

New methodology for air mercury surveys over mineralization

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<https://doi.org/10.70499/ZXZS8433>

ABSTRACT

Mercury (Hg) is one of the most mobile elements in the Earth's crust and is involved in all geological processes, including ore formation. Elevated mercury concentrations in primary and secondary scattering haloes are found in most endogenous ore deposits. High contents of this element are also found in some hydrocarbon deposits. The consequence is the high informativeness of geochemical exploration based on studies of primary and secondary litho-, hydro- and atmospheric mercury haloes. The latter are formed in soil gas and atmospheric air due to the unique physical and chemical properties of mercury. In various geological settings, mercury concentrations span more than six orders of magnitude, which requires dedicated analytical solutions. Gaseous elemental mercury (GEM or Hg⁰) is the only metal that is currently detectable in air by direct instrumental analysis. Portable instruments based on Zeeman atomic absorption spectroscopy have been developed to directly measure mercury concentrations in ambient air. Recent data obtained at sediment-covered mineralization and ore-controlling fault zones show the effectiveness of real-time Hg⁰ vapour measurement in near-surface air above Hg, Au, Au-Ag-Cu, polymetallic, and rare-metal mineralization under various geochemical and climatic conditions.

MERCURY IN EXPLORATION GEOCHEMISTRY

In geochemistry, mercury is classified as a trace element. Mercury abundance in the Earth's crust (the Clarke value) is estimated as 45 (30–80) ppb (µg/kg) (Saukov et al. 1972). Mercury is a scattered element with approximately equal near-Clarke content in igneous, metamorphic and sedimentary rocks (Saukov 1946). Mercury ore deposits contain only 0.02% of the total Hg planetary budget (Smirnov et al. 1976). Elevated levels of mercury have been documented in ore minerals of various magmatic-hydrothermal deposits (Saukov 1946; McCarthy et al. 1969; McCarthy 1972; Fursov 1983; Carr et al. 1984; Ozerova 1986; Rasmussen 1994). Mercury forms its own minerals and may be isomorphically incorporated into the crystal lattice, or absorbed as elemental mercury Hg⁰ into pores and on the surface of minerals in ores and host rocks.


The concentration of mercury in a mineral depends on the temperature of formation, with the mercury content ranging from the Clarke value to percent by weight. Other than mercury minerals, mercury (maximum concentration, wt %) has been reported in sphalerite (36), tetrahedrite/tennantite (21.5), native gold (16) and silver (3), pyrite (6), realgar (0.6), gudmundite (0.24), antimonite (0.2), and other ore and non-metallic (barite, carbonates, dickite, fluorite, quartz, sericite) minerals (Saukov et al. 1972). The general trend of mercury behaviour in endogenic deposits is an increase in mercury content from higher to lower temperature minerals. In hydrothermal ore deposits, mercury enrichment is observed in minerals comprising the final, low-temperature stages of ore formation (Saukov et al. 1972; Ozerova 1986).

In fossil fuels, mercury concentration also can vary in a wide range, covering six orders of magnitude: coal 1–300,000 ppb; oil 1–600,000 ppb; natural gas 1–5,000,000 ng/m³ (Ozerova 1986; Ozerova et al. 1999; Yudovich and Ketris 2005). The mercury and mercury-containing ore and hydrocarbon deposits are mainly located within global mercury belts such as the Mediterranean, Central Asia, and Circum-Pacific (Smirnov et al. 1976; Ozerova et al. 1999).

The physicochemical properties of mercury determine its presence in the Earth's crust in elemental form. The main mechanisms for the formation of elemental mercury are its evaporation from mercury-containing minerals, chemical and electrochemical reactions in the hypergene zone releasing bound mercury from sulphides (Saukov 1946; Smirnov 1955; Sveshnikov 1967; Kothny 1973; Fursov 1983). In 1946, A.A. Saukov defined the state of scattered mercury in the Earth's crust as "quasi-gaseous" and predicted the existence of a "mercury atmosphere" of ore deposits and a high informativity of lithochemical and gaseous mercury surveys (Saukov 1946). Subvertical transport of mercury from buried sources to the

continued on page 5

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New methodology for air mercury surveys over mineralization *continued from page 1*

surface, as with other geogenic gases, is driven by vertical gradients of pressure, temperature and concentration (Fig. 1).

Subsequent studies around the world have revealed the presence of lithochemical mercury haloes at almost all types of hydrothermal ore deposits. These Hg haloes are much wider than ore alteration zones and haloes of other ore elements. They can extend for hundreds of metres horizontally and vertically (Ozerova 1986; Fursov 2006). In contrast to lithochemical dispersion haloes, superimposed mercury haloes form directly above mineralization or associated fault zones in overlying transported overburden due to the high mobility of gaseous mercury.

The “mercury atmosphere” of ore deposits offers the possibility of using gaseous mercury haloes for geochemical exploration. Mercury is the only metal that forms vapour haloes in soil gas and near-surface atmosphere that can currently be directly detected. The first registration of mercury haloes in near-surface air and soil gas was made by E.A. Sergeev at the Khaidarkan mercury deposit using air sampling to the I₂ in KI solution and colourimetric Hg determination (Sergeev 1957, 1961). Soon after, instrumental methods, based on atomic absorption (AAS) and atomic fluorescence spectroscopy (AFS), were developed for the determination of mercury in atmospheric and subsurface (soil gas) air (Hawkes and Williston 1962; Stepanov et al. 1965; Barringer 1966; McCarthy et al. 1969; Robbins 1973; Fursov 1977; Zherebtsov et al. 1992). Most instrumental solutions require mercury preconcentration on a sorbent (mainly gold traps) to collect enough mercury for analysis and separate it from interfering

