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STIFTELSEN BERGTEKNISK FORSKNING
ROCK ENGINEERING RESEARCH FOUNDATION

METHODOLOGY FOR GEOCHEMICAL SCREENING OF THE BEDROCK – A CASE STUDY OF AN ARSENIC-RICH BASEMENT AT ARLANDA-ROSERSBERG

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BeFo Report 253

Cover photo: Silvery arsenic minerals [arsenopyrite (FeAsS); löllingite ($FeAs_2$)] in a quartz-feldspar matrix in a pegmatite dyke at Arlanda.

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Metodologi för geokemisk kartläggning av berggrund – en fallstudie från arsenikrik berggrund i Arlanda-Rosersbergsområdet

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PREFACE

The potential environmental impact of acidifying or toxic elements in the bedrock has become a major concern for handling aggregates within construction projects.

A general lack of knowledge about the mineral chemistry of the bedrock, and its reactivity when exposed to water and air, challenge exploitation of the bedrock and effective recycling of rock masses in the circular economy. As a first step to reduce this lack of knowledge this study has investigated where, and in what minerals, elements hazardous to health and the environment occur in an arsenic rich bedrock in the Arlanda-Rosersberg area, north of Stockholm with the aim to find a time- and cost-efficient methodology for identification of potentially contaminating rock materials in construction projects to be applied early in the planning process.

One of the findings is that a close correlation between enrichments of arsenic in the bedrock and arsenic contamination of the groundwater confirm that the geochemistry of the groundwater can be a powerful predictive tool for the recognition of high-arsenic bedrock domains. In addition, handheld X-Ray fluorescence measurements at outcrops in the field turned out to be an effective tool for reconnaissance mapping of arsenic enrichments in the bedrock.

The main participants in the study were Per Nysten and Mattias Göransson (SGU), Jenny Andersson (SGU and NRM), Paul Evins (WSP), and Iwona Klonowska and Daniel Buczko (Uppsala University); also a master's thesis (Skoog 2022) and a bachelor's thesis (Nilsson 2024), both at Uppsala University, were carried out in connection with the study.

The study and the associated investigations were funded by BeFo, the Swedish Environmental Protection Agency, the Geological Survey of Sweden (SGU), and the Swedish Museum of Natural History (NRM). The engagement from a reference group consisting of 18 actors, including representatives from the academia, government expert authorities, supervisory authorities, consultants, and industry is acknowledged.

Stockholm

Patrik Vidstrand

FÖRORD

Bygg och anläggningsarbeten i berg med höga halter miljö- och hälsofarliga ämnen kan medföra miljöpåverkan och problem vid masshantering.

Brist på kunskap om berggrundens mineralkemi och dess reaktivitet vid exponering för vatten och luft har skapat en grogrund för oro för omgivningspåverkan vid exploatering av berggrunden. Osäkerhet om bergmaterialens miljöpåverkan kan medföra kostsam översanering medan produktion av ersättningsmaterial leder till ökad exploatering, ökade transporter och riskerar att leda till utdragna tillståndsprocesser och vidare oro i civilsamhället.

Som ett första steg i att reducera denna kunskapsbrist har denna studie undersökt var, hur och i vilken form arsenik och några andra potentiellt försurande och toxiska ämnen förekommer i bergmaterial i syftet att ta fram tids- och kostnadseffektiv metodik för identifiering av potentiellt kontaminerande bergmaterial i bygglinjen. Undersökningarna har fokuserats till bygg- och anläggningsintensiva områden omkring Arlanda flygplats där ställvis höga halter miljö- och hälsofarliga ämnen i bergmaterial resulterat i kostsamma saneringsåtgärder och förseningar i byggprocessen.

Ett av resultaten är att ett nära samband mellan anrikning av arsenik i berggrunden och arsenikförorening av grundvattnet bekräftar att grundvattnets geokemi kan vara ett kraftfullt prediktivt verktyg för igenkänning av högarsenikhaltiga berggrundsdomäner. Undersökningarna visade även att analyser med handburen röntgenfluorescens på berghällar var ett effektivt verktyg för rekognoserande kartläggning av arsenikanrikad berggrund.

Deltagare i projektet var Per Nysten och Mattias Göransson (SGU), Jenny Andersson (SGU och NRM), Paul Evins (WSP) samt Iwona Klonowska och Daniel Buczko (Uppsala universitet). Ett magister- (Klara Skoog) och ett kandidatarbete (Cornelia Nilsson), samtliga Uppsala universitet, har genomförts inom ramen för studien.

Studien och de genomförda undersökningarna har finansierats av BeFo, Naturvårdsverket, SGU och NRM. Engagemanget från en referensgrupp bestående av 18 personer med representanter från universitet, statliga expertmyndigheter, tillsynsmyndigheter, konsulter och ballastproducenter erkänns.

Stockholm

Patrik Vidstrand

SUMMARY

Local high background levels of toxic and acidifying elements in the bedrock have become a concern for handling of rock masses in the construction intensive Stockholm region, Sweden. In this study, we have investigated where and in what minerals elements hazardous to health and the environment occur in an arsenic (As) rich bedrock in the construction-intensive Arlanda-Rosersberg area, Sweden. Targeted sampling of rocks for bulk rock geochemical analysis were guided by handheld X-Ray Fluorescence (HXRF) analysis, and followed up by detailed mineralogical, mineral chemical, and petrographical analyses. Significant enrichments of As of >100-1000 ppm, locally up to near 1 wt%, were encountered in a ca 1.3 km wide north-northeast trending zone inside a metasupracrustal belt in the south-eastern Arlanda area. Such high As concentrations are highly anomalous for a non-mineralised quartz-feldspathic bedrock. No other elements showed extreme enrichments. High As concentrations, >100 ppm, occurred in metasedimentary, preferentially mica-rich units, in intermediate- to mafic sills and dykes, and in fractionated tourmaline bearing pegmatites. In many places, tourmaline bearing pegmatites were spatially associated with As-enrichments in the bedrock and may be tracers for high-As units. Granitoid rocks were low risk lithologies.

Arsenic was hosted in arsenopyrite (FeAsS) and arsenide (löllingite, $FeAs_2$) and encountered in all enriched lithologies. Such widespread occurrence of arsenide has not previously been documented and indicates that the presence of arsenide in quartz-feldspathic rocks has been hugely underestimated. Pyrrhotite ($Fe_{(1-x)}S$) was the most common sulphide. Pyrite (FeS_2) was more common in granitic rocks and in structurally young discrete low temperature deformation zones.

Reconnaissance geochemical mapping of outcrops in the field was an effective method for initial tracing of high-As units, suitable for strategic selection of samples for whole rock analysis, and petrographic and mineralogical investigations. Successful application of the method, however, depends on geological expertise and high frequency of HXRF measurements that includes all lithologies. Arsenic is, however, a highly versatile element and the strategy for surveying the bedrock depends on the geological setting.

