Applied Geochemistry 145 (2022) 105423

Contents lists available at ScienceDirect

Applied Geochemistry

journal homepage: www.elsevier.com/locate/apgeochem

Biogeochemical prospecting for gold at the Yellowknife City Gold Project, Northwest Territories, Canada: Part 1 - Species optimization

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ARTICLE INFO

Editorial handling by: Patrice de Caritat

ABSTRACT

The Yellowknife City Gold Project (YCGP), located close to the city of Yellowknife, Northwest Territories, Canada, is exploring and drilling the extension of the Au-bearing shear zones that host the Giant and Con Mines. A biogeochemical survey was conducted over the Crestaurum and Barney deposits, which are hosted within two well-known high Au grade shear zones at the YCGP. A total of 625 samples were collected from the alder (*Alnus incana*) (n = 76), Labrador tea (*Ledum groenlandicum*) (n = 40), black spruce (*Picea mariana*) (n = 189), and juniper (*Juniperus*) (n = 320), and analyzed using ICP-MS. Robust statistical analyses were applied to biogeochemical data to (1) identify the biogeochemical response in plant samples with respect to the underlying mineralogy, (2) detect the most preferred plant species in the study area, and (3) identify Au pathfinder elements and subsequently zones of Au enrichment. Principal component analysis (PCA) and K-means clustering analysis were employed to achieve these objectives. The preliminary statistical analysis showed that plant samples can successfully accumulate anomalous Au values up to 147 ppb. Also, it showed that black spruce and Labrador tea can accumulate higher concentrations of Au than alder and juniper. According to the PCA, mineralization/geological (PC1) and physiological (PC2) factors control the distribution of elements in plant samples. According to K-means clustering, plant samples were classified into three clusters based on the vegetation type and their affinity to accumulate suites of elements. Black spruce samples were identified as the optimum sample media for biogeochemical exploration at the YCGP as they were clustered in proximity to the mineralization/geological factor (PC1) responsible for loading vectors of Au, Ag, As, Se, Bi, Pb, Cu, Co, Fe, Zn, Cd, Tl, and Cr. Au is strongly correlated with its pathfinder elements (Ag, As, Se, Bi, Tl and Sb), specifically in the south of the Crestaurum Shear. The presented results in this study demonstrate that zones of Au enrichment can be detected using biogeochemical and robust statistical methods.

1. Introduction

The Yellowknife City Gold Project (YCGP) is located within the well-studied Archean-age Yellowknife Greenstone Belt (YGB) in the south-west corner of the Slave Craton (Armitage, 2021; Cousens, 2000). The YCGP encompasses 791 square km of contiguous land to the north, south, and east of the city of Yellowknife in the Northwest Territories, Canada (Yellowknife City Gold project, 2015). Interest in exploration of the Yellowknife City Gold Property commenced in the 1930s in sediment-hosted quartz veins of the Burwash Formation due to the presence of the nearby Giant (8.1 Moz @ 16.0 g/t Au) and Con (6.1 Moz @ 16.1 g/t Au) Mines (Armitage, 2021; Shelton et al., 2016). Although these mines were closed in the early 2000s, an unexplored, highly prospective district-scale land position in the north portion of the YCGP was actively explored (Shelton et al., 2016). The Northbelt was originally explored in 2013 by Gold Terra to assess the continuity of the shear zones and associated mineralization related to the Giant and Con Mines. This area is interpreted to represent the northern extension of the Giant Mine structure (North Giant Extension: NGX) (Kelly, 1993). Gold Terra has subsequently expanded the potential and prospective geology of the

Abbreviations: YCGP, Yellowknife City Gold Project; NGX, North Giant Extension; YGB, Yellowknife Greenstone Belt; YRFZ, Yellowknife River Fault Zone; CV_{avg}, Average Coefficient of Variation; PCA, Principal Component Analysis; ICP-MS, Inductively Coupled Plasma-Mass Spectrometry; QA/QC, Quality Assurance/ Quality Control; LLD, lower limit of detection.

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https://doi.org/10.1016/j.apgeochem.2022.105423
Received 23 March 2022; Received in revised form 21 June 2022; Accepted 4 August 2022
Available online 13 August 2022

project by the acquisition of the Southbelt (extension of the Campbell and Con shears) and Eastbelt (contiguous with the Northbelt). Since these acquisitions, various exploration techniques, including multi-media geochemical, geophysical, and geological surveys have been carried out for Au exploration in an attempt to better understand alteration assemblages and Au mineralization (Armitage, 2021; van Hees et al., 1999; Cousens et al., 2006; Baragar, 1966; Cassidy et al., 2006).

Starting in 2015, Gold Terra expanded and delineated Au potential at the YCGP using biogeochemistry. The application of plant biogeochemistry to mineral prospecting is a well-established approach in mineral exploration (Dunn and HaleSeries, 2007), using chemical analysis of vegetation to measure the presence and character of underlying mineralization (Yellowknife City Gold project, 2015). In Canadian boreal forests, coniferous trees (e.g. spruce, pine, fir, and juniper) and shrubs (e.g. alder, willow, and Labrador tea) are widespread and able to accumulate relatively high concentrations of metals (Cohen et al., 1987; Dunn, 1981, 2007). These species are considered the preferred sample medium to provide reconnaissance biogeochemical mapping of concealed bedrock. According to surveys conducted by Dunn and others in 2001 and 2002 for the Yellowknife EXTECH program, black spruce (*Picea mariana*) and Labrador tea (*Ledum groenlandicum*) demonstrated a high ability to uptake anomalous concentrations of Au and its pathfinder elements (As and Sb) (Dunn and HaleSeries, 2007; Dunn et al., 2002; DunnD., 2001). Generally, a Pathfinder element for Au refers to a non-ore element associated with Au, and its spatial relationship to ore systems is of value in providing vectors during exploration efforts. Arsenic, Bi, Se, Sb, Cd, Pb, Hg, Zn, Cu, and Tl have been reported widely in the literature as well-known pathfinders for Au (Dunn and HaleSeries, 2007; Dunn et al., 2005; Aitchison, 1982; Kovalevsky and Kovalevskaya, 1989; Ghorbani et al., 2020, 2022; Warren and Horsky, 1986; Reid et al., 2008; Boyle, 1979; Girling et al., 1979; Erdman and Olson, 1985; Anand et al., 2019; Lintern and Anand, 2017).