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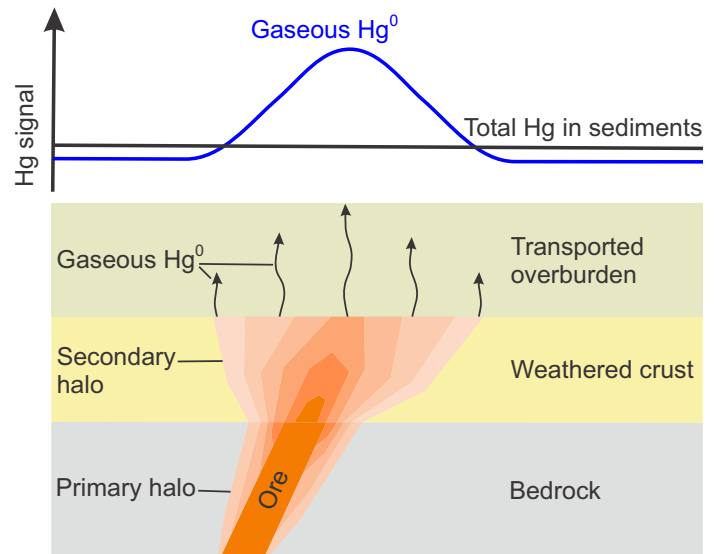


Fig. 1. Schematic model of mercury haloes above ore mineralization. Note that lithochemical anomalies can be lacking or displaced in transported overburden. In contrast, gaseous Hg⁰ haloes in near-surface atmosphere occur directly above mineralization.

components. However, humidity and some organic and non-organic volatiles can influence sorption efficiency of the gold trap.

Soil gas sampling is becoming a common technique in prospecting. The method has proven successful in the discovery and mapping of polymetallic, gold, and rare metal deposits (e.g. Hawkes and Williston 1962; McCarthy et al. 1969; McCarthy 1972; Robbins 1973; Zonghua and Yangfen 1981; Zherebtsov et al. 1992; Rehn and Rehn 1996; Fursov 2006). However, soil gas sampling requires homogeneous surface sediments and cannot be carried out in areas covered by waterbodies, bogs, permafrost, felsenmeer, or at outcrops of bedrock. In contrast to soil gas, direct analysis of Hg in air can be performed above any surface.

Most atmospheric mercury is gaseous elemental mercury (GEM or Hg⁰), which represents 90 to 99% of the total mercury, with a background concentration of 1.5–1.6 and 0.97–1.2 ng/m³ for the northern and southern hemispheres, respectively. Background concentration of gaseous oxidized mercury (GOM) and particulate bound mercury (PBM) is in the pg/m³ level (Sprovieri et al. 2016; Bencardino et al. 2024). Elemental mercury is the most mobile form, tending to move from buried sources to the natural land surface and then dissipate into the atmosphere as atomic vapour Hg⁰. This movement enables the direct detection of mercury in air by atomic absorption spectroscopy. Mercury concentration in air haloes above ore deposits can range from a few to hundreds of ng/m³ (Mashyanov 1980). New instrumental solutions have revived interest in the use of the air mercury survey for ore deposit prospecting. Recently, the RA-915M analyzer was used to study mercury haloes in air at Au,

New methodology for air mercury surveys over mineralization *continued from page 5*

Hg-Ag, and polymetallic occurrences at Salair Ridge, south-central Russia (Nevolko and Fominykh 2017) and on Vancouver Island in British Columbia, Canada (Rukhlov et al. 2021, 2022). In our work, we have evaluated different methods of air sampling to optimize the survey methodology.

Analyzers for field gaseous mercury surveys must be portable and have a detection limit at, or below, the atmospheric background mercury content ($\sim 1 \text{ ng/m}^3$), as well as high selectivity. These requirements are met by the analyzers based on atomic absorption spectroscopy with Zeeman correction for background absorption (ZAAS).



Fig. 2. The RA-915M portable Zeeman AAS analyzer.

MATERIALS AND METHODS

Mercury ZAAS analyzers and monitors in the RA-915 family (Lumex Instruments, Mission, British Columbia, Canada) have been designed for direct mercury determination in air, gases, liquids, and solids (Sholupov and Ganeyev 1995; Sholupov et al. 2004). The combination of the multipath analytical cell with Zeeman correction for background absorption enables direct and continuous measurement of the background mercury concentration in ambient air in real time. The RA-915AM monitor is used for the long-term non-attended Hg measurement in air monitoring networks, such as GMOS (Global Mercury Observation System) and GOS4M (Global Observation System for Mercury; Sprovieri et al. 2016; Mashyanov et al. 2021; Martino et al. 2022; Sholupov et al. 2022; Bencardino et al. 2024). The portable analyzer RA-915M (Fig. 2) is widely used for background air monitoring, indoor and outdoor pollution detection and mapping, continuous measurements from vehicles (Higueras et al. 2014; Ciani et al. 2021; Mashyanov et al. 2021; Cabassi et al. 2022), and geochemical surveys (e.g. Rukhlov et al. 2021, 2022).

For continuous measurements, analyzed air is continuously pumped at a flow rate of 8–10 L/min through the multipath analytical cell with the effective optical length of 9.6 m.

Readings are collected every 1 s and stored in the built-in, non-volatile memory for further processing (12 hours of 1 s data storage). An alternative air intake inlet with a built-in, high-efficiency (>98%) sorption filter (activated carbon) removing mercury from the sampled air is periodically switched on for baseline (zero) checks to correct the instrumental drift. To detect subtle anomalies near the background mercury concentration of 1.0 to 1.5 ng/m^3 , zero checks can be made when moving to the next point (i.e. every 1 to 2 minutes), or at least every 10 to 20 minutes. The limit of detection (LoD) defined as three times the standard deviation of the blank measurement, was 0.5 ng/m^3 Hg. The built-in battery provides at least 8 hours of operation, making this analyzer a convenient tool for field work. Raw data can also be processed in real time, with the individual measurements (one second each) normalized to standard pressure (101.3 kPa) and temperature (20°C), corrected for instrumental drift, and displayed on a 'live' graph in terms of time (hh:mm:ss) versus Hg concentration (ng/m^3) diagram using an external PC Notebook or Palm PC running a dedicated RAPID software (Lumex Instruments 2018).