A close correlation between enrichments of As in the bedrock and As contamination of the groundwater confirm that the geochemistry of the groundwater can be a powerful predictive tool for the recognition of high-As bedrock domains.

Keywords: arsenic, bedrock, aggregates, handheld X-ray fluorescence analyses, lithogeochemistry, löllingite, arsenopyrite

SAMMANFATTNING

Naturliga anrikningar av potentiellt toxiska och försurande ämnen i berggrunden skapar oro för omgivningspåverkan vid exploatering av berggrunden i den bygg- och anläggningsintensiva Stockholmsregionen. I denna studie har arsenikanrikad berggrund i Arlanda-Rosersbergsområdet undersökts i syfte att utveckla en tids- och långsiktigt kostnadseffektiv metodik för geokemisk kartläggning av berggrunden. Rekognoserande kartläggning med handburen röntgenfluorescensanalys (HXRF) var en effektiv metod för att spåra och geografiskt avgränsa arsenikanrikade zoner och som stöd för strategisk provtagning för totalgeokemisk, petrografisk och mineralogisk analys. Arsenikhalter på mellan 100 och 1000 ppm, lokalt upp till nästan 1 vikt%, kunde påvisas i en cirka 1,3 km bred zon i ett ytbergartsbälte i sydöstra Arlandaområdet. Inga andra ämnen uppvisade extrema anrikningar. Den kraftigt arsenikanrikade berggrunden utgjordes av metasedimentära bergarter, intermediära till mafiska magmatiska bergarter och turmalinförande pegmatiter. Granitoida bergarter hade genomgående låga arsenikhalter. Turmalinförande pegmatiter var på många ställen rumsligt associerade med arsenikanrikningar i sidoberget och kan vara en indikatorbergart för arsenikanrikad berggrund. Arsenikanrikningar liknande de upptäcks i Arlandaområdet är inte tidigare kända i det icke-mineraliserade urberget.

Arsenik förekom som arsenopyrit (FeAsS) och som arsenid (löllingit, $FeAs_2$). Löllingit påträffades i samtliga anrikade litologier. Utbredd förekomst av arsenid i icke mineraliserad berggrund har inte tidigare dokumenterats vilket indikerar att förekomst av arsenid kan ha förbisetts. Bland sulfider i berget var magnetkis ($Fe_{(1-x)}S$) vanligast förekommande, förutom i granitiska bergarter och i strukturellt unga, diskreta lågtempererade deformationszoner där pyrit (FeS_2) dominerade. Inga pyritförekomster innehöll mätbara halter arsenik.

Framgångsrik tillämpning av rekognoscerande HXRF mätningar är beroende av geologisk expertis och en hög frekvens av mätningar som inkluderar alla litologier. Arsenik kan förekomma i många olika former och uppträder därför på olika sätt i olika geologiska miljöer. Strategin för kartläggning av berggrunden måste därför anpassas till den geologiska miljön.

Ett nära samband mellan anrikningar av As i berggrunden och As-föroreningar i grundvattnet bekräftar att grundvattengeokemin kan vara ett kraftfullt verktyg för att hitta områden med arsenikanrikad berggrund

Nyckelord: arsenik, berggrund, ballast, handburen röntgenfluorescenseanalys, litogeokemi, arsenopyrit, löllingit

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1. INTRODUCTION

Parts of the Swedish bedrock include mineralisations that contribute to naturally elevated background levels of elements that may contaminate the environment if leached and spread at harmful concentrations. Natural enrichments of sulphides may cause acidification of the environment and high concentrations of heavy metals (e.g., lead, nickel, copper), radioactive elements (uranium, thorium) or metalloids such as arsenic (As) may contaminate surface- and groundwater (cf. Fig. 1; Maxe 2021). Also, low-grade mineralisations (non-mineralised bedrock), i.e., mineralisations at concentrations too low to be of an economical interest have the potential to cause high background levels of harmful elements. These are, however, as a rule, poorly investigated due to their insignificant economic value. A poor understanding of the contamination potential of low-grade mineralisations in the bedrock has become a problem in aggregate production and for handling rock masses in construction and tunnelling projects. It hampers effective incorporation of construction and tunnelling waste in a circular economy, and result in costly dumping of rock masses, increased production of virgin aggregates, and increased transports. The problem is further accentuated as the lack of understanding of the contamination potential of rocks hampers governmental authorities in their efforts related to environmental monitoring, guidance for the handling of rock masses and permit processes. As a result, construction projects are facing huge costs for surveying, remediation efforts and delays. The potential problems with elements hazardous to health and the environment leaching from rock is now pointed out as a prioritized area for investigations by the Toxicological Council (2022).

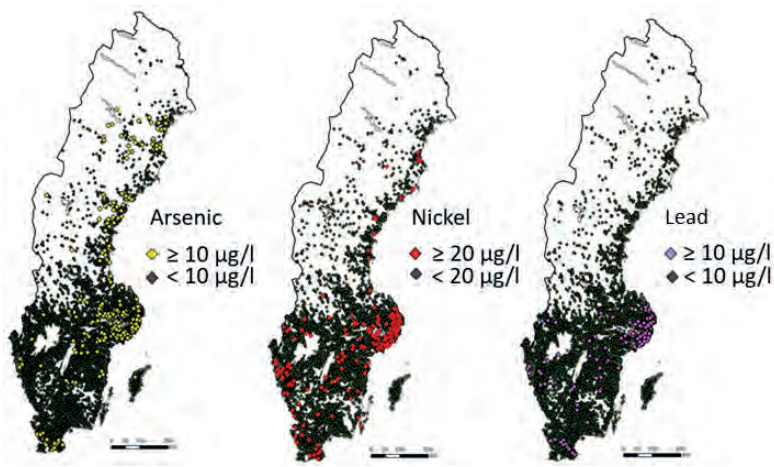

Figure 1. *Geochemistry of unfiltered raw water from tubed wells penetrating the bedrock. Data from Maxe (2021). Green symbols = safe drinking water; yellow, red and purple symbols denote geogenic contamination of groundwater at levels unsuitable for drinking according to the recommendations by Swedish Food Agency (2024) for households exceeding 50 users.*

1.1 The bedrock as a primary source of arsenic contamination

Arsenic (As) is a well-known toxic and carcinogen element that in places occur at high background levels in the bedrock from which it may be mobilized and spread to the environment by natural processes, anthropogenic activity, or climate change (e.g., Yang et al. 2012; Bretzler et al. 2017; Masuda 2018). Sulphides in mineralised rocks and volcanic settings are identified as the primary source of As, and mining operations as a main source of contamination (e.g., Smedley and Kinniburgh 2002; Ravenscroft et al. 2011; Hug et al. 2020). Oxidation of arsenopyrite (FeAsS) and pyrite (FeS_2) is regarded as the principal source of As (e.g., Bowell et al., 2014; Mukherjee et al., 2024). Dissolution of As from the bedrock takes place in several steps. Oxidation of primary As-bearing minerals leads to formation of arsenite [As(III)] and arsenates [As(V)], compounds that are mobile in the hydrosphere (Hug et al. 2020). Secondary hydrous As-minerals can be long transported to sedimentary aquifers, glacial deposits or deposited in deformation zone systems (Hatakka et al. 2014; Pedretti et al. 2019). In quarries and at construction sites rocks are crushed to fine fractions which may significantly increase the potential of As-leaching due to the increase of mineral surfaces that are exposed to weathering (Backman et al. 2006; Parviainen et al. 2015).

