

Responses to referee comments on "**Application of lithogeochemical and pyrite trace element data for the determination of vectors to ore in the Raja Au-Co prospect, northern Finland**" by Sara Raič et al., Solid Earth Discuss., 2021

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Referee # 1 | Solid Earth Discussion

Response to referee comment (RC) on "Application of lithogeochemical and pyrite trace element data for the determination of vectors to ore in the Raja Au-Co prospect, northern Finland" by Sara Raič et al., Solid Earth Discuss., <https://doi.org/10.5194/se-2021-119-RC1>, 2021

General Comments

RC: The manuscript by Raic et al. on the vectors to ore in the Raja project, Finland, is an excellent study on the use of mineralogical and geochemical vectors in exploration – especially since it deals with orogenic gold systems, which have not been so extensively studied as other mineral systems. However, the paper is rather long and sometimes reiterative so I would suggest the authors to try to synthesize it.

Response: Following these suggestions, the reiterative sections of the manuscript have been reduced or merged (see sections such as PCA and implications to mineralization in the discussion, as well as the conclusion section).

Specific Comments

All comments and suggestions made by the reviewer regarding typo, spelling and expression have been rephrased, accepted, and changed in the manuscript. These comprise the comments in the following lines: 11, 41, 46, 47, 49, 56, 60, 170, 785 and 851.

Comments made by the reviewer that would require a detailed response, clarification, or discussion, are addressed below.

Line 12:

As a new exploration tool for such terrains, we test the vectoring capacities of trace element and sulfur isotope characteristics of pyrite, combined with quantitative tools for whole-rock geochemical datasets.

RC: Which tools?

Response: Sentence rephrased to: [...] characteristics of pyrite, combined with quantitative statistical methods of whole-rock geochemical datasets.

Lines 16–17:

The major lithologies at Rajapalot comprise variously altered and deformed calcsilicate rocks that alternate with albitized metasedimentary units, mafic volcanic rocks, mica schist and quartzite.

RC: Which alteration? Albitization has affected all these rocks?

Response: The major lithologies at Rajapalot comprise amphibolite facies metamorphosed and polydeformed calcsilicate rocks that alternate with albitic units, mafic volcanic rocks, mica schist and quartzite.

Line 40:

More than five prospects make up the broader Rajapalot Au-Co project, and this study focuses on a single prospect, Raja.

RC: “More than five” looks a bit strange.

Response: Rephrased to: Seven prospects make up the broader Rajapalot Au-Co project, and this study focuses on a single prospect, Raja.

Lines 59–60:

[...] suitable for this type of analysis, considering its capability of hosting a variety of elements of economic interest (Au, As, Ni, Co, Ag, Te, W, Sb, Mo and Se) and its compositional adaption to changes in the physicochemical parameters of a hydrothermal system (Liu et al., 2018; Large et al., 2009; Ulrich et al., 2011; Deditius et al., 2014; Keith et al., 2018; Liu et al., 2018).

RC: Not all these elements are of economic interest. Delete “of a hydrothermal system”.

Response: Rephrased to: [...] suitable for this type of analysis, considering its capability of hosting a variety of elements of economic interest (Au, Ag, Bi, Co, Cu, Mo, Ni, Sb, Se, and Te) and its compositional adaption to changes in the physicochemical fluid parameters (Liu et al., 2018; Large et al., 2009; Ulrich et al., 2011; Deditius et al., 2014; Keith et al., 2018; Liu et al., 2018).

Line 66:

Several authors have shown that a combination of aforementioned analytical techniques also highly support refining of genetic models of ore deposits.

RC: References

Response: The following references are added in text, as well as in the reference section: Several authors have shown that a combination of aforementioned analytical techniques also highly support refining of genetic models of ore deposits (Barker et al., 2009; Hodkiewicz et al., 2009; Ulrich et al., 2011; Liu et al., 2018; Mukherjee et al., 2019; Voute et al., 2019; Meng et al., 2020; Vasilopoulos et al., 2021).

Meng, X., Li, X., Chu, F., Zhu, J., Lei, J., Li, Z., Wang, H., Chen, L., Zhu, Z.: Trace element and sulfur isotope compositions for pyrite across the mineralization zones of a sulfide chimney from the East Pacific Rise (1-2°S), *Ore Geol. Rev.*, 116, 103209, <https://doi.org/10.1016/j.oregeorev.2019.103209>, 2020.

Mukherjee, I., Large, R.R., Bull, S., Gregory, D.G., Stepanov, A.S., Ávila, J., Ireland, T.R. and Corkrey, R.: Pyrite trace-element and sulfur isotope geochemistry of paleo-mesoproterozoic McArthur Basin: Proxy for oxidative weathering, *Am. Mineral.*, 104, 1256–1272, <https://doi.org/10.2138/am-2019-6873>, 2019.

Vasilopoulos, M., Molnár, F., O'Brien, H., Lahaye, Y., Lefèbvre, M., Richard, A., André-Mayer, A.S., Ranta, J.-P., Talikka, M., Geochemical signatures of mineralizing events in the Juomasuo Au-Co deposit, Kuusamo belt, northeastern Finland., *Miner. Deposita*, 56, 1195–1222, <https://doi.org/10.1007/s00126-020-01039-8>, 2021.