continued on page 9

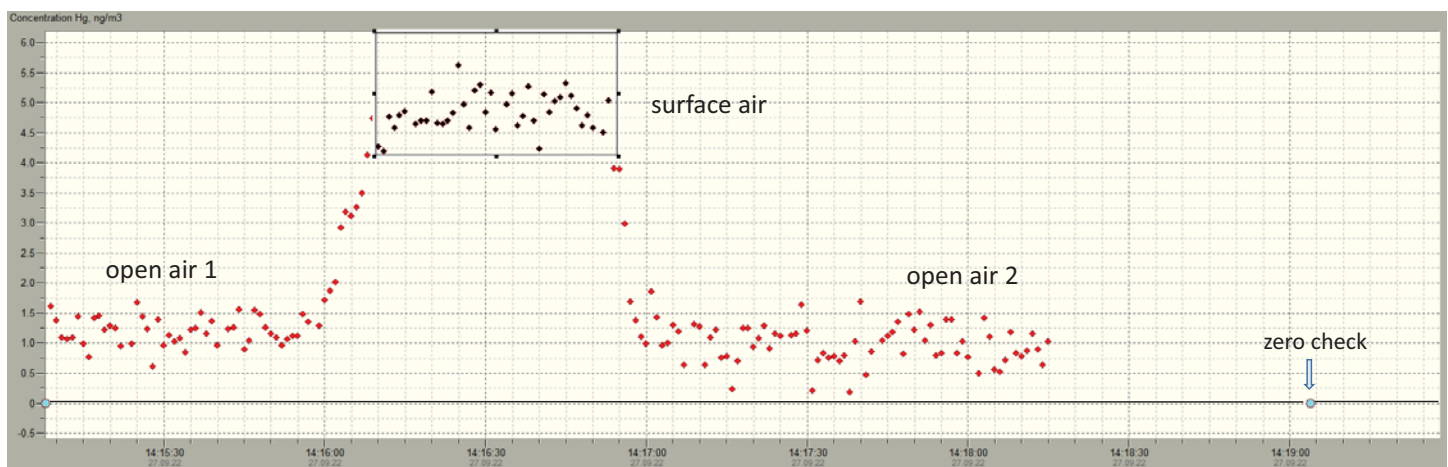
New methodology for air mercury surveys over mineralization *continued from page 8*



Fig. 3. Near-surface air sampling (a) 1 cm above undisturbed surface and (b) after removing the top 5 to 15 cm layer across an area of ca. ~20 cm in diameter. White tubing transports air collected from the bucket probe to the mercury analyzer inside the black bag.

Real-time measurements of mercury vapour (Hg^0) mass concentrations (in ng/m^3) were made using the RA-915M standard 'Monitoring mode', sampling both open air (0.1 to 0.5 m above ground) and near-surface air (1 cm above ground; Rukhlov et al. 2021, 2022). For near-surface air sampling, a 4-litre bucket probe is placed on the ground to minimize the influence of wind (Yasutake et al. 2011; Rukhlov et al. 2022). With the probe bucket, air was sampled above the undisturbed surface (Fig. 3a) and immediately after mechanically agitating or removing the top 5 to 15 cm layer of surface material (Fig. 3b).

Once the probe bucket has been placed in position for sampling, it takes 15 to 20 s to fill the multipath cell with the sampled air plus 40 to 60 s to collect the data. The raw data are stored in the built-in, non-volatile memory and allow further processing by the RAPID software. The data file related to each data collection point is identified by the time stamp; the time interval of 40 to 60 s with a steady signal level is selected on the graph, for which the mean concentration and the standard deviation are calculated using RAPID software (Fig. 4).



Statistics

N	Beginning of the range	End of the range	X mean, ng/m^3	SD, ng/m^3	X min, ng/m^3	X max, ng/m^3	Comment
1	27.09.2022 14:15:10	27.09.2022 14:15:50	1.20	0.23	0.62	1.68	open air 1
2	27.09.2022 14:16:05	27.09.2022 14:16:54	4.71	0.50	3.12	5.63	surface air
3	27.09.2022 14:17:37	27.09.2022 14:18:08	1.02	0.36	0.22	1.70	open air 2

Fig. 4. Real-time measurement and calculation of average gaseous elemental mercury concentration (X mean, ng/m^3), which was carried out using the RAPID software. 'Open air' – air sampled 0.1 to 0.5 m above ground; 'surface air' - air sampled 1 cm above ground under a bucket probe. Mean mercury concentration (\pm standard deviation, SD, ng/m^3) in the open air: 1.20 ± 0.23 (point 1) and 1.02 ± 0.36 (point 2); in the surface air under a bucket probe: 4.71 ± 0.50 .

continued on page 11

New methodology for air mercury surveys over mineralization *continued from page 9*

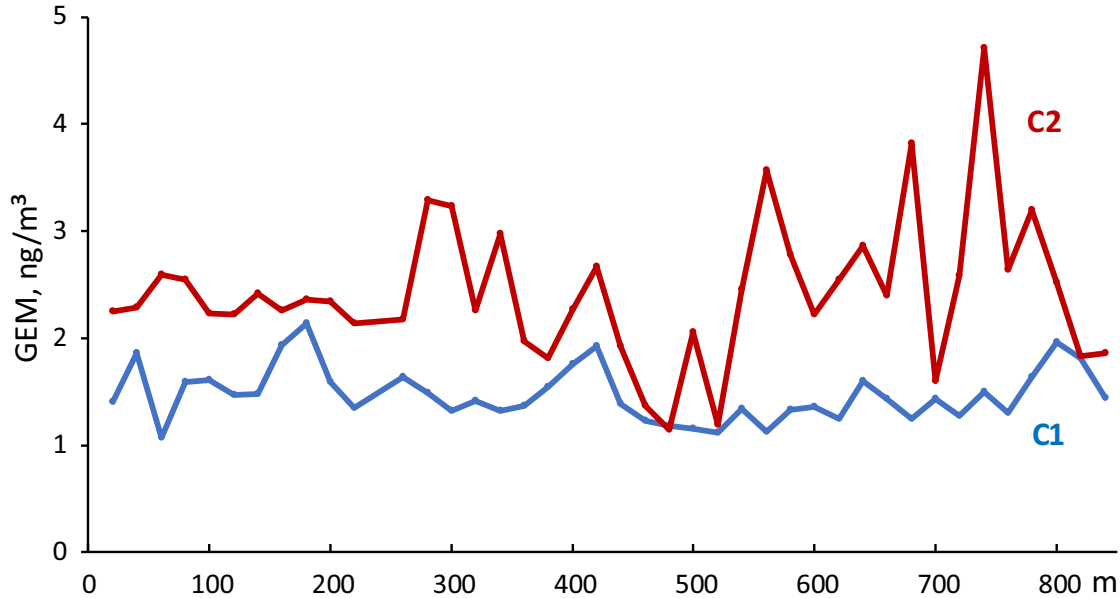


Fig. 5. Distance (m) along traverse versus mean gaseous elemental mercury (GEM) concentration (ng/m^3) in air 1 cm above undisturbed surface (blue line C1) and immediately after removing the top 5 to 15 cm layer (red line C2); air temperature 8–12°C, wind speed 10–13 m/s; central Kazakhstan, September 2022.