In order to understand the geochemical processes dominating the plant-substrate relationship, biogeochemical data should be treated, assessed, and analyzed statistically using the same procedures that would be applied to any set of geochemical exploration data. Quality assurance/quality control (QA/QC) is a critical component in assessing geochemical data quality before data interpretation. Abzalov (2008) introduced the average coefficient of variation, defined as CV_{avg} (%), to calculate the measurement errors in geochemical data (Abzalov, 2008). The compositional nature of geochemical data must also be considered in advance of any statistical interpretations. The compositional nature of geochemical data is expressed as part of some whole such as (part per billion) ppb, (part per million) ppm, or weight percentage (wt.%). It means that the sum of the elemental composition in any geochemical sample of interest (e.g., rock, water, or plant) is a constant. The constant sum or data closure problem is the most important limitation related to the compositional data that may lead to bias in correlations between two variables (Rollinson, 1992). This issue can be addressed using logarithms of ratios (Aitchison, 1982; Filzmoser et al., 2009; Buccianti and Grunsky, 2014).

The present study was conducted to (1) identify the biogeochemical characterization of Au and its pathfinders in plants that retain a record of the concealed mineralization setting and (2) apply robust statistical techniques to evaluate multi-element associations and reveal their interrelationships in plants with respect to the geology beneath, both of which assists in delineating zones of Au enrichment at the YCGP. Multivariate statistical analyses are well-known robust methods to evaluate the associations, correlations, and trends between variables (elements) and observations (any geochemical sample such as plants or rocks). Principal component analysis (PCA) and K-means clustering are recognized as practical geostatistical approaches in geochemical prospecting for buried mineralization (Ghorbani et al., 2022; Pratas et al., 2005; Dunn and Heberlein, 2020; Benz, 2017; Zuo, 2011). PCA is a linear dimension reduction technique for large datasets and a well-known method for identifying multi-element associations and interrelationships related to geological processes (Chen, 2015). K-means clustering is also an unsupervised data classification technique that is useful for identifying the controls on element variations (Hartigan, 1975). The combination of these methods provides an insight into the dominant geochemical processes controlling the distribution of elements in plant samples in respect of the underlying mineralization. Also, it highlights the robustness of biogeochemical and statistical techniques for follow-up studies.

2. Regional geology of the YCGP

Orogenic Au deposits (Bohlke, 1982), also known as lode-Au deposits, are an important source of Au worldwide. These deposits are dominantly associated with metamorphic terranes formed at variable depths ranging from 2 to 20 km and associated temperature-pressure regimes of 200–700 °C and <1–5 kbar, respectively (Groves et al., 1998). Generally, orogenic Au deposits occur at or above the brittle-ductile transition in compressional settings where hot Au-bearing fluids from deeper levels are the source of the mineralization (Groves et al., 1998; Goldfarb et al., 2005). Au in orogenic deposits is commonly hosted in quartz-carbonate veins and iron-enriched sulphidized wall rocks (Dubé, 2007). Silver, As, B, Bi, Mo, Sb, Te, W, and sometimes anomalous Cu, Pb, and Zn, may accompany Au in orogenic Au systems (Groves et al., 1998; Dubé and Gosselin, 2007).

The Yellowknife City Gold Project (YCGP) extends for 10–60 km to the north, east, and south of the city of Yellowknife, Northwest Territories, Canada (Armitage, 2021). This project is located in the southern part of the Slave Province (Fig. 1). The Slave Province is an Archean granite-greenstone terrane containing several ore deposits, including base metals, Ag, and Au (2.7–2.55 Ga) (Isachsen and Bowring, 1997; Ootes et al., 2011). In Particular, the YCGP encompasses a prolific orogenic Au trend situated along shear zones and faults related to the north-south trending Archean Yellowknife Greenstone Belt (YGB). The YGB is one of several greenstone belts with primarily low-grade greenschist facies metamorphism exposed in the southern portion of the Slave Province (Cousens, 2000; Cousens et al., 2006; Isachsen and Bowring, 1994; Botor et al., 2019). The YGB consists of a north-south trending metavolcanic sequence of mafic to felsic volcanics of the Kam and Banting Groups, which are unconformably overlain by a conglomerate package of the Jackson Lake Formation (Armitage, 2021; Cousens et al., 2006; Henderson and Brown, 1966; Helmstaedt and Padgham, 1986) (Fig. 1). The YGB is intruded by the Ryan Lake pluton, Defeat Plutonic Suite, Duckfish Granite, and the Anton Complex to the west (Armitage, 2021; Henderson, 1985) and is conformably overlain by the dominantly sedimentary packages of Duncan Lake Group, including the Walsh Lake and Burwash Formations to the east (Armitage, 2021; Helmstaedt and Padgham, 1986; Henderson, 1985).