Lines 86–88:

[...] and summarized the volcano-sedimentary successions and intrusions with the following generalized lithostratigraphy (from bottom to top): (i) basaltic mafic volcanics and minor conglomerates (ii) clastic sedimentary rocks, (iii) subaerial mafic volcanics, komatiites and carbonate rocks; (iv) greywackes, carbonaceous material and sulfur-rich pelitic rocks and (v) phyllites and greywackes.

RC: Basaltic mafic is redundant. Material is not a really good geological term. Carbonaceous-bearing rocks?

Response: Rephrased to: [...] and summarized the volcano-sedimentary successions and intrusions with the following generalized lithostratigraphy (from bottom to top): (i) mafic volcanics and minor conglomerates (ii) clastic sedimentary rocks, (iii) subaerial mafic volcanics, komatiites and carbonate rocks; (iv) greywackes, carbonaceous-bearing rocks and [...].

Line 133:

Inferred mineral resources at Raja only, are estimated at 2.97 million tonnes @ 2.9 g/t gold with 383 ppm cobalt, which form 46 % of the gold-cobalt resources at Rajapalot (Mawson Gold Ltd., 2021).

RC: Perhaps I would say “averaging 383 ppm of Co”

Response: Rephrased to: Inferred mineral resource-estimate at Raja only are at 2.97 million tonnes @ 2.9 g/t gold and covers average 383 ppm cobalt, which form 46 % of the gold-cobalt resources at Rajapalot (Mawson Gold Ltd., 2021).

Lines 142–145:

[...] (i) Sequence 1 comprises siliciclastic, albitized and carbonatized, largely oxidized metasedimentary rocks from a continental margin setting; (ii) Sequence 2 represents a metamorphosed sedimentary sequence formed under reduced conditions consisting of pelitic turbidites, sandstones, carbonates and sulfidic carbonaceous material-bearing rocks. Mafic rocks (lava flows, dykes and volcanoclastic sediments) are common within both sequences. In the mineralized zones, domains of retrograde alteration to chlorite or epidote occur.

RC: Could you explain these oxidized sediments? I think this deserves a more detailed explanation.

Response: The oxidized part of this package most likely formed during the later stages of the Great Oxygenation Event. The oxidized state of the rocks could also have been influenced by oxidizing saline fluids, derived from the interaction with hematitic rocks that formed during the Great Oxygenation Event. Also, within the stratigraphy of the Peräpohja Belt (Kivalo Group) are inferred evaporites of the Petäjaskoski Formation (Kyläkoski et al., 2012; see reference in ms).

Lines 158–163:

The sampled drill cores are from two profiles (profile 1 and profile 2; Fig. 2b–c) from Sequence 2 of the Raja prospect. Major lithologies are mafic metavolcanic rocks (e.g. pillow basalt to amphibolite), albite-calcisilicate rocks, biotite-calcisilicate rocks, albitite, mica schist and muscovite-bearing quartzite (Fig. 2c). The zones of high-grade Au-Co mineralization are characterized by sulfide disseminations adjacent to linear, or sub-linear near-vertical structures (Cook and Hudson, 2018; Cook et al., 2020). The Raja Au-Co resource extends 240 meters parallel to strike, 950 meters down plunge reaching a vertical depth of 560 meters (Mawson Gold Ltd., 2020).

RC: I think that there is some contradiction here.

Response: The Raja Au-Co resource is formed owing to the intersection of largely vertical structures intersecting with a stratabound reactive host package resulting in a flattened prolate shape to mineralization with the long axis extending 950 meters down plunge and the in-strata intermediate axis of the ellipse up to 240 meters parallel to strike.

Line 310:

This rock commonly contains quartz and albite porphyroclasts that are rimmed by mica-rich seams and bands, which often exhibit replacement of biotite by chlorite in sheared zones.

RC: mica-rich seams and bands... don't understand

Response: The pyrite-porphyroclasts are enveloped by micas (muscovite and biotite). These sheetsilicates either form foliated seams or fine laminations. Rephrased the sentence to: This rock commonly contains quartz and albite porphyroclasts with enveloping muscovite and biotite, which often exhibit replacement of biotite by chlorite in sheared zones.

Lines 328-338:

Section 4.2.1 Alteration types

RC: This is a repetition. The term sericitic alteration is too vague. Sericite is a fine grained white mica that you don't know what is. Why not, phyllic?

Response: The metamorphic white mica is a muscovite, while the alteration white mica is more phengitic. At Rajapalot, the hanging wall shows good evidence for the equivalent to the classic porphyry staged overprinting the amphibolite-facies rocks.

The section 4.2.1 was incorporated into 4.1 and section 4.2 was changed from "Alteration types" to "Ore textures and Mineralogy". The following paragraph is added to 4.1.: The most common alteration types include albitization (throughout the pre-post-syn-orogenic stages), retrograde alteration assemblages of chloritized biotite, chlorite and Fe-Mg amphiboles (syn-orogenic), sulfidation-induced formation of massive pyrrhotite and subordinate pyrite and chalcopyrite (syn-to post-orogenic), as well as sections where plagioclase is affected by the sericitic alteration characterize the latest alteration event (post-orogenic; see Table 1).

Line 418:

To achieve stable results, it is recommended to enter the principal component analysis with variables selected based on geochemical reasoning, and not with the full set of analyzed elements.

RC: By whom is recommended?

Response: reference added: [...], and not with the full set of analyzed elements (see Le Maitre, 1982; Reimann et al., 2008).

