements was revealed in the LIBS maps, which show telluride inclusions and cobaltiferous pyrite rims associated with gold (Fig. 1).

Despite our initial research plan that had to be scaled down because of the Covid crisis, our study based on optical microscopy and LIBS already provides a wealth of information and opens promising perspectives for fast and effective gold exploration using LIBS technology, including commercially available handheld systems.

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