The Kam Group, from base to top, includes the Chan, Crestaurum, Townsite, and Yellowknife Bay (host of the Giant and Con Mines) Formations. All conformable formations have a mafic to intermediate volcanic composition, mainly subalkaline basalts and basaltic andesites, excluding the Townsite Formation, which is composed of felsic to intermediate volcanic rocks (Armitage, 2021; Cousens et al., 2006). The Banting Group is primarily comprised of sheared calc-alkaline intermediate to felsic volcanic and volcanoclastic sedimentary rocks (Armitage, 2021). The Jackson Lake Formation is the youngest. Conglomerates of the Jackson Lake Formation have infilled the Yellowknife River Fault Zone (YRFZ), which is a faulted contact zone between the Kam Group and the Banting/Duncan Lake Groups indicating post-faulting sediments deposition (Armitage, 2021; Martel and Lin, 2006; Falck, 1990). The Walsh Lake and Burwash Formations of the Duncan Lake Group consist of a thick pile of greywacke and mudstone turbidites that conformably overlie the Banting Group (Armitage, 2021; Helmstaedt and Padgham, 1986; Henderson, 1985; Martel and Lin, 2006). Au mineralization at the YGB is similar to other mesothermal,

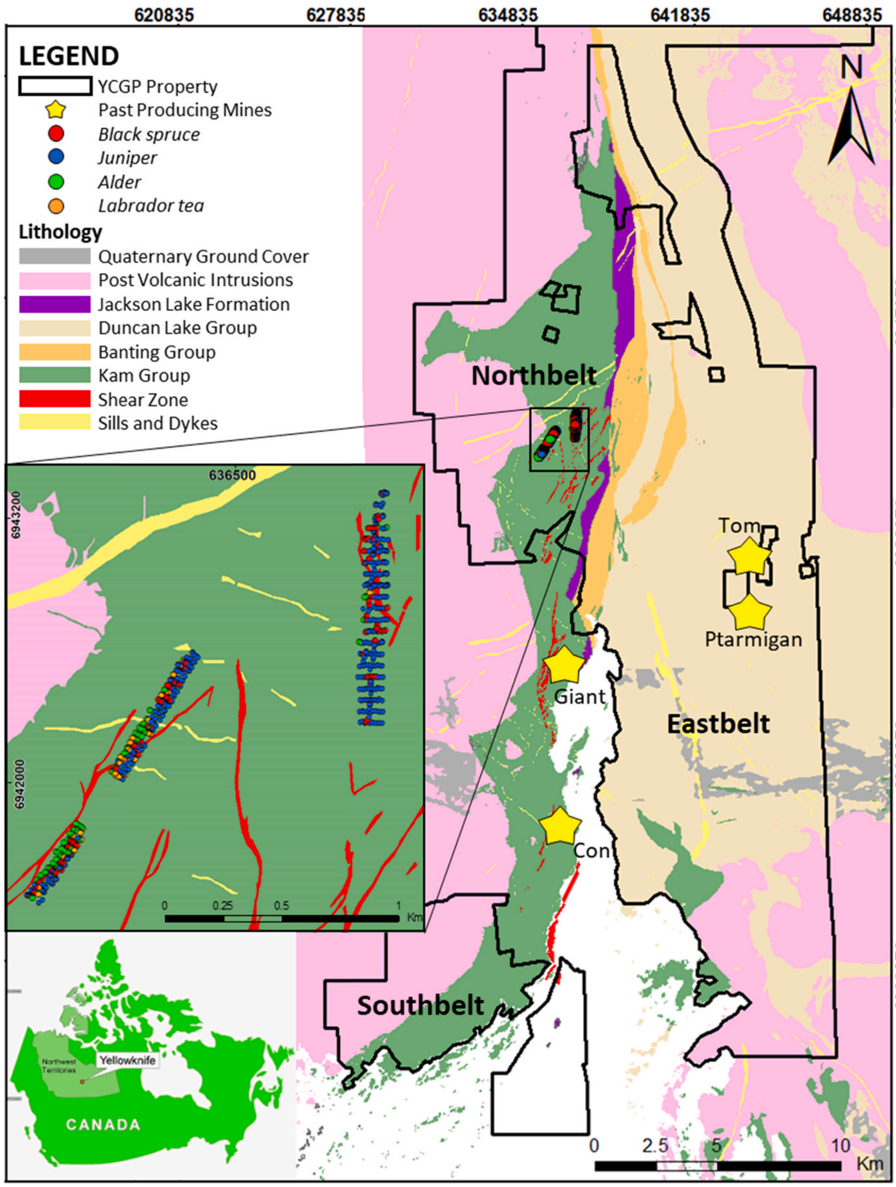

Fig. 1. Regional geology of the Yellowknife City Gold Project (YCGP) and the location of samples collected at the 2015 biogeochemical survey (Yellowknife City Gold project, 2015). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

orogenic, quartz-carbonate or Au-only deposits, which are mainly hosted in shear zones that transect mafic volcanic and metasedimentary rocks (Goldfarb et al., 2005).

The YCGP is divided into three zones, including the Northbelt, Eastbelt and Southbelt (Fig. 1). The Northbelt has the same characteristics and stratigraphy that underlies the historic Giant and Con mines, mainly hosted in quartz-carbonate-bearing shear zones that crosscut primarily mafic volcanic-dominated rocks (Groves et al., 1998; Ootes et al., 2011; Siddorn et al., 2006). This study covers the two well-known Au-bearing shear zones within the northern portions of the property, including the Crestaurum and Barney Shears. Crestaurum is a narrow discrete north-northeast trending shear zone that crosscuts the mafic volcanics of the Kam Group (Chan and Crestaurum Formations). Mineralization at the Crestaurum Shear is characterized by low to moderate pyrite, arsenopyrite, visible Au, stibnite, chalcopyrite, sphalerite, galena, and other minerals associated with multi-stage quartz--carbonate veining (Armitage, 2021). The Barney Shear is a north-south multi-kilometer wide deformation zone within the Chan Formation affected by a northeast-trending crossing structure. The Barney Shear hosts mineralization within quartz-carbonate veins with several intervals characterized by moderate to high levels of coarse sulphides, including arsenopyrite, pyrite, galena, chalcopyrite, pyrrhotite, and sphalerite (Armitage, 2021).