LeMaitre ref. added to Reference section:

Le Maitre, R.W.: Numerical Petrology. Elsevier Scientific Publishing Company, Amsterdam, The Netherlands, 281 pp. ISBN: 978-0-444-42098-5, 1982.

Line 506:

You do not discuss at all, the mechanisms of ore precipitation despite you quote it several times along the paper.

RC: Statements on ore-formation models are included in the rephrased section 5.4 Signatures of sulfur isotopes in pyrite and ore-forming processes.

Line 623:

Pyrite A, which has the highest Co/Ni ratios, displays the heaviest $\delta^{34}\text{S}$ signatures $\delta^{34}\text{S}$ (1.3 ‰ to +5.9 ‰), contrasting with pyrite B (-1.8 ‰ to +7.3 ‰) and pyrite C (-1.2 ‰ to +7.4 ‰; Fig. 13b; Table 3).

RC: Perhaps highest instead heaviest...

Response: Rephrased: Pyrite A, which has the highest Co/Ni ratios, displays positive only $\delta^{34}\text{S}$ signatures (+1.3 ‰ to +5.9 ‰), contrasting with pyrite B (-1.8 ‰ to +7.3 ‰) and pyrite C (-1.2 ‰ to +7.4 ‰; Fig. 13b; Table 3).

Line 643.

Regional pre-orogenic albitization, caused by diagenetic and basinal brines, prepared the country rocks for a possible pre-enrichment of metals and for later Au-mineralizing processes.

RC: I am a bit surprised that you state that albitization is caused by sedimentary brines during diagenesis. I guess that many people would disagree with this. Could you expand this discussion a bit, perhaps in other place of the paper? Or at least add some references.

Response: In Eilu et al., (2003): Several deposits in the CLGB, the Kuusamo belt and the Peräpohja belt are associated with albitization along major structural features (Pankka and Vanhanen, 1992). In these greenstone belts the early spilitization and diagenetic albitization are then overprinted by a more extensive albite alteration, which is interpreted to be the result of a of greenstone belt-scale hydrothermal circulation of magmatic and/or basinal brines prior to or during the peak deformation (Pankka and Vanhanen, 1992; Vanhanen, 2001).

At Rajapalot we can see the preservation of fine laminar structure in some rocks that are not fabric destructive. The driving force for country rock pre-preparation are basinal saline-fluids, along with the probably localized heating around large mafic lavas and sills.

Line 643 is rephrased to: Regional pre-orogenic or early-metamorphic albitization, which predates the peak deformation and gold mineralization, was caused by greenstone belt-scale hydrothermal circulation of magmatic and/or basinal brines (see Eilu et al., 2003), which prepared parts of the country rocks for a possible pre-enrichment of metals and for later Au-mineralizing processes (Pankka and Vanhanen, 1992; Vanhanen, 2001).

Reference added to references section:

Pankka, H.S. and Vanhanen, E.J.: Early Proterozoic Au-Co-U mineralization in the Kuusamo district, northeastern Finland, *Precambrian Res.*, 58, 387–400, [https://doi.org/10.1016/0301-9268\(92\)90126-9](https://doi.org/10.1016/0301-9268(92)90126-9), 1992.

Lines 659–661:

These rocks underwent several stages of alteration, such as albitization, chloritization and sericitization and have different rheological properties (brittle and ductile), indicating that rheology has an impact on the metallurgical processes (Farajawicz & Cook, 2021).

RC: Rheology and metallurgical processes... can you explain this a bit more?

Response: Rephrased to: These rocks underwent several stages of alteration, such as albitization, chloritization and sericitization producing widely varying rheological hosts to mineralization (Farajawicz & Cook, 2021). These contrasting host rheologies will play a significant part in the design of suitable crushing circuits capable of releasing gold and cobalt into chemical or flotation treatment cells

Lines 740–745:

The most abundant trace elements in pyrite are Co (up to 3.6 %), Cu (up to 1.1 %), As (up to 0.77%), Ni (up to 0.38 %), Se (up to 0.15 %), Mo (up to 713 ppm), Ag (up to 659 ppm), W (up to 242 ppm), Tl (up to 123 ppm), Te (up to 110 ppm), Bi (up to 94 ppm) and Au (up to 4.38 ppm). Considering that divalent cations, such as Cu²⁺ (0.54 Å), Co²⁺ (0.65 Å) and Ni²⁺ (0.69 Å) are involved in isovalent substitution with Fe²⁺ (0.61 Å), their anomalous concentrations within the pyrite structure are less surprising (George et al., 2018). Cations involved in coupled heterovalent substitution of 2Fe²⁺ are Cu⁺, As³⁺, Mo⁴⁺, Ag⁺, W⁶⁺ and Au⁺, Au³⁺, Tl⁺, Bi³⁺ (Chouinard et al., 2005 in George et al., 2018; Deditius et al., 2009). Elements involved in the anion substitution of S are As, Sb, Se and Te, as they can play the role of cations and anions (George et al., 2018).

RC: this repeats what was said above. Try to clear it.

Response: Following the suggestions of the reviewer this section was shortened to avoid repetition.

Line 767:

While the latter PCs are capable to describe substitution processes within the pyrite structure, which is a valuable asset for geometallurgical purposes, the PC3 points out that some grains could show an association between Au and As, when considering their negative loadings (see Fig. 12b).

RC: Again, I don't understand why geometallurgy is quoted here.

Response: The phrase "which is a valuable asset for geometallurgical purposes" is removed from the sentence, and rephrased to: "The PC3, however, points out that some grains could show an association between Au and As, when considering their negative loadings (see Fig. 12b)".