The calculated average mercury concentrations are plotted versus distance along a traverse on a graph and can then be evaluated in conjunction with geological, geophysical, or other geochemical information. At the background points, the mercury concentrations in open air and near-surface air are practically the same and are at the level of the background air concentration. In anomalous areas or in high wind conditions, more stable and contrasting values were recorded in air 1 cm above ground using the bucket probe, immediately after removing the top 5 to 15 cm layer of surface material. This technique allows measurements to be made even in unfavourable meteorological conditions. Figure 5 shows the results of an experimental air mercury survey at a prospecting site in central Kazakhstan. The survey was carried out at a wind speed of 10 to 13 m/s and an air temperature of 8 to 12°C. Compared to the mercury contents in open air 0.5 to 1 m above ground, more contrasting mercury anomalies are observed in the near-surface air 1 cm above ground when measured using a bucket probe, immediately after removing the top 5 to 15 cm of the soil layer (Fig. 5).

continued on page 12

New methodology for air mercury surveys over mineralization *continued from page 11*

Fig. 6. Sampling air mercury 1 cm above ground using a bucket probe after removing the top 5 to 15 cm layer across a sampling area of ~20 cm in diameter. **a)** Central Kazakhstan, 2022. **b)** Central British Columbia, Canada, 2023.



To detect weak anomalies, we recommend sampling air 1 cm above ground using a bucket probe immediately after removing or mechanically agitating the top 5 to 15 cm layer (Fig. 6). Measurements are made in continuous ‘Monitoring mode’ with 1 s data acquisition for about 1 min. The average mercury concentration at the sampling point is calculated using a 30 to 50 s steady-state signal fragment (Fig. 4). The periodicity of the zero check depends on the stability of the external conditions, such as air temperature. For low Hg concentrations, the baseline should be checked at least every 10 minutes or between every measurement point (e.g. Rukhlov et al. 2022). This technique has been developed and applied in experimental air mercury surveys at sediment-covered, polymetallic mineralization in British Columbia, Canada, as discussed below.

SURVEY RESULTS

The air mercury surveys using a RA-915M mercury analyzer were carried out in two areas in British Columbia, Canada: (1) Lara-Coronation Zn-Cu-Pb-Ag-Au occurrence on Vancouver Island, and (2) Silver Hope Cu-Ag-Au mineralization in central British Columbia (Fig. 7).

Lara-Coronation Zn-Cu-Pb-Ag-Au volcanogenic massive sulphide occurrence

The Lara-Coronation polymetallic occurrence comprises volcanogenic massive sulphide (VMS) Zn-Cu-Pb-Ag-Au mineralization hosted by the Sicker Group volcanic rocks on Vancouver Island (Rukhlov et al. 2022 and references therein). The mineralization consists of known mercury concentrator-minerals, such as sphalerite, pyrite, chalcopyrite, and galena, with minor tetrahedrite, tennantite, bornite,

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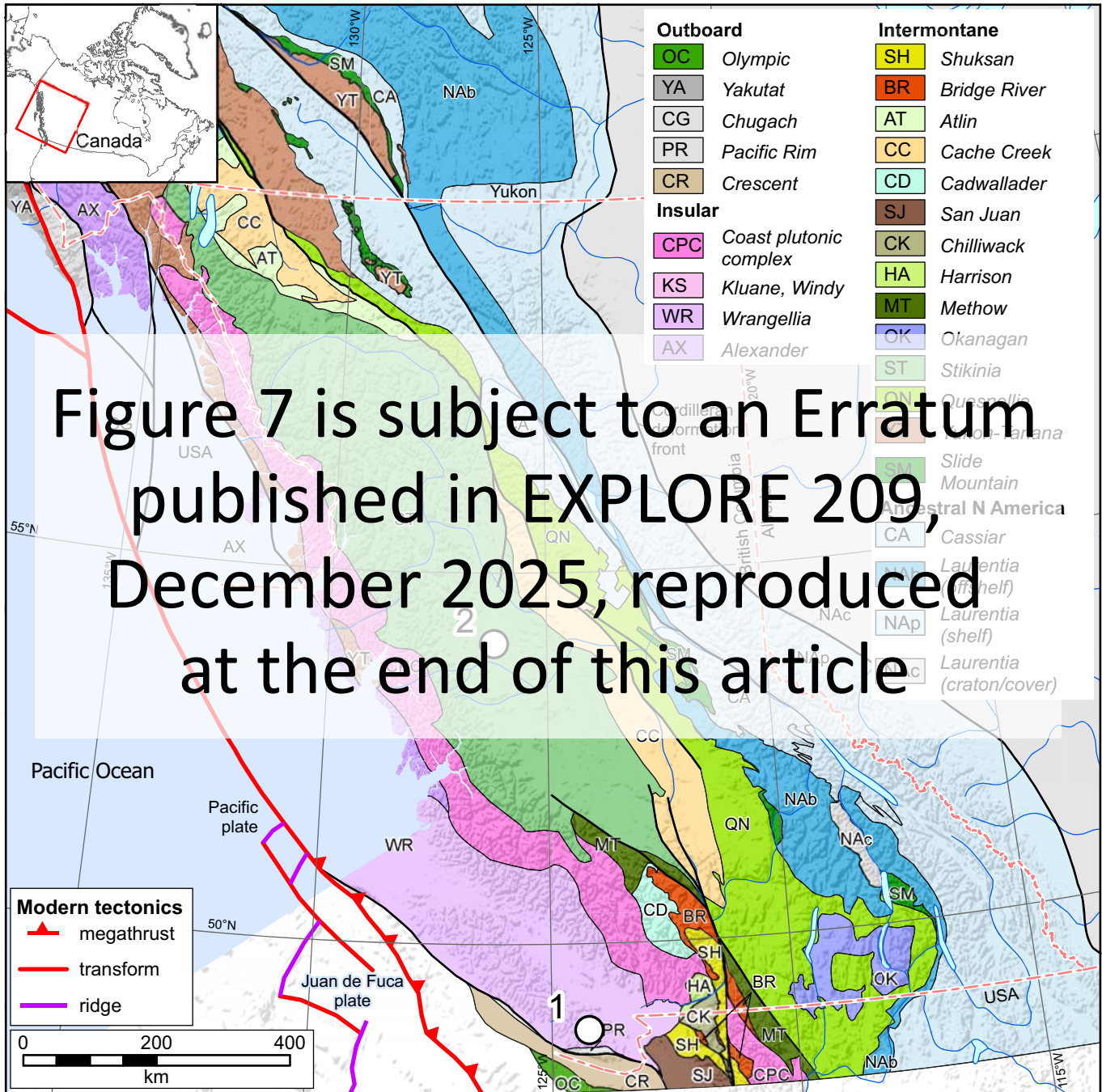