3. Methodology

3.1. 2015 biogeochemical survey

In 2015, a biogeochemical survey was conducted over the Crestaurum and Barney Shears, located in the north portion of the YCGP. The objective of this survey was to identify the optimum sampling medium for biogeochemical exploration at the YCGP and evaluate whether there is a plant-substrate relationship that is related to mineralization using statistical techniques. To achieve these objectives, 625 new growth needle and leaf samples, including field duplicates and standards, were taken from the dominant vegetative cover in the study area, including 320 juniper (*Juniperus)*, 189 black spruce (*Picea mariana*), 76 alder (*Alnus incana*), and 40 Labrador tea (*Ledum groenlandicum*) samples.

Samples were collected at 10 m station spacing along 25 m spaced cut lines oriented perpendicular to the Crestaurum shear; and at 10 m station spacing along 50 m spaced cut lines oriented perpendicular to the Barney shear, respectively. Needle and leaf samples were collected by snipping 25 cm lengths of branches, representing about the seventh growth season (Armitage, 2021), from chest height and around the circumference of trees. A minimum of 200 g wet sample of each plant was required for chemical analysis (Dunn and HaleSeries, 2007). The majority of samples in the study area were taken from areas of bedrock outcrop covered by thin (<30 cm) organic-rich soils with sparse tree cover to slightly more forested areas (Wolfe and Morse, 2017). Bedrock outcrop is commonly exposed to the surface over the entirety of the survey area (Palmer et al., 2021). Forested soils are thicker than the bedrock outcrop soils and are mainly covered by trees such as black spruce, consisting of a thin organic layer overlying poorly developed soils and sometimes thin tills (<2m) (Wolfe and Morse, 2017; Kerr and Knight, 2002).

Post-collection, needle and leaf samples were placed on a rack in a heated building overnight to dry out. Samples were prepared at ALS Labs in Vancouver, where the twigs and leaves were carefully separated and milled using a Wiley Mill to 100% passing 1 mm. Then 0.25-g aliquots of each ashed sample were digested in 75% aqua regia ($1HNO_3$:3HCl) using a digestion block operating. The final solution was analyzed by inductively coupled plasma-mass spectrometry (ICP-MS) using the ME-VEG41a package for ashed vegetations at ALS Global, Vancouver, British Columbia, Canada. A merged biogeochemical dataset including elements Au, As, Ag, As, B, Bi, Ca, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Mo, Ni, P, Pb, S, Sb, Se, Tl, and Zn was imported into the geochemical analysis software, ioGAS, for further data validation, including control of analytical data quality, data treatment, censoring, log-ratio transformation, multivariate data analysis and interpretations.

3.2. Data preparation and assessment

A systematic approach was applied to interpret biogeochemical data effectively and meaningfully. First, quality assurance/quality control (QA/QC) was performed to evaluate the measurement errors attributed to different stages of field sampling, lab sub-sampling, preparation, and analytical processes (Heberlein et al., Rice). The QA/QC of data was computed using the average coefficient of variation, CV_{avg} (%), to assess the effect of the measurement error and the quality of biogeochemical data (Table 1) (Ghorbani et al., 2022). In the following formula, N is the number of duplicated pairs, and a and b are the concentration of element i in the original and duplicate samples, respectively (Abzalov, 2008).

$$\mathbf{CV_{avg}}(\%) = 100 \times \sqrt{\frac{2}{\mathbf{N}}\sum_{i=1}^{\mathbf{N}}\frac{(\mathbf{a_i}-\mathbf{b_i})^2}{(\mathbf{a_i}+\mathbf{b_i})^2}}$$

Then, 1/2 detection limit was used to censor values below the lower limit of detection (LLD) with a fraction of the detection limit (Carranza, 2011). As biogeochemical data are compositional data, a log-ratio transformation was employed to address the closure problems (Aitchison, 1982; Filzmoser et al., 2009; Buccianti and Grunsky, 2014; Chen, 2015; Grunsky and de Caritat, 2020; Demšar et al., 2013). Robustication was also applied using M-estimator to mitigate the effect of outliers. This method reduces the weight of the observation with a significant error to cope with outliers (Filzmoser et al., 2009; Filzmoser and Hron, 2008; Stanimirova et al., 2007).

Table 1
The quality assurance/quality control (QA/QC) results using the average coefficient of variation (CV_{avg} (%)) indicate a marginal range for Cd, Tl, Se, Au, and S in 2015 biogeochemical data.

QA/QC of the 2015 Biogeochemical data					
Elements	CV_{avg} (%)	**Elements**	CV_{avg} (%)	**Elements**	CV_{avg} (%)
Cd_ppm	48.06	**Sb_ppm**	22.6	**Zn_ppm**	11.08
Tl_ppm	47.44	**Ni_ppm**	22.17	**P_pct**	9.94
Se_ppm	39.59	**Pb_ppm**	20.78	**As_ppm**	9.89
Au_ppm	33.52	**Ba_ppm**	19.11	**Fe_ppm**	9.62
S_pct	30.5	**Mo_ppm**	17.82	**Co_ppm**	9.27
Mn_ppm	27.97	**K_pct**	17.09	**B_pct**	9.19
Mg_pct	23.98	**Cr_ppm**	15.66	**Cu_ppm**	7.63
Bi_ppm	23.93	**Ag_ppm**	14.01	**Ca_pct**	1.43

Robust RQ-mode principal component analysis (PCA) and K-means clustering were applied to identify the elemental association in the needle and leave samples and their inter-relationship with different vegetation types over the Crestaurum and Barney Shears. PCA is a linear dimension reduction technique for large datasets that transforms original data into a new lower-dimensional representation using a linear projection with a minimum loss of information (Demšar et al., 2013; Singh et al., 2017). RQ-mode PCA was introduced by Zou et al. (1983) to geochemical data, signifying the interrelationship between elements (R-mode) and the combinations of samples (Q-mode) that explain variation among variables simultaneously (Chen, 2015; Neff, 1994; Zhou et al., 1983). K-means clustering analysis is a simple and powerful unsupervised algorithm that has been widely used in data mining to discover the hidden spatial pattern and structure of unlabelled samples (Zuo, 2017; Xu et al., 2021). In this method, samples are classified into different clusters, exhibiting all samples in the same cluster with relatively similar characterization.