Line 780:

According to Dmitrijeva et al. (2020), the significant W and Mo signals in pyrite represent newly precipitated sub-micron-scale scheelite and molybdenite phases that are the products of a coupled dissolution-reprecipitation reaction of a parent W-Mo-bearing hematite

RC: I think that here there is a misunderstanding of the paper by Dmitrijeva et al.

(2020) paper. I don't understand what is the link here with hematite – not present in Raja to my knowledge.

Response: When considering the bituminous and graphitic rocks at Rajapalot, the molybdenum is clearly tied to organic material (e.g. flakes of Mo in peculiar bitumen shapes; chemistries also point to a reduced black shale precipitation of Mo). Regarding the W, it is possible that the scheelite and wolframite are sourced from the granitoids (along with boron). Another W-source could be from W-evaporites as discovered in Searles Lake (California). This issue is currently being studied and investigated by another working group at the GTK and will hopefully soon provide more clarity.

Line 789–791:

In order to determine whether the relatively high concentration of all analyzed trace elements in Raja pyrites are due to nanoparticles would require detailed high-resolution transmission electron microscopic measurements, which however is beyond the scope of this study.

RC: This is not true – with LA ICPMS you can usually detect nano-inclusions unless evenly disseminated.

Response: The reviewer is correct, nanoparticles are not likely going to be seen in the laser profile, since we are cutting down about 1 micron per second. Bigger inclusions will be seen, however, if they are a few nanometers, it is difficult to determine whether they are dispersed uniformly.

Line 802:

The negative shift in sulfur isotopic values and the large within-pyrite variations of $\delta^{34}\text{S}_{\text{VCDT}}$ values may indicate variation in relatively oxidizing and reducing conditions in the fluid system, related to local magmatic-hydrothermal and sedimentary sources, respectively (Hodkiewicz et al. 2009; Molnár et al. 2016b).

RC: This is highly unlikely – for having such variations in the SO₄-H₂S ratios you need to be in the stability field of sulfates... check some relevant references. Perhaps reflects mixing between magmatic and sedimentary sources. The positive values perhaps due to the inheritance of sulfur from a system with biogenic reduction of sulphate in a partially closed system.

Response: Sentence is rephrased to: The significant changes in Co/Ni ratios in pyrite B, and the wider range in sulfur isotopic values (-1.8 ‰ to +7.3 ‰), as well as the erratic within-grain variations of $\delta^{34}\text{S}_{\text{VCDT}}$ (up to 4‰), may be caused by the variation in the redox conditions in the fluid system (Ohmoto, 1972; Hodkiewicz et al. 2009; Molnár et al. 2016b). Such a variation is not uncommon in magmatic hydrothermal systems with

predominantly magmatic origin of sulfur. However, mixing of sulfur from different sources (e.g., from magmatic fluids and from metasediment-related fluids) cannot be excluded and in this latter case the elevated $\delta^{34}\text{S}_{\text{VCDT}}$ values may indicate influx of sedimentary sulfur produced by sulphate-reducing bacteria.

Ohmoto (1972) reference added to reference section:

Ohmoto, H.: Systematics of Sulfur and Carbon Isotopes in Hydrothermal Ore Deposits, *Econ. Geol.*, 67, 551–578, 1972.

Line 809:

The documented negative $\delta^{34}\text{S}_{\text{VCDT}}$ values for pyrite C (-1.2 ‰ to +7.4 ‰), as well as the elevated concentrations of redox sensitive elements (Ni, Cu, W, Ag, Hg, Mo and Zn) may also indicate the occasional shift of conditions to a more oxidizing state.

RC: The $\delta^{34}\text{S}$ depletion is very low and within the range of magmatic rocks, either leached from or degassing.

Response: The reviewer is correct with the statement that the $\delta^{34}\text{S}$ variation is small and still within the magmatic source range. The statement in line 809 is related to the variation in redox-sensitive element concentrations that is used to strengthen the argument that S-isotope variation reflects minor changes in redox conditions.

Line 816-onwards:

Section 5.5 Implications for mineral exploration

RC: This is dominantly a repetition of what has said before and a lot of vague statements. I would delete most of it and merge the key info with the conclusions

Response: Following this suggestion, the sections 5.5 and 6 are merged and shortened to section 6. Conclusions and implications to mineral exploration.

Referee # 2 | Solid Earth Discussion

Response to referee comment (RC) on "Application of lithochemical and pyrite trace element data for the determination of vectors to ore in the Raja Au-Co prospect, northern Finland" by Sara Raič et al., Solid Earth Discuss., <https://doi.org/10.5194/se-2021-119-RC2>, 2021

General Comments

RC: This manuscript, focussing on the Raja prospect, Finland, is an excellent study demonstrating the use of geochemical techniques and vector determination to aid in targeted mineral exploration. There is an abundance of detail throughout, though as a result the manuscript is longer than necessary. The authors should reduce the length of the text and avoid unnecessary background information. They should also avoid repetition, particularly in relation to principal component analysis discussions.

Response: Taking the reviewers comment regarding the length, repetition and too detailed background information into account, section of the manuscript to which this comment applies to have been reduced, such as the PCA and implications to mineralization parts of the discussion, as well as the conclusion.

RC: Within all figure captions, the analytical data sources should be stated – eg. In Figure 11, it should be stated that the data come from LA-ICP-MS analysis of pyrites. This allows the reader to assess the figures without reference to the text (they should be able to stand alone). Do this for all figure captions, where appropriate.