In Sweden, high concentrations of As in the bedrock accompanied by contamination of the hydrosphere occur in the Skellefteå and Bergslagen ore districts and in areas with alum shales of the Lower Palaeozoic platform cover (Hildebrand et al. 2009). High-As rocks also occur in Mälardalen and Västernorrland (op cit.), but due to the absence of an economic interest in these rocks, their geology is not well known. Studies of As-contamination of groundwater in these areas conclude that the source of As is likely to be As-rich sulphides in the bedrock, but clear relationships with a specific mineralogy, texture and rock type are not addressed (Fig. 2; Thunholm et al. 2009; Bhattacharya et al. 2010).

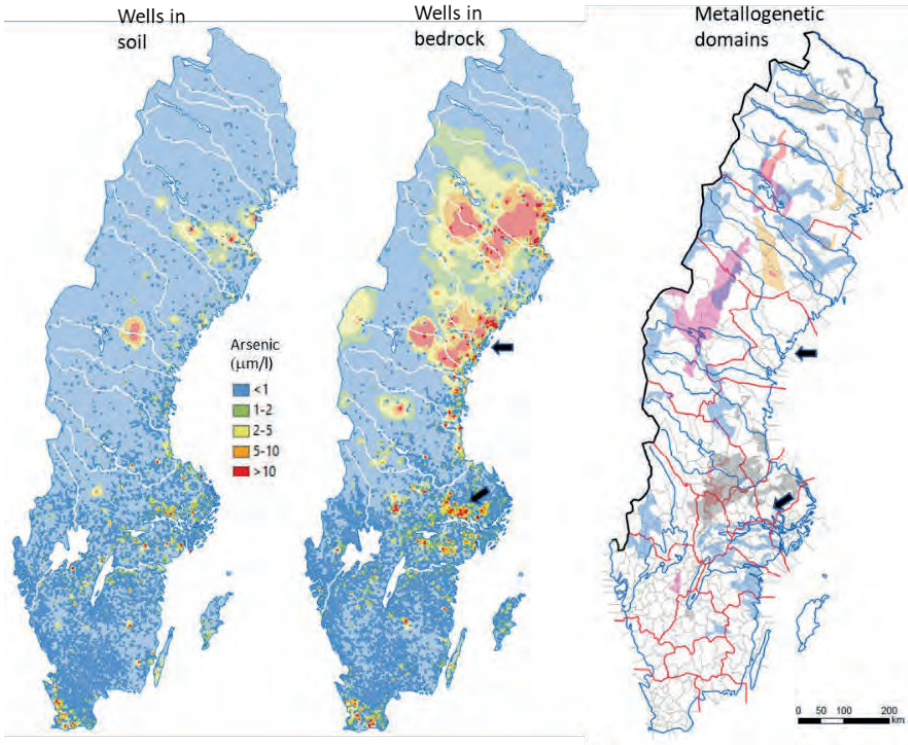

Figure 2. *As-geochemistry of unfiltered raw water in tubed wells (private wells) in soil (A) and in the bedrock (B). Data from Maxe (2021). Map made by interpolation, Inverse Distance Weighting, (IDW). Areas with more than 3 km distance to the nearest sample point (well) are shown in faded colour. Legend follows thresholds for the concentration of As in potable water at 10 µg/l following the recommendations given by Swedish Food Agency (2024), for households exceeding 50 users. (C) Metallogenetic domain map from the SGU ore and minerals map data base as of June 2024.*

1.2 Geogenic arsenic contamination in the Arlanda-Rosersberg area

At present, fear for As-contamination related to exploitation of the bedrock in construction intensive parts of the Stockholm region have set focus on the bedrock as a primary source for As. The wake-up call was the discovery of 500 ppm As in crushed rock aggregates in a sandbox at a day-care in Sigtuna (Länsstyrelsen Stockholm 2019). The material was produced at Arlanda where several construction projects have encountered problems with high concentrations of As in the bedrock (e.g., Law and Jakobsson 2014, Svea Hovrätt 2021; Swedavia 2022). Arsenic contamination of freshwater in neighbouring low background areas caused by the use of rock masses

from Arlanda has also been reported (Sigtuna kommun 2021). The high concentrations of As in the bedrock has led to costly delays of construction and remediation actions and is a major challenge for construction and building activities (e.g., Bidros 2014; Law and Jakobsson 2014).

1.3 Scope of study

The goal of this study has been to pin down *where*, *how*, and *in what form* elements hazardous to health and the environment, and in particular As, occur at high concentrations in the bedrock in a construction intensive urban part of central Sweden with high demands for construction material. The aim has been to integrate this information to develop a methodology for effective prediction of the enrichment of toxic elements in the bedrock at construction sites and with special focus on As.

The study area is located in the construction intensive areas around the Stockholm Arlanda airport (Fig. 3). With its location between the large cities of Stockholm and Uppsala, including the key Nordic logistics centre around the airport, it is one of the most expansive regions in northern Europe, and in focus for intensive construction work in the forthcoming decades.

The investigations of the bedrock have included targeted sampling of rocks guided by handheld X-Ray Fluorescence (HXRF) analysis, followed up by detailed mineralogical, geochemical and petrographical analyses.