3.2.1. Data quality

This study expresses the average coefficient of variation (CV_{avg}(%)) as the total measurement error. A high total measurement error can obscure meaningful geochemical patterns and consequently undermine the biogeochemical technique to identify underlying mineralization (Abzalov, 2008). Coefficient of variation values below 20% indicate good data precision; between 20% and 30% acceptable precision; between 30% and 50% marginal precision; and over 50%, poor precision. In 2015 biogeochemical data, most elements are within the good and acceptable quality range (Table 1). Cadmium, Tl, Se, Au, and S are found within the marginal range (Table 1). These elements must be used with caution as their distribution patterns are likely to be influenced by noise caused by poor reproducibility (Dunn and Heberlein, 2020; Heberlein et al., Rice).

4. Results and discussion

4.1. Summary of statistics

Table 2 provides the statistical summary applied to 625 black spruce, alder, Labrador tea, and juniper samples. In this study, Au, its associated pathfinder elements, including Ag, As, Bi, Sb, Se, Tl, Pb, and Tl, as well as a suite of other elements, including Cr, Fe, Ni, Mn, Zn, Co, Ca, Cu, Cd, P, K, S, Mg, B, and Mo, were selected to identify potential biogeochemical responses in plants to subsurface variations in geology. The results of the univariate statistical analysis show that the lowest (0.4 ppb) and highest (147 ppb) Au values were identified in juniper needles. The average Au concentration for all vegetation types is ~9 ppb. Black spruce samples with average Au values of 18.6 ppb demonstrate an enhanced ability to accumulate Au and reflect the plant-substrate relationship compared to Labrador tea, alder, and juniper with lower average Au contents of 12.86 ppb, 5.3 ppb, and 4 ppb, respectively. In addition to Au, the average concentration of most elements, including Ag, As, Bi, Sb, Se, Tl, Pb, Tl, Cr, Fe, Ni, Mn, Co, Cu and Cd, are elevated in black spruce in comparison to other vegetation types, with the exception of Mo, K, Zn, S, and Mg, which are accumulated more in alder samples. Boron and Ca have higher values in Labrador tea and juniper compared to other vegetations.

In order to have a better understanding of the depletion and enrichment of each element, the average concentration of each element in all sample types was compared with a background level. In this study,

Table 2
Descriptive statistics of minimum, maximum, mean and median values of elements concentration for 625 vegetation samples collected at the YCGP. The number of collected samples for each plant species was shown in a bracket. Also, the average concentration of elements in different vegetation types, including alder, black spruce, juniper, and Labrador tea, are provided. The Median or 50 percentile is considered the background level.

Sample Type	All Vegetation Samples				Alder [76]	Black Spruce [179]	Juniper [320]	Labrador Tea [50]
Elements	Min	Max	Mean	Background	Mean			
Au_ppb	0.4	147	9.1	4.4	5.3	18.61	3.92	12.86
Ag_ppm	0.02	6.08	0.64	0.05	0.06	1.98	0.04	0.13
Bi_ppm	0	0.282	0.03	0.02	0.01	0.07	0.02	0.04
As_ppm	1.94	185	19.15	10.1	13.85	40.22	6.72	29.02
B_ppm	76	1620	323	219	589	447	149	626
Ca_pct	13.55	26	24.3	26	21.78	23.31	25.97	21.15
Cd_ppm	0.02	8.27	0.82	0.62	0.6	1.09	0.77	0.41
Co_ppm	0.29	64.3	4.76	2.2	5.09	10.08	1.7	3
Cr_ppm	0.52	24.8	3.88	2	1.98	8.23	1.67	4.62
Cu_ppm	12.45	338	93.9	52.7	103	191	28	144
Fe_ppm	459	15400	2466	1300	1406	5263	1043	2650
K_pct	1.21	11	7.42	7.66	10.83	9.41	5.02	10.78
Mg_pct	0.2	7.69	2.33	1.99	5.44	2.71	1.17	3.89
Mn_ppm	261	27000	6185	4040	7977	10524	2562	11271
Mo_ppm	0.11	36.7	1.57	0.81	4.6	0.8	1.41	0.83
Ni_ppm	1.01	196.5	25.2	14	20	33.57	21.1	29.63
P_pct	0.29	1.001	0.94	1	1	1	0.88	1
Pb_ppm	1.19	170	7.56	3.91	3.51	15.81	3.43	9.22
S_pct	0.04	3.31	0.76	0.61	1.54	0.99	0.36	1.39
Sb_ppm	0.3	44.6	2.87	1.35	1.98	5.95	1.04	4.65
Se_ppm	0	1.185	0.16	0.1	0.09	0.32	0.07	0.18
Tl_ppm	0	6.73	0.16	0.03	0.04	0.4	0.03	0.18
Zn_ppm	87	8790	1344	456	3608	2423	432	843

no suitable background samples were collected as the entire area of the survey is underlain by a similar geology. Therefore, the median or 50th percentile was used as background (Dunn and HaleSeries, 2007). The average concentrations of Au in plant samples (9.1 ppb) collected at the YCGP are almost two times higher than the background level (4.4 ppb). Particularly, black spruce and Labrador tea are highly enriched in Au. Mean Au content in black spruce (18.61 ppb) is more than four times and in Labrador tea (12.86 ppb) is more than two times higher than the background level (4.4 ppb). Alder is slightly enriched in Au (5.3 ppb), while juniper is depleted (3.9 ppb) compared to the background value (4.4 ppb). Comparing the Ag and Bi mean values to the background levels shows no difference to slight enrichments in alder and juniper. Silver is enriched up to 40 times in black spruce. Silver and Bi are accumulated more than two times in Labrador tea samples. Arsenic, Pb, Sb, Se, Cr, Fe, Mn, Cu, Cd, Zn values are enriched in all sample types compared to the background level. Thallium values in black spruce, juniper, and Labrador tea are three times more than the background level. Cobalt is accumulated at twice the background level in black spruce and alder. Juniper and alder have accumulated Ni two times higher than the background level. No significant difference was identified for Ca in juniper between the mean content and background value, while Ca is depleted in the other vegetation types. Molybdenum shows more than five times enrichment in alder, while black spruce and labrador tea contain the same mean Mo content as the background value.