Response: The analytical data sources have been stated within figure captions that it applies to (Figs. 11 and 12), as suggested by the reviewer.

Specific Comments

All comments and suggestions made by the reviewer regarding typo, spelling and expression have been rephrased, accepted, and changed in the manuscript. These comprise the comments in the following lines: 20, 25, 28, 81, 144, 416, 422, 432, 509, 519, 552, 576, 620, 814, 851, 857, 872 and 883.

Comments made by the reviewer that would require a detailed response, clarification, or discussion, are addressed below.

Line 40:

An example of this kind of gold mineralization has recently been discovered in the Paleoproterozoic Peräpohja belt, within the Rajapalot project area owned by the Mawson Gold Ltd (centered at coordinates 3,408,707E by 7,372,441N of the Finnish coordinate system KKJ3).

RC: Given the international scope of the journal, please express coordinates using a globally utilized coordinate system.

Response: Changed to: [...] (centered at coordinates 408750.412294E by 7369184.18986N of the EUREF_FIN_TM35FIN coordinate system).

Lines 82–84:

The depositional history of these Karelian basins coincides with the Great Oxygenation Event, which created favorable conditions for a pre-concentration of metals (e.g. Co, Cu, Ni, Mo and Au) in sulfidic sediments, as well as the deposition of carbonaceous material (Melezhik et al. 2013).

RC: Numerous peer reviewed publications support the statement regarding the favorable conditions for the concentration of metals in basinal deposits during periods of atmospheric oxygenation, including Lyons et al., 2014; Large et al., 2014, 2015, 2017; Gregory et al., 2017; Johnson et al., 2017; Armstrong et al., 2018. Reference to some of these works should be made.

Response: Suggested references were included in the text, as well as in the reference section:

Armstrong, J.G.T., Parnell, J., Bullock, L.A., Perez, M., Boyce, A.J., Feldmann, J.: Tellurium, selenium and cobalt enrichment in Neoproterozoic black shales, Gwna Group, UK: Deep marine trace element enrichment during the Second Great Oxygenation Event, *Terra Nova*, 30, 244–253, <https://doi.org/10.1111/ter.12331>, 2018.

Gregory, D.D., Lyons, T.W., Large, R.R., Jiang, G., Stepanov, A.S., Diamond, C.W., Figueroa, M.C., and Olin, P.: Whole rock and discrete pyrite geochemistry as complementary tracers of ancient ocean chemistry: An example from the Neoproterozoic Doushantuo Formation, China, *Geochim. Cosmochim. Ac.*, 216, 201–220, <https://doi.org/10.1016/j.gca.2017.05.042>, 2017.

Johnson, S.C., Large, R.R., Coveney, R.M., Kelley, K.D., Slack, J.F., Steadman, J.A., Gregory, D.D., Sack, P.J., Meffre, S.: Secular distribution of highly metalliferous black shales correspond with peaks in past atmosphere oxygenation, *Miner. Deposita*, 52, 791–798, <https://doi.org/10.1007/s00126-017-0735-7>, 2017.

Large, R.R., Halpin, J.A., Danyushevsky, L. V., Maslennikov, V.V., Bull, S.W., Long, J.A., Gregory, D.G., Lounejeva, E., Lyons, T.W., Sack, P.J., McGoldrick, P.J., Calver, C.R.: Trace element content of sedimentary pyrite as a new proxy for deep-time ocean-atmosphere evolution, *Earth Planet. Sc. Lett.*, 389, 209–220, <https://doi.org/10.1016/j.epsl.2013.12.020>, 2014.

Large, R.R., Gregory, D.D., Steadman, J.A., Tomkins, A.G., Lounejeva, A., Danyushevsky, L. V., Halpin, J.A., Maslennikov, V., Sack, P.J., Mukherjee, I., Berry, R., Hickman, A.: Gold in the oceans through time, *Earth Planet. Sc. Lett.*, 428, 139–150, <https://doi.org/10.1016/j.epsl.2015.07.026>, 2015.

Large, R.R., Mukherjee, I., Gregory, D.D., Steadman, J.A., Maslennikov, V. v., Meffre, S.: Ocean and Atmosphere Geochemical Proxies Derived from Trace Elements in Marine Pyrite: Implications for Ore Genesis in Sedimentary Basins, *Econ. Geol.*, 112, 423–450, <https://dx.doi.org/10.2113/econgeo.112.2.423>, 2017.

Lyons, T.W., Reinhard, C.T., Planavsky, N.J.: The rise of oxygen in Earth's early ocean and atmosphere, *Nature*, 506, 307–315, <https://doi.org/10.1038/nature13068>, 2014.

Lines 99–101:

RC on Figure 1: Excellent figure and very detailed, though this should be a full page landscape figure to ensure detail is not illegible in final publication.

Response: Following the suggestion of the reviewer, Figure 1 is rearranged, in order to be completely clear and legible for the reader.

RC on Figure 1 caption: Data sources for maps (a) and (b) require citations.

Response: Sources added: (a) Geologic map of the main bedrock units of the eastern part of the Fennoscandian Shield and the Peräpohja belt as highlighted area (black rectangle). Simplified geological map is based on Koistinen et al. (2001) (b) Geologic map of northern Finland showing Archean and Proterozoic rocks. The Rajapalot Au-Co project is located within the shaded rectangle. Simplified map is modified from the Bedrock of Finland – DigiKP (2021; digital map database of the Geological Survey of Finland).