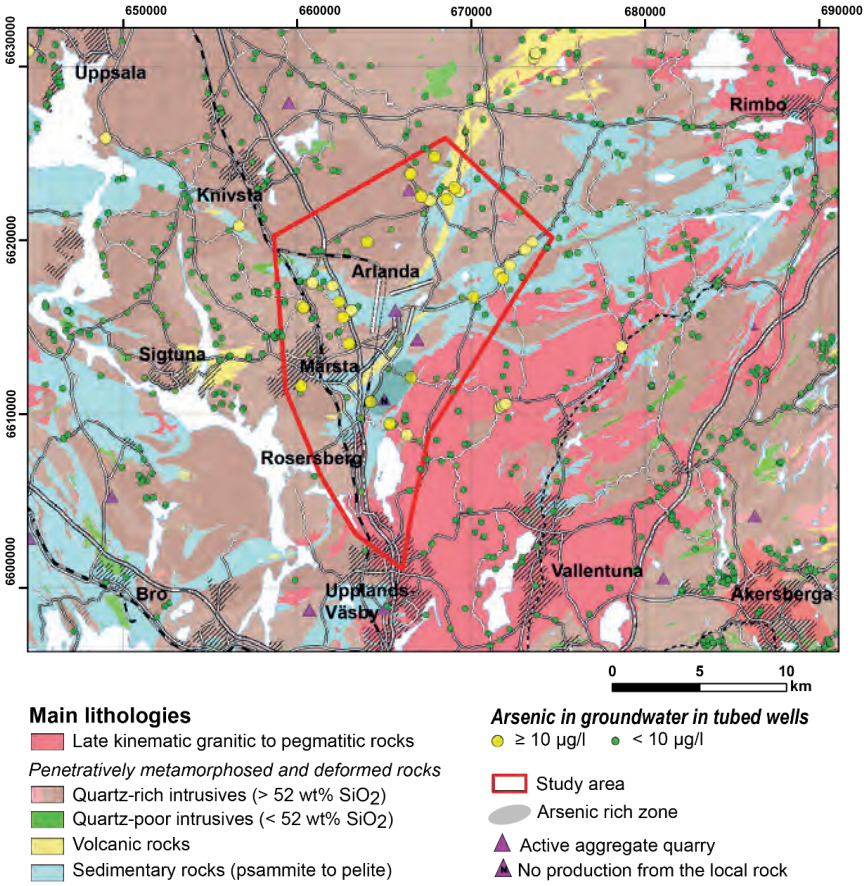

Figure 3. *Bedrock map of the Arlanda-Rosersberg area with study area marked as a red polygon. Zone with high-As enrichments in the bedrock is shown in shaded grey. Bedrock map based on the SGU bedrock map database at scale 1:50 000. Arsenic geochemistry of unfiltered raw water from Maxe (2021). Reference value for As concentration in potable water for large drinking supplies (>50 persons) after Swedish Food Agency (2024).*

Disclaimer

This study is focused on the primary source of harmful elements in the bedrock. Aspects on weathering, leaching and mobility of elements to the environment, and the influence from microbiological activity is beyond the scope of this research. In this study, the contamination potential of the bedrock refers only to the primary concentration of elements in the bedrock, i.e., the primary source of the elements.

1.4 Reference values for arsenic in the bedrock

In this study, low concentrations of As are referred to as concentrations of As that fall below 10 ppm, which is the reference value considered by the Swedish Environmental Protection Agency to entail less than a minor risk for potential contamination when rock masses are recycled for use in constructions (Swedish Environmental Protection Agency 2010). This guideline value is based on the concentration of As in Swedish soil which has a 90-percentile of 10 ppm (based on 12,815 soil analyses available in the SGU data base year 2007). The corresponding value for less sensitive land use is 25 ppm. The 90-percentile for the Swedish bedrock gives a value of 5.5 ppm As based on 4910 bulk rock geochemical analyses of rocks classified as non-mineralised in the lithogeochemical database of SGU [data used by Miškovský et al. (2020)].

2. GEOLOGICAL SETTING OF THE ARLANDA AREA

The bedrock in the Arlanda area constitutes a strongly deformed and folded NNE trending steeply dipping ca 5-10 km wide metasupracrustal belt intruded by at least two generations of granitoids (Fig. 3; plutonic rocks and sub-volcanic intrusives; Stålhös 1991). The supracrustal complex and the early intrusive rocks have been deformed and metamorphosed at amphibolite facies conditions. Geothermobarometry coupled with phase equilibrium modelling indicate that the amphibolite facies metamorphism reached pressures of 0.45-0.55 GPa and temperatures of 550-600 °C (Sjöström and Bergman 1998; Skoog 2022; Andersson et al. 2024b). Amphibolitized mafic sills and dykes are common in the sedimentary units but have not been observed in the sub-volcanic intrusives and the plutonic rocks. It is unclear whether the amphibolites belong to one or multiple generations of mafic intrusions. Structurally younger granites (Vallentuna granite), pegmatitic granites, and pegmatites dominate the bedrock in the south-eastern part of the area (Fig. 3). Pegmatite in the form of lenses, dikes, and smaller intrusions are common throughout the area. Some pegmatites have a high content of the boron-bearing mineral tourmaline. Structurally young granites have also been observed in the central parts of the area, not shown on the bedrock map of Stålhös (1972). The structurally younger granites and pegmatites lack the penetrative amphibolite facies deformation seen in the older supracrustal and intrusive rocks.

The Arlanda area forms part of the Bergslagen lithotectonic domain (cf. Stephens and Jansson, 2020). The supracrustal complex at Arlanda, however, is lithologically distinct from the characteristic Bergslagen supracrustal deposits. The volcanic rocks have a dacitic to andesitic composition, and quartz-rich porphyries with granitic (rhyolitic) composition, typical of Bergslagen volcanism, have not been observed. The sedimentary rocks are also poorer in quartz, and horizons with limestone are absent. A few occurrences of skarn are indicated in the bedrock map of Stålhös (1972) but not encountered by mapping in this study. The area lacks known sulphide mineralisations characteristic of neighbouring regions in Bergslagen, and there are no records of ancient mines. The principal rock units composing the bedrock in the Arlanda-Rosersberg area are described below. In the description of lithologies below, all rocks that have been deformed and metamorphosed have the prefix “meta“ to denote a metamorphic overprint.

Metasedimentary rocks (≥1.91-1.90 Ga)

The Arlanda Supracrustal Complex is dominated by rocks of a sedimentary origin (psammite to pelite), but with intercalations of volcanic rocks, typically less than 1 meter wide, and bands, dikes, and lenses of mafic and intermediate intrusive rocks, aplites, pegmatites, and pegmatitic granites (Fig. 4A). The sedimentary rocks occur as fine-grained rocks, with a banding defined by variations in the proportions of mica (biotite and/or muscovite), quartz and feldspar (Fig. 4B). Mica-rich zones occur as mica schists, occasionally with a high proportion of cordierite. Cordierite-rich zones are common along the contact to the central volcanic unit (Fig. 4C). In places, these zones also expose metasedimentary rocks with 0.5-2 cm large andalusite porphyroclasts (Fig. 4D). The mica-rich metasedimentary rocks are schists composed of biotite, plagioclase, quartz and muscovite with accessory ilmenite and apatite (Fig. 4E). Muscovite- and quartz-rich metasedimentary rocks often contain tourmaline. The tourmaline-bearing metasedimentary rocks are always muscovite bearing (Fig 4F).