Fig. 7. Air mercury survey sites at mineral occurrences in British Columbia, Canada: 1 - Lara-Coronation, 2 - Silver Hope. Terranes after Colpron (2020).

electrum, pearceite, and arsenopyrite. The area is mostly covered by up to 30 m of glacial sediments (Kapusta et al. 1988; Bodnar 2017).

The air mercury survey data and methodology are described in detail in Rukhlov et al. (2021, 2022). We performed orientation surveys across known mineralization, sampling air 1 to 50 cm above ground while simultaneously measuring meteorological parameters, such as air temperature, relative humidity, absolute atmospheric (station) pressure, wind speed, and wind direction, in August 2020 and July 2021. The initial reconnaissance survey in 2020 involved continuous (one per second) measurements,

New methodology for air mercury surveys over mineralization *continued from page 13*

sampling air 2 to 50 cm above ground during foot traversing. We used an average of ten consecutive readings, which were automatically stored in the analyzer's memory. Near-surface air was analyzed for at least 20–30 seconds at each stop every 2 to 50 m along a traverse (Rukhlov et al. 2021). In contrast, the 2021 survey sampled air 1 cm above ground using a 4.2 L bucket probe (21 cm diameter), collecting 60 to 120 readings (1 second each) per sampling point above both undisturbed surface and immediately after mechanically agitating or removing the top 5–15 cm layer across an area of ~20 cm in diameter. Sampling points were spaced 4 to 36 m along traverses (260 to 500 m long) and spaced 25 to 50 m apart to enable areal gridding of the results (Rukhlov et al. 2022). We found that the mineralized zone was marked by a strong anomaly of up to 250 ng/m³ Hg in near-surface air. Background mercury concentration outside of mineralized zones was 1.2 ng/m³ (Fig. 8).