Both references are added to reference section:

Koistinen, T., Stephens, M. B., Bogatchev, V., Nordgulen, Ø., Wennerström, M. & Korhonen, J. (comp.): Geological map of the Fennoscandian Shield, scale 1:2 000 000. Trondheim:

Geological Survey of Norway, Uppsala: Geological Survey of Sweden, Moscow: Ministry of Natural Resources of Russia, Espoo: Geological Survey of Finland. <https://gtkdata.gtk.fi/kalliopera/index.html#>, 2001.

Bedrock of Finland – DigiKP: Digital Map Database, Geological Survey of Finland (GTK): <https://gtkdata.gtk.fi/kalliopera/index.html#>, last access: 06.12.2021.

Lines 205–222:

Trace element analyses in pyrite were performed by a laser ablation-inductively coupled plasma-mass spectrometry (LA-ICPMS) at the Geological Survey of Finland (GTK), using a Nu AttoM single collector (SC)-ICPMS (Nu Instruments Ltd., Wrexham, UK) and an Analyte Excite 193 ArF laser-ablation system (Photon Machines, San Diego, USA). The laser was run at a pulse frequency of 10 Hz and a pulse energy of 5 mJ at 30% attenuation to produce an energy flux of 2.5 J/cm² on the sample surface with a 50 µm spot size. Each analysis was [...].

RC: Was a hydrogen reaction cell utilized in the LA-ICP-MS analysis, to remove selenium analysis interferences? If not, the potential influence of this interference needs to be accounted for in the discussion. If a reaction cell was utilized in the analyses then this should be stated.

Response: We are aware of the formation of Ar-based polyatomic ions that affect the determination of ^{77,78,80}Se+ (⁴⁰Ar³⁶Ar¹H+, ⁴⁰Ar³⁸Ar+, and ⁴⁰Ar⁴⁰Ar+). However, all of these interferences are gas blank related, i.e., they are generated in the ICP and do not result from the laser ablation process. Consequently, since all of our analyses were acquired using TRA (Time Resolved Analysis) that in every case starts with a gas blank measurement, all gas-related polyatomic interferences are removed prior to concentration calculation. No reaction cell is required.

Lines 229–235:

One pyrite standard was used for external standard bracketing (PPP-1; Gilbert et al, JAAS, 2014) and the in-house standards Py1 and Py2 were used for quality control, both previously measured by gas mass spectrometry. For Py1, with a $\delta^{34}\text{S}_{\text{CDT}}(\text{‰})$ reference value of $-0.6 \pm 0.3 \text{ ‰}$ (1s) we measured an average value of $-0.6 \pm 0.5 \text{ ‰}$ (1s, n=26). For Py 2 with a $\delta^{34}\text{S}_{\text{CDT}}(\text{‰})$ reference value of $-0.3 \pm 0.3 \text{ ‰}$ (1s), we measured an average [...].

RC: Insert the reference value and analysed value for the PPP-1 external standard. The internal standards utilised in the S-isotope analysis calibration/QC span a very narrow $\delta^{34}\text{S}$ range (-1.2 to 0 ‰, including errors). The $\delta^{34}\text{S}$ values found during the sample analyses range outside the range of the standards (-1.76 to 7.40 ‰), making the calibration at the farthest ranges less reliable. This is a minor point as the difference between the standards and sample values is not substantial, but it is not good practice.

Response: The external standard has a certified value of $\delta^{34}\text{S}_{\text{CDT}} = +5.3 \pm 0.2 \text{ ‰}$ and the quality control standard are $-0.6 \pm 0.5 \text{ ‰}$ and $-0.3 \pm 0.3 \text{ ‰}$, so the isotopic range (about 6‰) is very close to the range of the samples (-1.76 to 7.40 ‰). We also routinely analyze an additional in-house quality control standard of pyrrhotite composition, having a value of +5.6‰, with similar accuracy to the two in house pyrites standards analyzed in this study.

Line 272:

In practice it turned out to work well, when only considering eigenvectors with eigenvalues > 1.

RC: “In practice it turned out to work well” – this is an opinion with no clear justification or scientific merit to the statement. Please rephrase.

Response: Sentence is deleted, but the statement eigenvalue >1 is included in the previous sentence (and Jolliffe, 2002 reference is added): After this standardization, the

data matrix Z is used for the computation of the correlation matrix PCA. In order to assure that most of the data variability is preserved, only principal components with large **eigenvalues** (> 1) are considered (see Kaiser, 1960; Jolliffe, 2002).

Jolliffe, I.T.: Principal Component Analysis, Second Edition, in: Springer Series in Statistics, edited by: Bickel, P., Diggle, S., Fienberg, K., Krickeberg, K., Olkin, I., Wermuth, N., Zeger, S., Springer, New York, USA, pp. 487, ISBN 0-387-95442-2, 2002.

Lines 277-279:

Out of the 50 elements analyzed within the whole-rock geochemical dataset, a subset of 12 elements was used (As, Au, Co, Cu, Mo, Ni, Se, Sn, Te, Tl, U and W). From the 61 elements analyzed in pyrites, a total of 12 were used to produce geochemical groupings with the PCA (Ag, As, Au, Bi, Co, Cu, Mo, Ni, Se, Te, Tl and W).