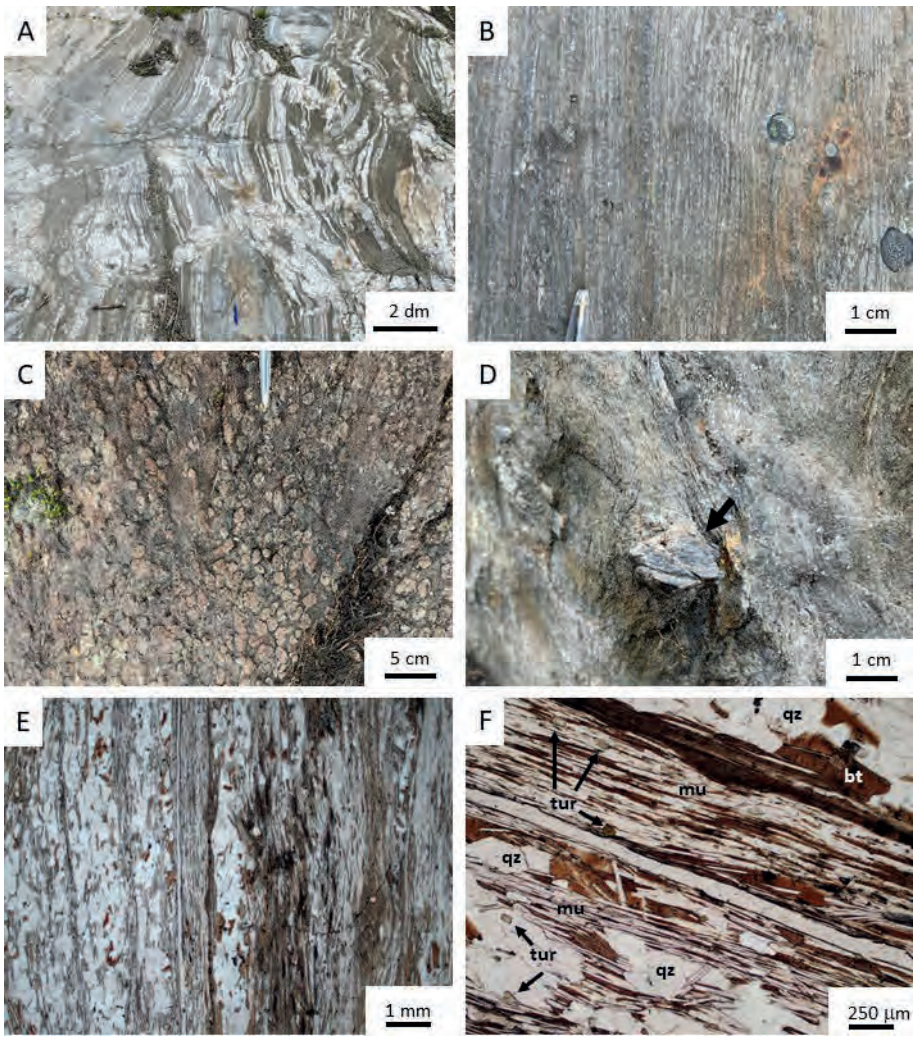

Figure 4. *Metasedimentary rocks in the Arlanda area. A) Field photo of lithololgically banded metasedimentary rocks injected and in places cut by tourmaline-rich pegmatitic veins and dykes. Skånela, northern shore of Fysingen. B) Banded metasedimentary rock close to runway 3 at Arlanda. C) Cordierite blastesis in a metasedimentary rock at the contact to a sub-volcanic metaintrusion, Good Morning hotels, Arlanda. D) Andalusite megacryst (reddish brown marked by arrow) at Nybygget, Arlanda. E-F) Photomicrographs from optical microscope (parallel nicols) of thin sections of muscovite and tourmaline-rich As-enriched (50 ppm As) micaschist at Laggatorp. Abbreviations: qz-quartz, mu-muscovite, bt-biotite, tur-tourmaline.*

Volcanic Rocks (1.91-1.90 Ga)

The main NNE-trending volcanic unit in the central Arlanda area (cf. Fig. 3), consists of fine- to medium grained grey to dark grey volcanic (extrusive) to subvolcanic (intrusive) rocks, in places with plagioclase or plagioclase-amphibole porphyritic textures (Fig. 5A and B). The composition of the volcanic rocks ranges from dacite to andesite (granodiorite to tonalite). They are composed of plagioclase, biotite, quartz and amphibole with accessory ilmenite, apatite, zircon, opaques (Fig. 5C) and occasional garnet (Fig. 5E). They are relatively poor in quartz and K-feldspar. Lenses (enclaves) and schlieren of darker or lighter composition are common (Fig. 5B). In places, banded sections with variations in colour and grain size occur reflecting variations in the proportions of plagioclase and biotite, and locally, amphibole. The dominant ore minerals are pyrrhotite and pyrite. In places, rare concentrations of pyrite occur together with K-feldspar, albite and chlorite in 2–5 mm wide discrete deformation zones with a bleach margin to the side rock (Fig. 5D).

The main volcanic unit in places also contains garnet (Fig. 5E; almandine garnet; cf. Skoog, 2022). They are about 3–10 mm in size and occur sporadically in the rock. Garnet- and/or cordierite and/or andalusite-bearing schists are found along the contact between the main volcanic unit and surrounding sedimentary rocks (Fig. 4D, 5E and F).