Silver Hope Cu-Ag-Au occurrence

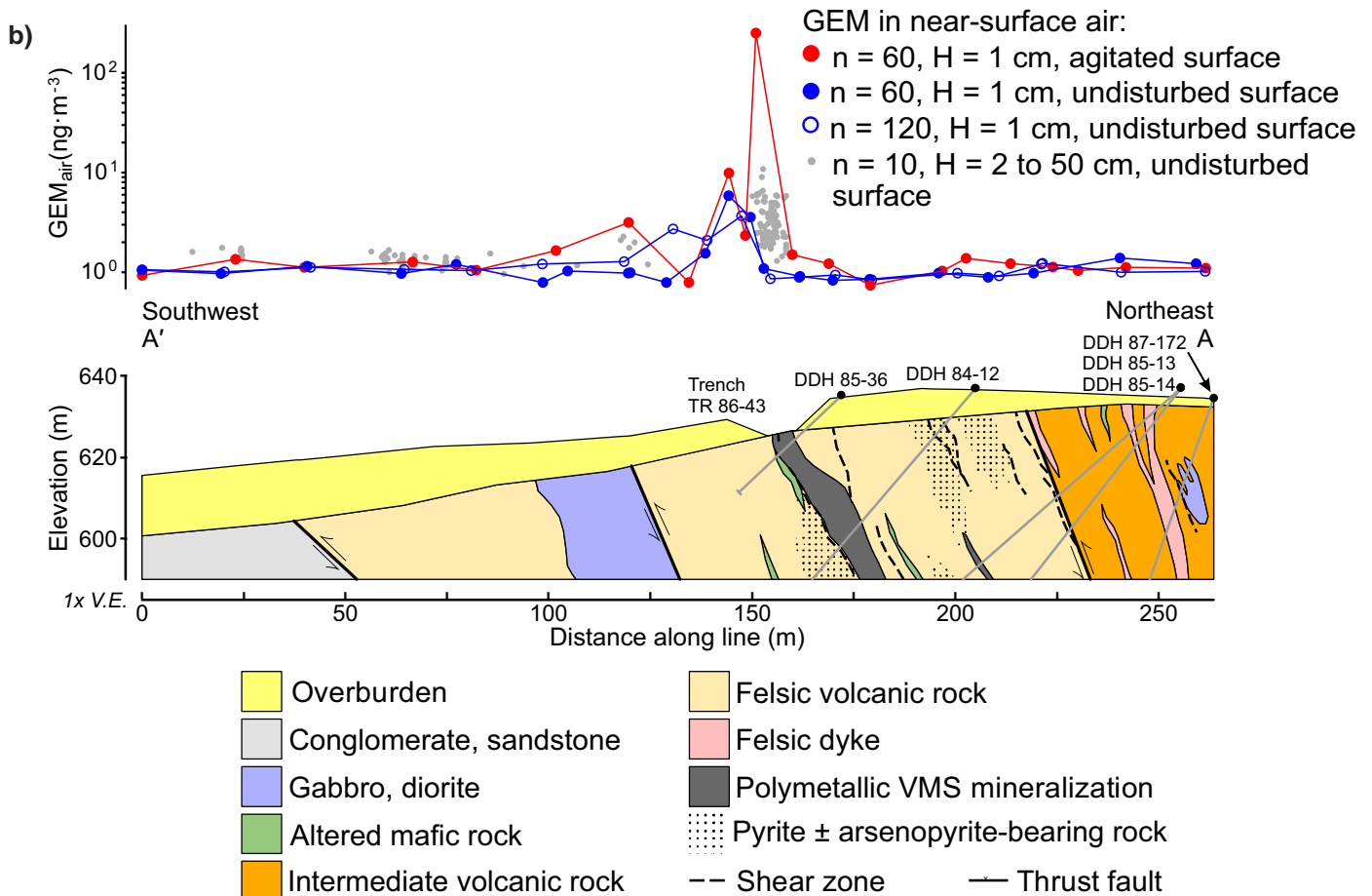
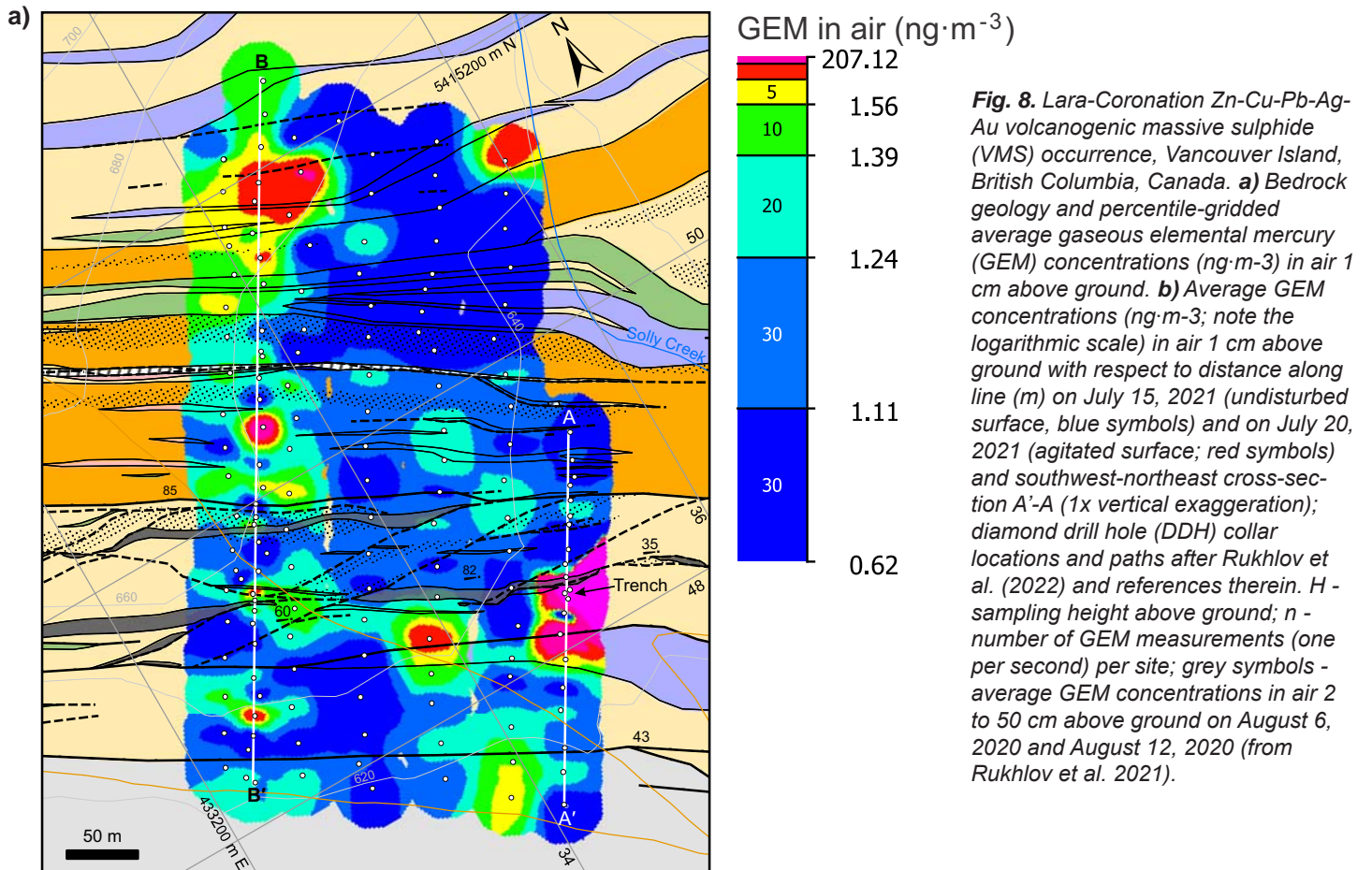
At the Silver Hope occurrence in central British Columbia, Canada (Fig. 9a), an air mercury survey was carried out along 14 profiles on the flanks of known porphyry Cu-Ag-Au mineralization (Tamburri and Barnes 2024). The main ore zone is clearly identified by a halo in the near-surface air with a maximum of 24 ng/m³ Hg and a background concentration of 1.3 ng/m³ Hg (Fig. 9b). The thickness of the glacial sediments along profile 6006200N varies from 10 to 30 m and thins to the east of the main zone.

CONCLUSIONS

The Lumex RA-915M portable analyzer enables direct mercury determination in air starting from the background level of 1–2 ng/m³, even under unfavourable meteorological conditions such as strong wind and low ambient temperature. This analytical technique detects weak gaseous Hg emissions directly above sulphide mineralization, including sediment-covered orebodies and related fault zones. In contrast to soil gas sampling, gaseous Hg haloes in near-surface air can be detected over outcrops, wet soils, bogs, salt marshes, permafrost, snow, and other types of surface materials.