Au concentration in vegetation types collected over the Crestaurum and Barney Shears are compared in Figs. 2 and 3. Needle samples collected over the Crestaurum shear contain higher Au values than those collected over the Barney Shear in all vegetation types except alder, which contains similar Au values in both localities (Fig. 2). High Au values (89–147 ppb) were identified in all vegetation types located in close proximity to each other over the south of the Crestaurum Shear. Black spruce indicates a significant Au enrichment in both shears compared to other vegetation species (Fig. 3).

4.2. Robust RQ-mode principal component analysis (PCA)

The biogeochemical study at the YCGP was carried out to document element concentrations and spatial variations in plant samples. A robust

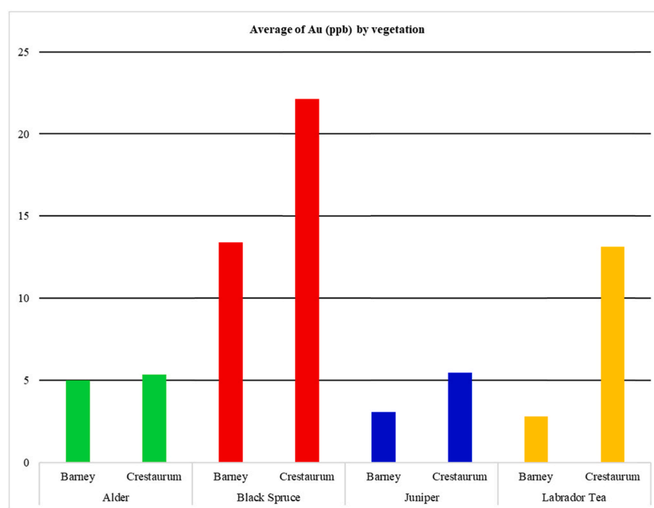

Fig. 2. The comparison of the average concentration of Au (ppb) by different vegetation collected over the Barney and Crestaurum Shears.

RQ-mode principal component analysis (PCA) inspection of this data was applied to identify the geochemical processes dominating the element distribution in vegetation samples in the survey area (Fig. 4). PC1, PC2, and PC3 with eigenvalues greater than one account for ~83% of the total variance covering the most important data information. PC1 and PC2 are the main geochemical factors controlling the distribution of elements in vegetation samples collected in the 2015 biogeochemical survey.

PC1 (~65% of total variance) consists of a set of elements that primarily represent the bedrock composition, including Au, Ag, As, Se, Bi, Pb, Cu, Co, Fe, Zn, Cd, Tl, and Cr. The association of Fe, Co, Cr, Ti, and Ni with Au, As, and Sb in one component of the PCA demonstrates a strong plant-substrate relationship in the Au mineralized area (Ghorbani et al., 2022; Dunn et al., 1995). Therefore, the biogeochemical signature controlling PC1 is termed the mineralization/geological factor. The

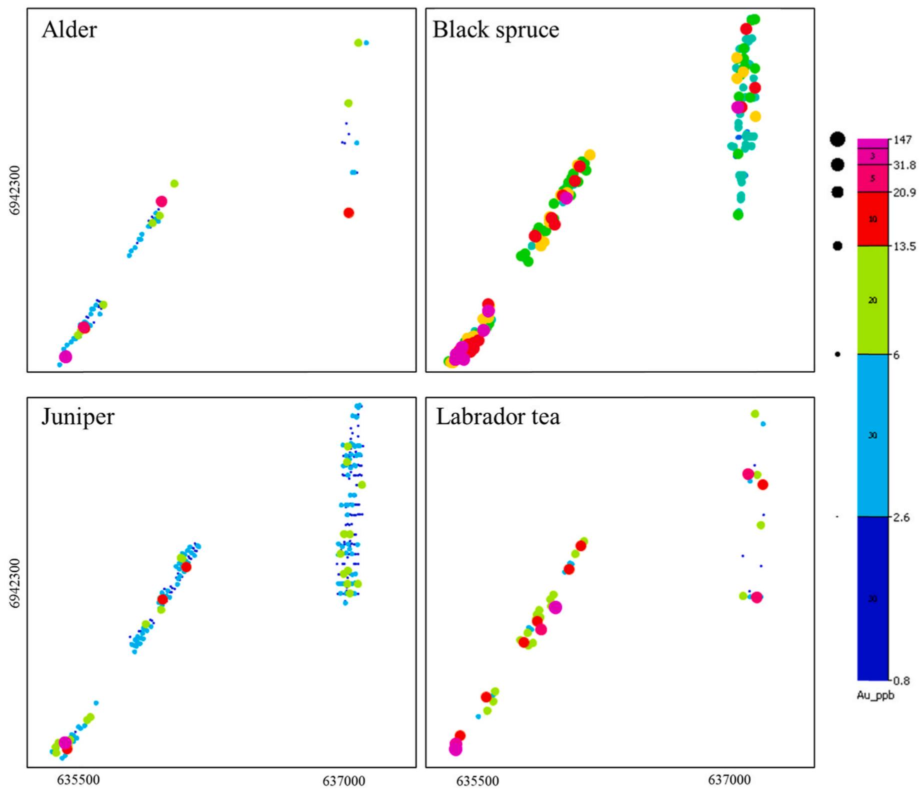

Fig. 3. The concentration of Au in alder, juniper, black spruce, and Labrador tea samples collected over the Crestaurum and Barney Shear. Anomalous Au values up to 147 ppb were identified in all vegetation types located over the southern portion of the Crestaurum shear. Black spruce needles show a significant Au enrichment compared to other vegetations.