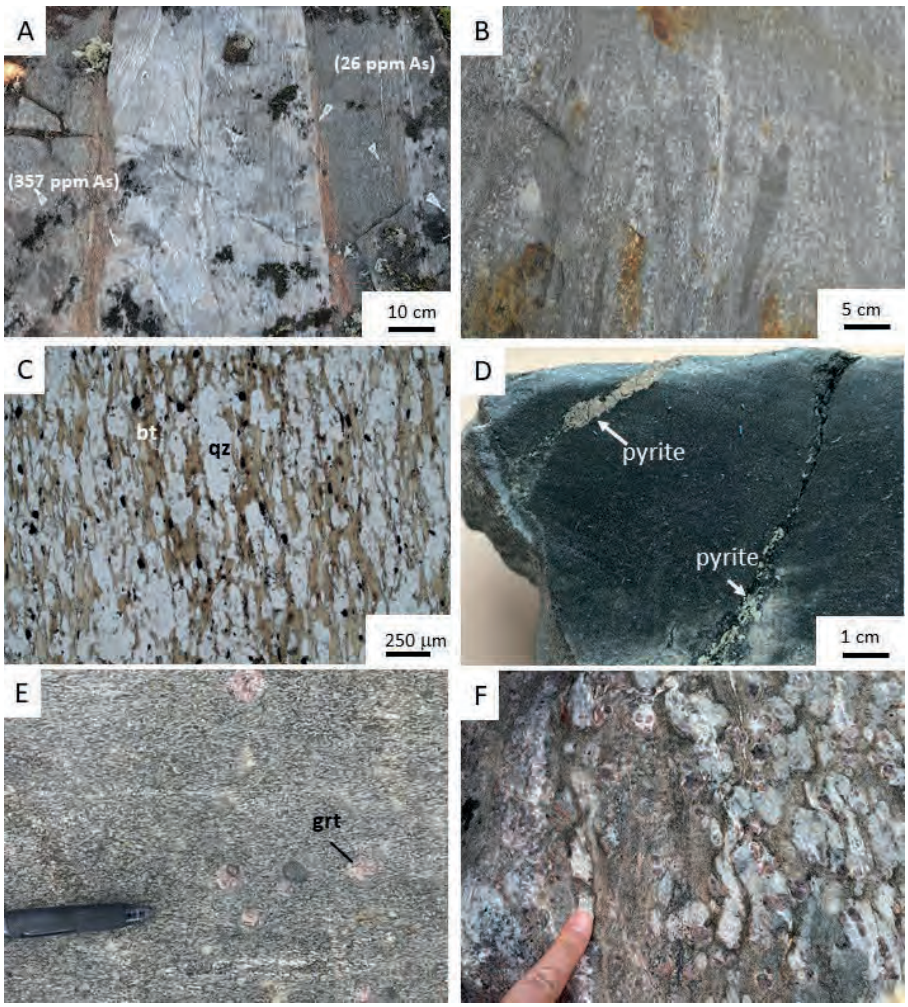

Figure 5. *Volcanic and sub-volcanic rocks at Arlanda of andesitic to dacitic composition. A) Plagioclase-amphibole porphyritic volcanic sills in low-angle discordant relation to the lithological banding and veining in a metasedimentary rock (Västerbytorp quarry). B) Plagioclase porphyritic sub-volcanic rock with schlieren and enclaves of mafic composition. C) Characteristic texture and mineralogy of intermediate volcanic rocks at Arlanda composed of plagioclase, biotite (bt), quartz (qz) and ilmenite (opaque mineral). D) Pyrite in discrete semi-brittle fractures in mafic volcanic rock (Västerbytorp). E) Garnet (grt; pale pink) bearing dacitic sub-volcanic rock (Fire station Arlanda). F) Garnet and cordierite (severely altered) rich metasedimentary rock in a megaxenolith inside the central volcanic unit at Arlanda (Benstocken).*

Early quartz-poor mafic intrusives (1.91-1.87 Ga)

Pervasively amphibolitized dykes, sills and larger bodies several meters across of gabbroic to dioritic composition are intercalated in the metasedimentary complex (Fig. 6A and B). The metagabbro occurs as fine- to medium grained isotropic to weakly foliated rocks composed of amphibole, plagioclase and ilmenite and accessory apatite (Fig. 6B-D). Some amphibolites have four different amphiboles: light coloured grunerite, iron and magnesium pargasitic hornblende and tschermakite (Fig. 6C). Some samples preserve a relict igneous texture (Fig. 6D).

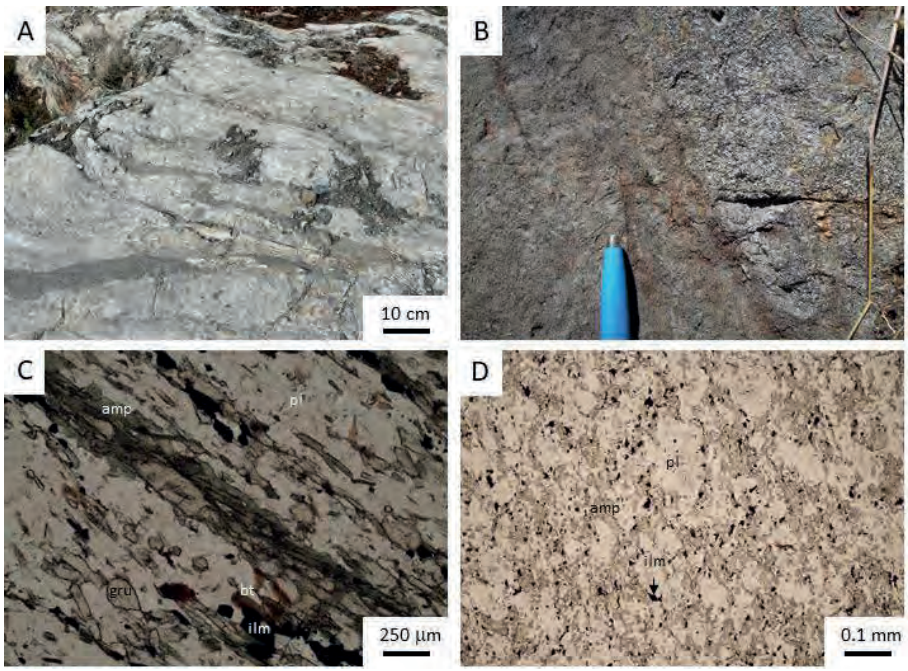

Figure 6. *A) Folded amphibolite dyke in the metasedimentary complex (Västerbytorp quarry, JAN220002). B) Two varieties of medium grained metagabbro that differ in texture and plagioclase content (road cut, Kimsta road). C-D) Thin section photomicrographs (parallel nicols) showing amphibolite enriched in As with dark and light-coloured amphiboles (C; Sample JAN220010C, Kimsta road) and with relict igneous texture (D, sample JAN220010E, 191 ppm As). Abbreviations: amp-amphibole, bt-biotite, ilm-ilmenite, gru-grunerite, pl-plagioclase.*

Early felsic intrusive rocks (1.90-1.87 Ga)

The early granitoid intrusive rocks are pervasively deformed, grey to light grey, medium-grained predominantly equigranular granitic to granodioritic rocks in places with mafic enclaves (Fig. 7A and B). They are composed of plagioclase, quartz and biotite with accessory ilmenite, apatite, zircon, and chlorite (Fig. 7C). Occasionally, they contain fine grained banded xenoliths of the surrounding volcanic rocks (Fig. 7D). Dominating ore minerals are ilmenite, pyrrhotite, and pyrite.

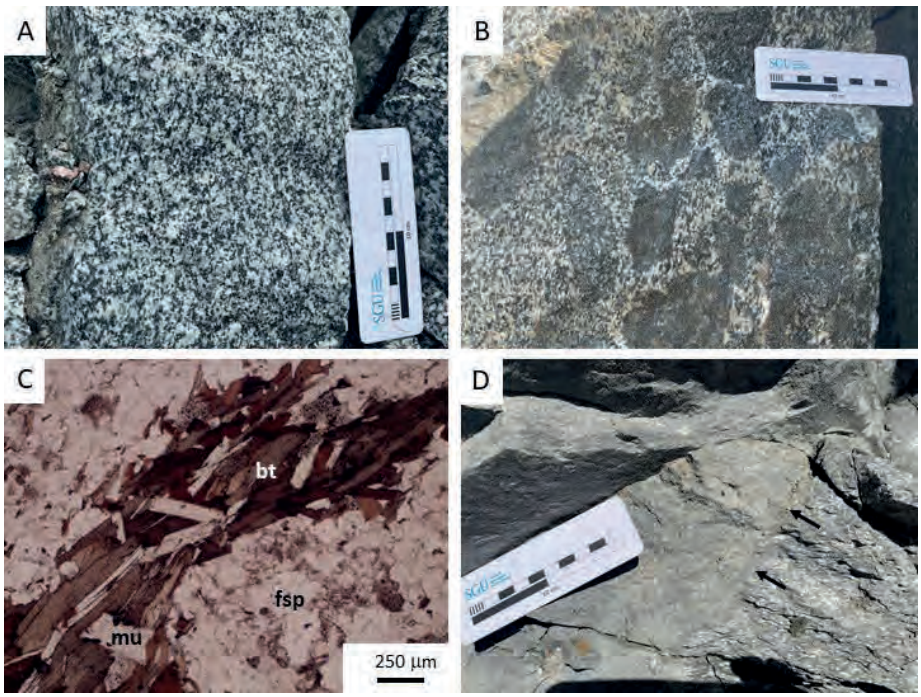

Figure 7*. A) Characteristic field appearance of arsenic-free Uppsala type granite. B) Enclave bearing variety of As-free Uppsala granite. C) Photomicrograph (optical microscope, parallel nicols) of two-mica bearing Uppsala granite. D) Fine grained mafic xenoliths enriched in As (25 ppm) in Uppsala granite low in As (4 ppm). Arrows denote contact between granite and xenolith. Abbreviations: tur-tourmaline, mu-muscovite, bt-biotite, fsp-feldspar.*