RC: Why were the subsets of 12 elements used different in the whole rock vs the LA-ICP-MS data? Did the statistical method not work on certain elements, or were these analyses not available from the given analytical methods?

Response: For the two different datasets, only elements were used that would contribute to the building of a predictive model when using the algorithms of the PCA. For this purpose, one has to consider a dimensionality reduction of our data matrix. To achieve dimensionality reduction, elimination of a subset of variables must be considered, rather than the elimination of entire observations. For this purpose, the following must be considered: missing data (gaps in data collection, high proportion of missing values, other errors), low or zero variance throughout (normalized) observations, or high correlations between variables (too many similar features are not useful in enhancing the predictive power of the model). When considering all these features ("pre-modelling" → covariance matrix, correlation matrix, missing values, etc.), the two different subsets of variables that were used in this study, could be identified as the "key" elements for the PCA.

Lines 327-335:

Section 4.2.1 Alteration types

RC: This section does not appear to be providing results of the new analyses, but summarising the alteration events as identified in previous works. If there are no new results, this should be in the intro and/or discussion sections. If evidence of this alteration has been observed, then this information should be incorporated into sections 4.1 or 4.2.2. Either way, this section should be incorporated elsewhere within the text or removed.

Response: Following the suggestion of the reviewer, the section 4.2.1 was incorporated into 4.1 and section 4.2 was changed from "Alteration types" to "Ore textures and Mineralogy". The following section is added to 4.1.: The most common alteration types include albitization (pre-post-syn-orogenic), retrograde alteration assemblages of chloritized biotite, chlorite and Fe-Mg amphiboles (syn-orogenic), sulfidation-induced formation of massive pyrrhotite and subordinate pyrite and chalcopyrite (syn-to post-orogenic), as well as sections where plagioclase is affected by the sericitic alteration characterize the latest alteration event (post-orogenic; see Table 1).

Line 358:

Pentlandite occurs as flame-like exsolution lamellae (few microns) or as anhedral inclusions in pyrrhotite.

RC: Provide a numerical value for the pentlandite exsolution lamellae.

Response: The size of exsolution lamellae is $<20\mu\text{m}$; changed to: Pentlandite occurs as flame-like exsolution lamellae ($<20\mu\text{m}$) or as anhedral inclusions in pyrrhotite.

Line 360:

The Bi-Te-rich phases also occur along grain contacts with biotite (Fig. 5d). Given the small grain size of the Bi-Te-rich phases, no useful analyses could be produced to define their respective mineral names.

RC: Do the numerous Bi-Te-rich phases have a consistent composition (and thus potentially mineralogy?), or are they highly variable? As is mentioned, the size precludes accurate mineral identification, but does the composition appear similar across most/all of the Bi-Te-rich phases?

Response: Unfortunately, no useful analyses could be produced for the Bi-Te-rich phases given their small grain size. However, identification with the SEM gave a composition between 18.61 and 34.38 wt.% for Bi, and a Te-composition between 13.10 and 54.73 wt.%. Additional components that showed a signal are Co, Fe, Mo, As and S (when in assemblage with cobaltite, sulfides, and molybdenite). These estimates made by the SEM indeed show a compositional variation. It is however difficult to determine whether the varying chemistries reflect separate mineral phases or are the result of inaccurate signals due to grain size.

Figure 5: Above Line 365

RC on Figure 5: Are all 12 images necessary to convey the extent of mineralisation? This seems excessive.

Response: As suggested by the reviewer, the number of photomicrographs in Figure 5 was reduced from 12 to 6. The figure caption is adjusted accordingly-

Line 514:

Geochemical gradients recognized from the correlation of alteration mineralogy with Au- and Co-grades in MER dimensions exhibit distinct trends [...].

RC: "MER" – this acronym has not been stated in the text. This should be given in brackets after the term on line 508.

Response: The acronym "MER" is first mentioned in section 3.3.2 Molar element ratio analysis, **Lines 283–285:** Molar element ratio (MER) analysis and corresponding plots of $3K/(Al + Mg + (Fe - S))$ versus $Al/(Al + Mg + (Fe - S))$, and $(Na + K)/Al$ versus Na/Al , are the variables and ratios that best describe the observed alteration assemblages.

Figure 10: Above Line 525

RC on Figure 10: Figure 10 – (e) and (f) require legends for Au and Co. Presumably they are the same as those used in (b) and (c), but this is not clearly stated nor very obvious.

Response: Following the suggestion of the reviewer, it is clarified in the Figure caption that the legends from Figs 10 b-c are applicable for Figs. 10 e-f. Figure caption is rephrased to: Figure 10: Graphical presentation of mass transfer processes at Raja in molar element ratio diagrams in correlation with Au- and Co-grades. Axes are molar ratios. Diagrams are taken from Stanley (2017). Molar ratio plots of $3K/(Al+Mg+(Fe-S))$ versus $Al/(Al+Mg+Fe-S)$ classified according to (a) lithologies, (b) Au-concentrations (in ppm) and (c) Co-concentrations (in ppm). General element ratio diagrams using $(Na+K)/Al$ versus Na/Al and classified according to (d) lithologies, (e) Au-concentrations (in ppm) and (f) Co-concentrations (in ppm). **Lithology and concentration legends from (a), (b) and (c) are also used in (d), (e) and (f).** [Ab = Albite, Act = Actinolite, Bt = Biotite, Chl = Chlorite, Kf = K-Feldspar, Ser = Sericite].