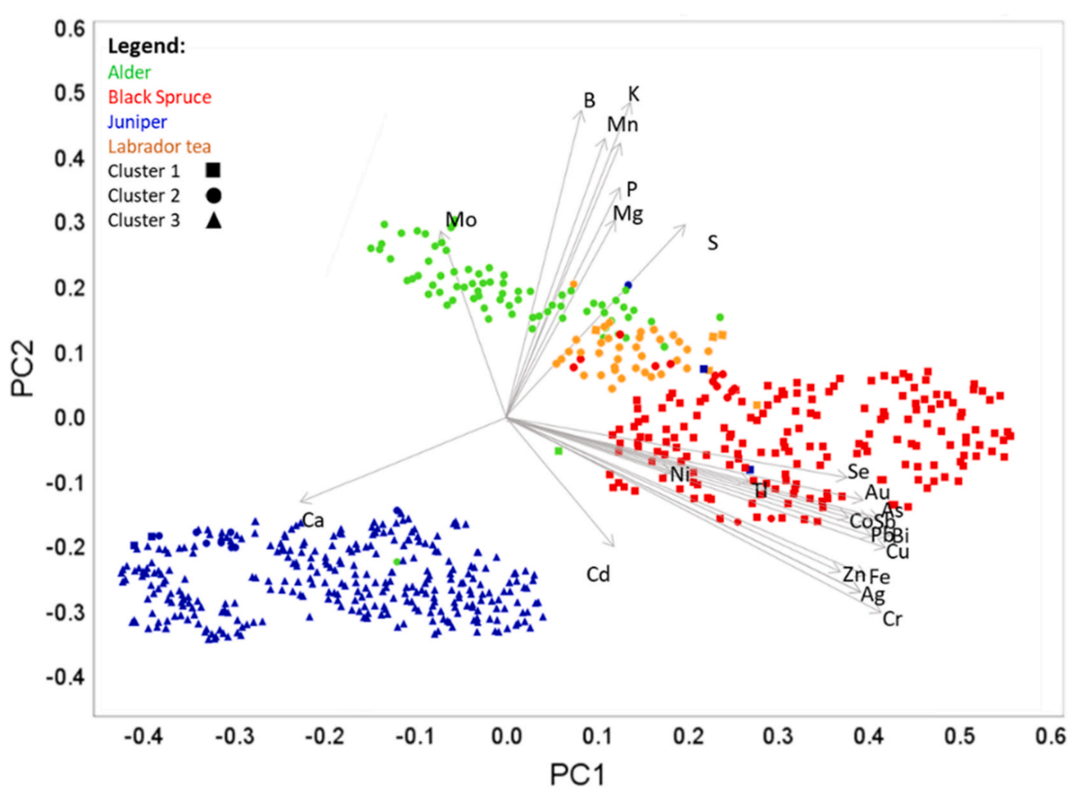

Fig. 4. Graphical representation of the robust RQ-mode principal component analysis (PCA) and K-means clustering applied to the 2015 biogeochemical sample. The PCA results indicate that the mineralization/geological factor (PC1) and physiological factor (PC2) are the contributing factors controlling the distribution of elements in plant samples collected at YCGP. PC3 includes only Mo that is positively loaded in PC2. Based on the K-means clustering, samples are grouped into three clusters shown by square, circle, and triangle shapes, demonstrating the plant-substrate relationship with respect to the lithology/mineralogy beneath.

loadings of PC1 are vectored close to black spruce samples shown in red (Fig. 4). PC2 (~13% of total variance) is dominated by elements that are considered essential elements for plant metabolism. This component has a strong negative loading for Ca and strong positive loadings for B, Mn, Mg, K, P, and Mo. The plant physiological factor reflects the biogeochemical response (PC2) that controls the occurrence and distribution of essential elements in plant samples. The loadings of PC2 are positively vectored toward alder (green) and Labrador tea (orange) samples. Alder and Labrador tea have broader leaves than black spruce and juniper needles. Therefore, they indicate more physiological features related to the presence of essential elements such as Mg, K, and P. Calcium is negatively loaded to PC2, likely because of its structural function in plant metabolism and tendency to accumulate in bark than needles and leaves (Dunn and HaleSeries, 2007). PC3 is responsible for ~5% of the total variance, including only Mo, which is also positively loaded in PC2. PC3 loading is proximal to the alder samples (green), indicating a significant association between Mo and alder samples.

4.3. K-means clustering analysis

K-means clustering analysis was applied to the 625 needle samples collected in the study area. According to Fig. 4, needle samples are grouped into three clusters based on their vegetation type and affinity to the specific suites of elements. Clusters are presented in different shapes: cluster 1 = squares, cluster 2 = circles, and cluster 3 = triangles (Fig. 4). Cluster 1 is dominated by the majority of the black spruce samples (red squares), located proximal to the set of elements that compose PC1 (mineralization/geological factor). Therefore, black spruce can be considered the preferred sample medium to reflect a plant-substrate relationship by accumulating higher concentrations of metals, Au, and associated pathfinder elements. Cluster 2 includes samples from Labrador tea (orange circles) and alder (green circles). Labrador tea samples (orange circles) are clustered between PC1 and PC2, slightly more toward PC2, signifying the influence of both the mineralization/geological and physiological factors on the biogeochemistry of Labrador tea samples. Labrador tea is the second most preferred plant species for the biogeochemical survey at the YCGP. Alder samples (green circles) are located proximal to PC2 and PC3, where sets of elements responsible for the physiological factor are located. The association of alder and PC3, where the Mo vector is loaded, is related to the tendency of alders to accumulate Mo (Dunn and HaleSeries, 2007). The requirement of Mo in alder and the presence of anomalous values of Mo in alder have been reported in studies by Becking (1961), Dunn (1984), and Dunn et al. (1990) (Becking, 1961; Dunn, 1985; Dunn et al., 1990). Juniper samples (blue triangles) are predominantly associated with cluster 3, which is loaded negatively to PC1. These samples contain low values of Au and are considered the least suitable media for biogeochemical prospecting of Au compared to other vegetations in the study area. Also, the presence of the Ca vector near the juniper samples can be attributed to the higher concentration of Ca in juniper in comparison to the other vegetation types.