Younger Granitic and Pegmatitic Intrusive Rocks (1.85-1.80 Ga)

Late kinematic granite spatially associated with pegmatite occurs in the south-eastern part of the study area (Fig. 3 and 8A). It is part of a large granite to pegmatite granite complex exposed in the northern Stockholm area, the so-called Vallentuna and Stockholm granites (Fig. 3; Johansson and Stephens 2017; Johansson 2019). The presence of structurally younger granite has previously not been documented in the central airport area (cf. Stålhös 1972). During the sampling campaign of this study, however, a weakly foliated, structurally young, biotite- and garnet-rich granitic intrusion was found in a quarry in the central Arlanda area (Fig. 8B; Nysten 2022; Nysten and Klonowska 2024). It is possible that there are more occurrences of younger granite than those detected in previous mapping.

In contrast, several generations of structurally young pegmatites and granitic pegmatites are common in the bedrock near the airport. They form thin dikes (on the scale of decimeters to meters) and larger masses, often in the form of round, protruding lenses, 1-100 square meters in size (Fig. 8C). They have sharp contacts to the surrounding rocks and in places cross-cut early deformation structures at high-angles. The pegmatites vary from primitive so-called ceramic pegmatites (dominated by quartz, feldspar, and muscovite) to moderately differentiated pegmatites rich in black tourmaline, green apatite, and red garnet. The tourmaline rich pegmatites are typically characterised by light grey K-feldspar and with accessory minerals including beryl, columbite, and locally arsenopyrite/löllingite (Fig. 8D-E). In places, a ca 5-10 cm wide recrystallized contact zone to side rock to tourmaline rich pegmatitic veins are enriched in tourmaline (Fig. 8F). The chemically and mineralogically most complex dike found at Arlanda was encountered during the construction of runway 3, but is now inaccessible (Nysten and Jonsson 1998; Weisz 2012). A spatial correlation between the distribution of mineralogically different pegmatite dikes and the structurally young granites can be observed, where the most primitive dikes occur closer to the main granite massif while the more chemically evolved (fractionated) dikes occur farther away (cf. Černý 1991).

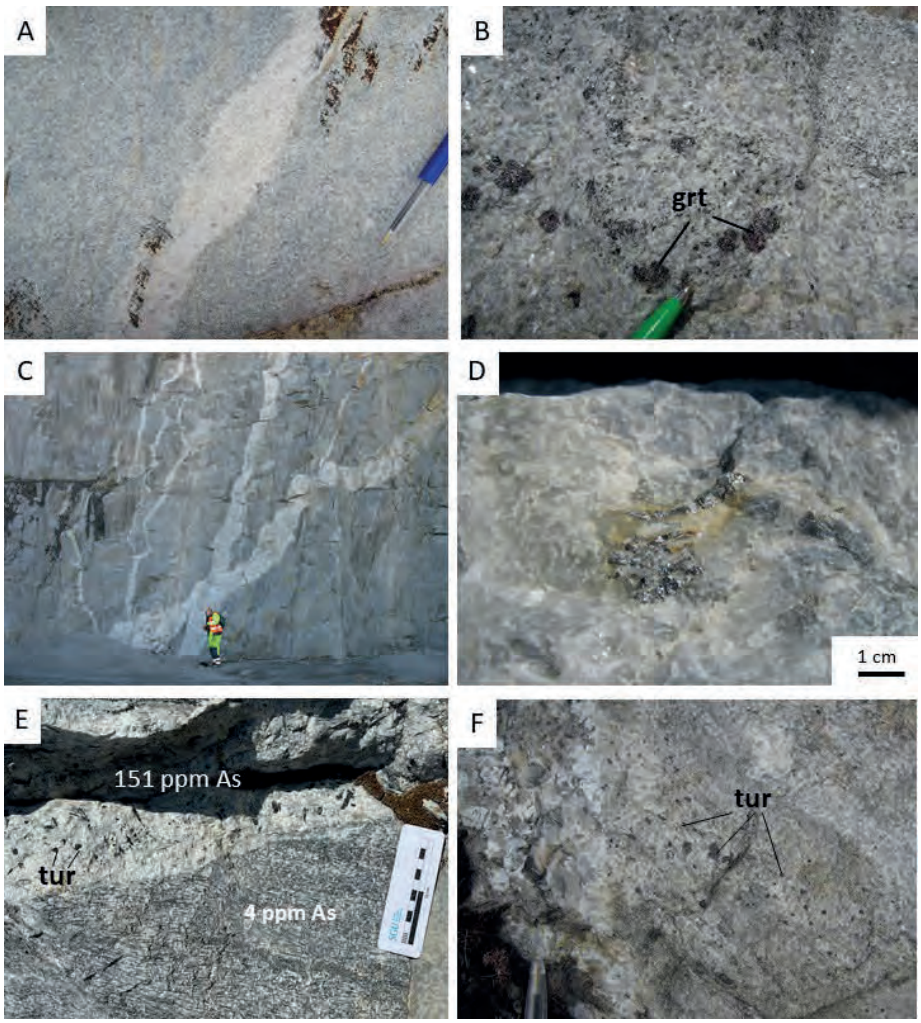

Figure 8. *Low As late kinematic granites and As-enriched pegmatites. A) Leucocratic weakly lineated pegmatite bearing granite at Kimsta (sample PNY230022A). B) Low As garnet-biotite granite at Långåsen (sample JAN210052B). C) Several generations of pegmatites enriched in As. D) close up of As-minerals (arsenopyrite/löllingite) in pegmatite dyke shown in B. E) Tourmaline-rich (black mineral) low angle discordant pegmatite-aplite dyke in penetratively schistose Uppsala granite (Airside Arlanda, Långåsen). F) Metasedimentary rock (to the right) with tourmaline rich contact to tourmaline and muscovite rich pegmatite (Västerbytorp, locality JAN220002). Abbreviations: tur-tourmaline, grt-garnet.*

Age and Metamorphic History of the Bedrock

At present there are no published radiometric age data for the bedrock in the Arlanda area. Based on field relationships, the sedimentary rocks are considered to be the oldest rocks. In the Västerbytorp area, a plagioclase and amphibole porphyritic metaandesite is observed to cross-cut a banding and ca 0.5 cm wide feldspar-rich layer parallel veins in a metasedimentary rock (Fig. 5A). The volcanic rocks are assumed to be of the same age as volcanic rocks in other parts of Bergslagen, dated at 1.91–1.89 Ga, while the early intrusions are dated at 1.91–1.87 Ga (Stephens and Jansson 2020). The youngest granites and pegmatites in the nearby Stockholm area have been dated at around 1.80–1.78 Ga (Öhlander and Romer 1996; Johansson 2019).