Lines 623–624:

Pyrite A, which has the highest Co/Ni ratios, displays the heaviest $\delta^{34}\text{S}$ signatures ($\delta^{34}\text{S}$ (1.3 ‰ to +5.9 ‰), contrasting with pyrite B (-1.8 ‰ to +7.3 ‰) and pyrite C (-1.2 ‰ to +7.4 ‰; Fig. 13b; Table 3).

RC: From the data discussed, Pyrite A has a ‘lighter’ (lower) max $\delta^{34}\text{S}$ (+5.9‰) than pyrite B (+7.3‰) or pyrite c (+7.4‰). This therefore does not constitute “the heaviest $\delta^{34}\text{S}$ signatures”. This statement needs to be changed. Also, use higher/lower, rather than lighter/heavier when discussing $\delta^{34}\text{S}$ values.

Response: Rephrased to: Pyrite A, which has the highest Co/Ni ratios, displays positive only $\delta^{34}\text{S}$ signatures (+1.3 ‰ to +5.9 ‰), contrasting with pyrite B (-1.8 ‰ to +7.3 ‰) and pyrite C (-1.2 ‰ to +7.4 ‰; Fig. 13b; Table 3).

Lines 787–789:

Although a careful data checking of LA-ICP-MS ablation profiles for possible ablated inclusions was carried out, the presence of nanoparticles containing portions of trace elements should not be excluded (Cook et al., 2009).

RC: What is the spatial resolution of the LA-ICP-MS analysis that was conducted? This would provide an approximate value for which the potential nanoparticles would be smaller, if present.

Response: With a spot size of 40 micron, a 5Hz repetition rate, an ablation rate of 0.5micron per second and a scanning speed in the order of 0.2s across the mass range, an inclusion of about 1 micron represent a volume of 0.06% of the total ablated mass, so we will not be able to see nanometer inclusions.

Line 805:

In addition, the Se/S ratios of this pyrite type display a much wider scattering (4.00×10^{-6} to 0.003) and a higher mean value (4.12×10^{-4}) compared to pyrite A (2.93×10^{-4}).

RC: As mentioned earlier in the text, the Se/S ratios for pyrite C are similarly wide ranging, so using mean Se/S for a comparison between Pyrites A and C is misleading.

Response: Sentence is misleading and is removed.

Lines 816–879:**Section 5.5 Implications for mineral exploration**

RC: Much of this section is repetition of previous sections with limited further discussion. Re-write and distil this section down to key points regarding the “Implications for mineral exploration”. Lists of positive and negative PC loadings of elements are not warranted here.

Response: Following this suggestion, the sections 5.5 and 6 are merged and shortened to section 6. Conclusions and implications to mineral exploration.

Lines 865–879:

RC: Given the overlapping ranges of $\delta^{34}\text{S}$ in all the pyrite types, it should be stated that sulphur isotopes are not a distinguishing factor between different pyrite generations (and thus mineralization zones).

Response: Following the suggestion of the review, the following was included in the rephrased section 5.4 Signatures of sulfur isotopes in pyrite and ore-forming processes: The overall $\delta^{34}\text{S}_{\text{VCDT}}$ data of pyrite grains at Raja are between -1.8 ‰ and +7.4 ‰, covering a common range of sulfur isotope composition for orogenic gold deposits (Molnár et al., 2016a). Considering the similarities of $\delta^{34}\text{S}_{\text{VCDT}}$ values in each pyrite type, these signatures

are not diagnostic, but rather indicative of anomalous Co- or Au-zones. However, the combined use of trace element geochemistry and S-isotopic signatures in pyrite can help to discuss the behavior of S, hydrothermal processes, fluid/rock interactions and accumulation of metals during the formation of the Raja Au-Co prospect.

Line 885:

The LA-ICP-MS study of sulfides, specifically the ubiquitously present pyrite, offers a high spatial resolution and a novel approach in the determination of vectors to ore.

RC: As mentioned before, you have not utilised the high spatial resolution of LA-ICPMS in this study by assessing the variation in trace element concentrations between pyrite rims and cores. You have averaged out the elemental concentrations within each pyrite, potentially missing important evidence for the timing of mineralizing fluids. I suggest revisiting this data, where feasible.

Response: The reviewer is correct with the statement that in the evaluation of the large-scale picture the focus is mostly put on the averaged data from texturally distinct pyrite B. However, when looking at each data point regarding the data point position within the grains, the core-rim variations showed a very erratic behavior that did not allow any systematic categorization or a general establishment of a core-rim trend. This, however, is what is to be expected when the variation is related to variable redox conditions, or fluid mixing in a dynamic system (where the actual value is depending on the variable masses of mixing fluids). Regarding the more detailed study of this pyrite type, the reviewer is correct with the statement. However, the aim of this study was to find endmembers that could provide potential vectors to exploration targeting, which is why more focus was put on the two endmember pyrite types.

Lines 880 onwards:

RC: Much of this section is repetition and/or unnecessary statements regarding the purpose of the study. Please rework this section to be more succinct.

Response: Following this suggestion, the sections 5.5 and 6 are merged and shortened to section 6. Conclusions and implications to mineral exploration.

Lines 893–897:

RC: These statements/ore formation models should be made explicitly in the discussion section and summarized in the conclusions. Ore-formation models should not be first proposed in the conclusions.

Response: Following the suggestion of the reviewer, these statements were included in the rephrased section 5.4 Signatures of sulfur isotopes in pyrite and ore-forming processes.