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New methodology for air mercury surveys over mineralization *continued from page 14*



New methodology for air mercury surveys over mineralization *continued from page 15*

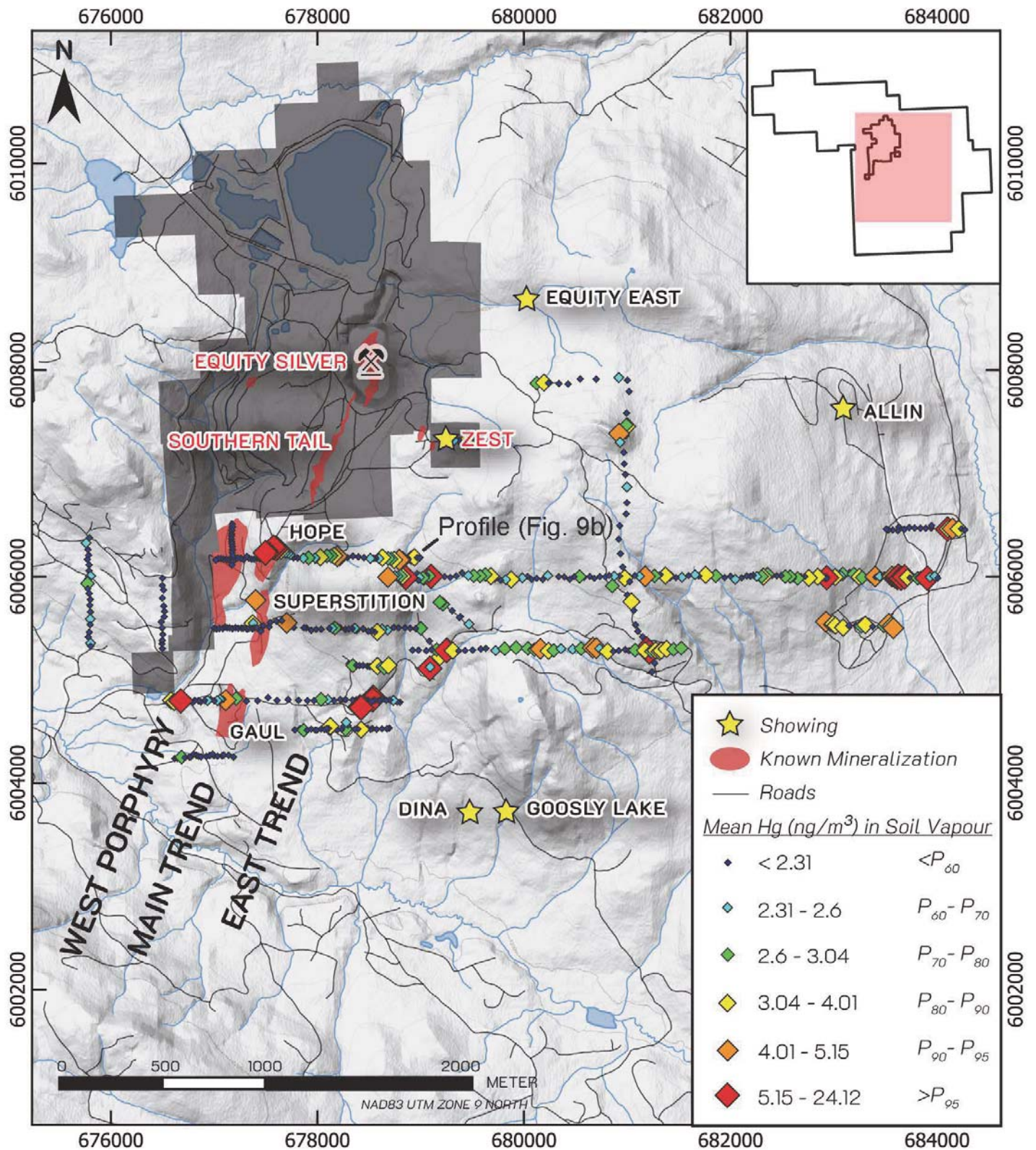


Fig. 9. Silver Hope Cu-Ag-Au occurrence, central British Columbia, Canada. **a)** Mineralized zones, Newmont Corporation's property (grey polygon) that hosts the past-producing Equity Silver and Southern Tail deposits, and air mercury survey results. The "P" values in the legend indicate percentiles. The inset map in the upper right corner shows the Silver Hope property boundary and the map footprint (pink area). Tamburi and Barnes (2024) (Fig. 9b on next page)

New methodology for air mercury surveys over mineralization *continued from page 16*