The PCA and K-means clustering indicate that mineralization/geological and physiological factors are the dominant biogeochemical signatures controlling the distribution of elements in plants at the YCGP. Also, black spruce was identified to be the best sample medium for biogeochemical reconnaissance prospecting for Au in the study area. In black spruce, Au is associated significantly with As and Sb ($R^2 > 0.8$) and somewhat less with Bi and Pb ($R^2 > 0.63$). These associations demonstrate the robustness of the plant-substrate relationship since mineralization in the region consists of low to moderate abundances of sulphides such as pyrite, arsenopyrite, stibnite, and galena at the Crestaurum and Barney shears (Armitage, 2021). Also, a high Au–Mo association (R^2: 0.56) was identified in black spruce. This relationship may be related to the presence of late felsic intrusions with strong Au ± Mo mineralization (Armitage, 2021). It has been claimed that Mo is a successful biogeochemical indicator of mineralization because plant tissues do not establish significant barriers to Mo uptake (Dunn and HaleSeries, 2007).

The PCA and K-means clustering analysis indicate that Ag, As, Bi, Sb, Se, and Tl are the best pathfinders of Au in the current study. Also, black spruce > Labrador tea were identified as the best vegetation medium for Au prospecting in the study area. All pathfinder elements are significantly enriched in the southern portion of the Crestaurum Shear, where high Au concentrations are identified (Fig. 3). The association of Au with As and Sb is related to the sulphide mineralization hosted within the mafic volcanics. Many studies reported the presence of As-bearing and Sb-bearing minerals such as pyrite, arsenopyrite, and stibnite within the Crestaurum and Barney Shears (Armitage, 2021; Botor, 2018). Also, positive associations between Au and Ag can possibly be related to the presence of porphyry-style mineralization with high Au and Ag content intersecting the Barney shear (Armitage, 2021). Thallium is a useful pathfinder of Au in biogeochemical exploration. A strong association between Au, Tl, As, and Sb is observed in polymetallic deposits (Dunn and HaleSeries, 2007). Also, a significant association between Au and Tl is identified in spruce samples located at the Highland Valley mine in Southern British Columbia (Dunn and HaleSeries, 2007; Warren and Horsky, 1986). Significant correlations between Au and Ag, Bi, Se and, to a lesser extent, As and Tl were observed north of the Barney Shear. Samples collected from the north of the Barney shear are located precisely over the mineralized shear (Fig. 1), where Au and its pathfinder elements are similarly enriched. These associations illustrate a crystal-clear example of the plant-substrate relationship and the robustness of the biogeochemical technique in delineating zones of Au enrichment. The results of this study can be applied to a wider survey across the YCGP to look for extensions of the advanced targets and possibly elucidate new targets at low cost and with significant reliability.

5. Conclusion

The outcomes of the biogeochemical prospecting for Au at the Crestaurum and Barney Shears using biogeochemical and robust statistical techniques were found to be positive and reliable. Statistical analysis indicates that needle samples can accumulate a significant amount of Au, up to 147 ppb. Black spruce > Labrador tea are confirmed as the optimum sample media among all plant species collected for this study. The comparison between Crestaurum and Barney Shear indicated that needle samples collected over the Crestaurum Shear are more enriched in Au in all plant samples except for alder, with the same Au values in both shears. Principal component analysis and K-means clustering analysis are used to discover the plant-substrate relationship and elemental association. Three factors controlling the geochemical distribution in plant samples are identified using PCA. PC1 includes a strong association between Au, Ag, As, Se, Bi, Pb, Cu, Co, Fe, Zn, Cd, Tl, and Cr, representing the mineralization/geological factor. Well-known Au pathfinder elements including Ag, As, Se, Sb, Bi, and Tl are attributed to PC1, indicating the mineralization association. PC2 includes essential elements for plant metabolism, including B, Ca, Mn, Mg, K, P, and Mo. Ca is negatively loaded into PC2 due to its structural function. PC2 represents physiological factors. PC3 consists of only Mo, significantly associated with alder samples. K-means clustering classifies samples into three clusters based on the vegetation type and their tendency to accumulate specific sets of elements. Black spruce samples are grouped into cluster 1, where PC1 or the mineralization/geological factor is vectored. This result verifies the ability of black spruce to provide insight into the geochemistry and composition of the underlying lithologies. Labrador tea and alder samples are classified into cluster 2, while juniper samples are grouped in Cluster 3. Au is strongly accompanied by its pathfinder elements, especially in the south of the Crestaurum Shear. The association of Au with its pathfinder elements indicates the reliability of the results and a strong plant-substrate relationship. The positive result of this survey verifies the robustness of the combined use of biogeochemical and statistical analysis, which can be used in a broader survey across the YCGP to find extensions to the advanced and early-stage Au targets.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

We thank Aaron Doan, Duncan Studd, and the entire Yellowknife City Gold Project team for conducting the survey, providing data, and for valuable discussions. Funding for this project was provided by Western University and the Natural Sciences and Engineering Research Council (NSERC).

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