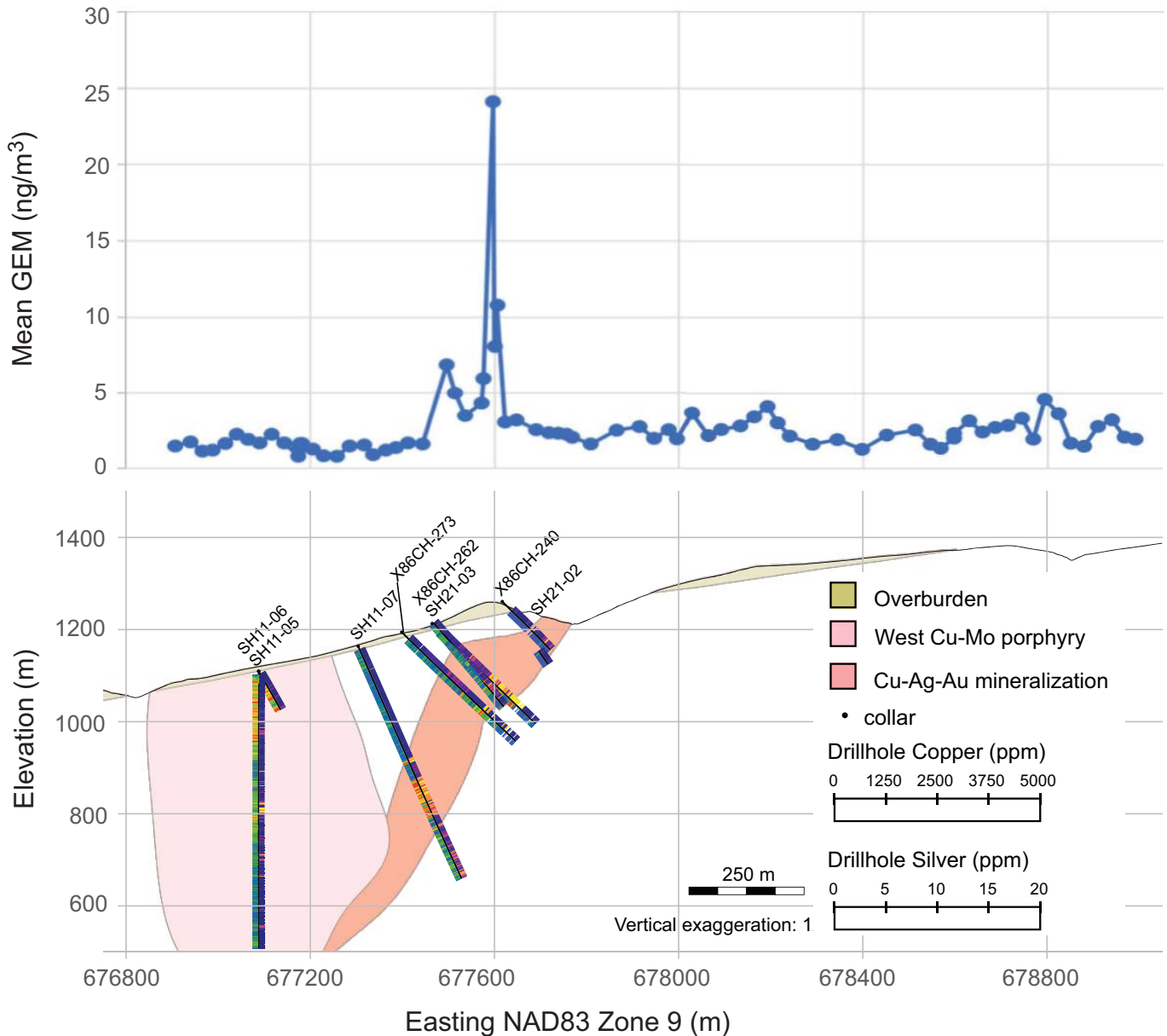


Fig. 9 continued. Silver Hope Cu-Ag-Au occurrence, central British Columbia, Canada. **b)** Average gaseous elemental mercury (GEM) concentrations in air 1 cm above ground and geological cross-section along profile 6006200N.

Rapid air mercury surveys help delineate mineralization under cover in real-time and are thus effective for mineral prospecting in underexplored areas. They can complement other techniques that may be less efficient (e.g. due to extensive transported overburden or the heterogeneous nature of the surface).

ACKNOWLEDGEMENTS

We thank all the partners who have contributed to the research. Lumex Instruments Canada kindly loaned the RA-915M Hg analyzer for the field work. Mercury surveys on Vancouver Island, British Columbia, were supported by the British Columbia Geological Survey (BCGS). Special thanks to the BCGS colleagues L. Ootes and A. Hickin, and summer students M. Golovetsky and B. Coates for their help in the field. Anomalous Exploration Ltd. conducted Hg vapour surveys for Finlay Minerals Ltd. at their Silver Hope property in central British Columbia, Canada. An experimental mercury survey in Kazakhstan in 2022 was supported by Polymetal (M. Morozov), Open Geology (P. Putilov), and Zhana Mys (D. Zhunusov). We very much appreciate the thorough reviews and comments of P.E. Rasmussen (Health Canada) and M.B. McClenaghan (Geological Survey of Canada).

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New methodology for air mercury surveys over mineralization *continued from page 17*

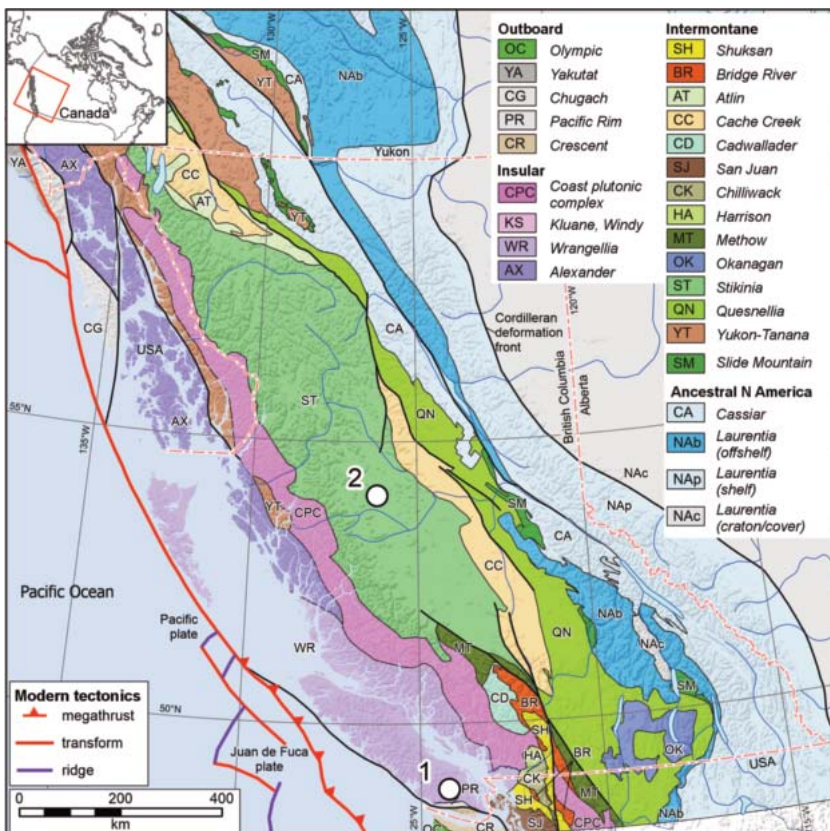
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New methodology for air mercury surveys over mineralization *continued from page 18*

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Erratum



Correction of Figure 7 in *New methodology for air mercury surveys over mineralization* by N.R. Mashyanov, A.S. Rukhlov, S.E. Sholupov, V.V. Ryzhov, A.D. Shashko, E. Perkins and W. Barnes. EXPLORE, 208, p. 13.

In the original article, Figure 7 on page 13 was presented with a portion of the ocean mask reproduced incorrectly. This error may affect the interpretation of the data presented in the figure.

The conclusions of the paper remain unchanged. The Editor sincerely apologizes for this error and any confusion it may have caused.

The online version of the article has been corrected to include this corrected version and is reproduced here for reference..



Fig. 7. Air mercury survey sites at mineral occurrences in British Columbia, Canada: 1 - Lara-Coronation, 2 - Silver Hope. Terranes after Colpron (2020).