During the mapping of the Arlanda area, Stålhös (1972) identified three metamorphic phases: The oldest phase includes localized partial melting (veining) in the supracrustal rocks near the contacts to the ≥ 1.87 Ga early intrusions. It was followed by a regional phase of deformation and metamorphism resulting in penetrative deformation structures in the supracrustal rocks and the early intrusions. The final metamorphic phase included localized melting (veining) of the supracrustal rocks near the younger granite-pegmatite intrusions. Direct (absolute) age determinations for the metamorphic evolution of the Arlanda area are lacking. U-Pb zircon ages at about 1.87–1.86 Ga attributed to the regional metamorphic event have been obtained from metasedimentary rocks to the east (Bergby) and to the west (Odensala) of the Arlanda area (Andersson et al. 2006; Johansson and Stephens 2017). A pre-1.86 Ga age for the main tectonometamorphic event is supported by an Ar-Ar hornblende cooling age of 1.85 Ga in a metagabbro east of Knivsta (Page et al. 2007).

3. METHODS

The areal extent of the study area was defined according to the distribution of As-contamination of groundwater in tubed wells penetrating the bedrock (cf. Fig. 3). The groundwater chemistry data used is from Maxe (2021). The classification of contamination based on the reference value of 10 µg/l As after the recommendations for maximum As content in large drinking water supplies (> 50 persons) as defined by the Swedish Food Agency (2024). A link between the As-geochemistry of the bedrock and contamination of groundwater had previously been suggested in studies of the groundwater geochemistry in Sigtuna municipality (Thunholm et al. 2009), and supported by SGU reconnaissance lithogeochemical mapping in the area 2020.

The field data, petrography and whole rock geochemical data used in this study also include data from Skoog (2022) and two geochemical mapping projects carried out by SGU: reconnaissance mapping in parts of the Sigtuna municipality with problems with As-contamination of groundwater in 2020 and a geochemical mapping campaign focused on high-As rocks at Arlanda in 2023 carried out in collaboration with the Swedish Environmental Protection Agency (Andersson et al. in press). These projects followed the same methodological protocol as described below, apart from Skoog (2022) and the SGU project in 2020 that did not include HXRF analyses.

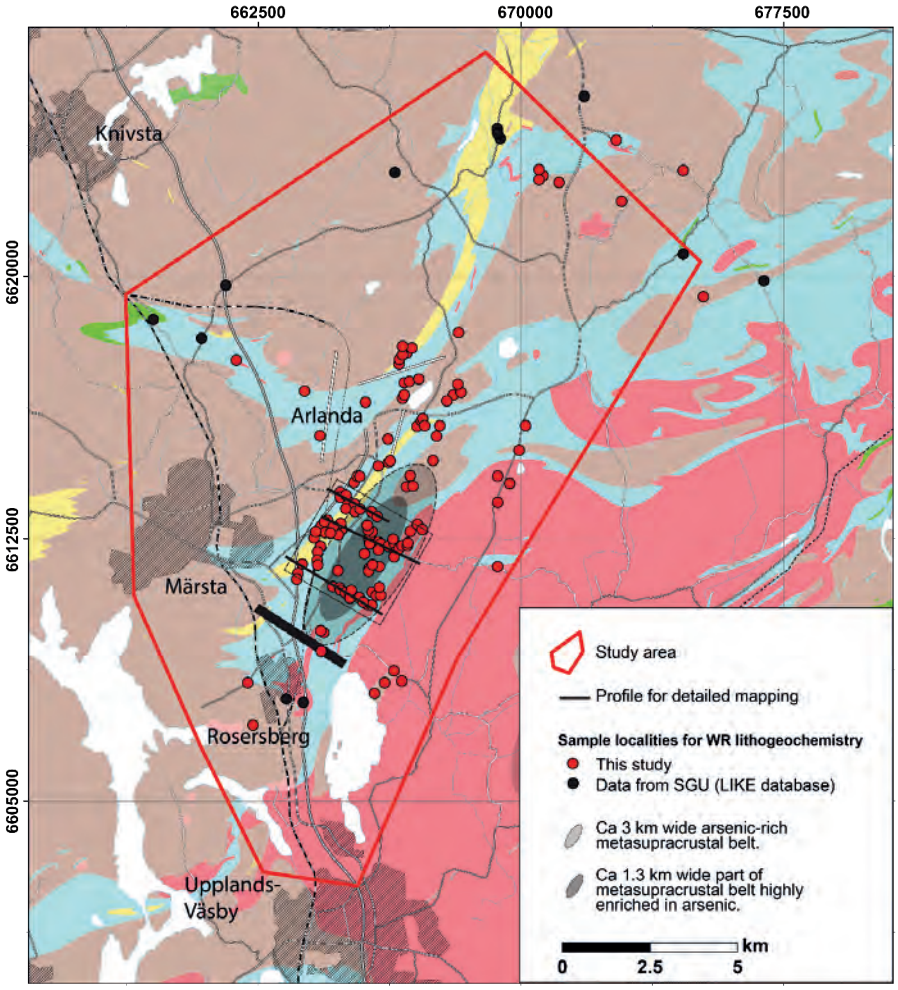

Figure 9. *Bedrock map with sample localities for whole rock geochemical analyses and location of the study area with profiles for detailed mapping of As in the bedrock indicated. Legend of bedrock map follows Figure 3.*

3.1 Field work and sampling

The field work was initiated by reconnaissance geochemical mapping including HXRF and selected WR analysis of representative rock units with a geographical cover of the study area. The early results showed that enrichment of As was concentrated in a more than ca 3 km wide NNE-trending zone in the central parts of the south-eastern metasupracrustal belt at Arlanda, while granitoids and coherent felsic volcanic to sub-volcanic intrusions had low contents. Following these results, sampling of the bedrock was concentrated in east-west trending profiles across the NNE-trending steeply dipping structural grain of the study area with a main focus on the metasupracrustal complex composed of a volcanosedimentary sequence intercalated with dykes and sills of quartz-poor metaintrusive rocks, pegmatite and pegmatite granite. In addition, detailed geochemical mapping was carried out in two ca 3 km long east-west trending profiles across the enriched zone (see section *3.1.1. Detailed geochemical transect mapping*, below): a northern profile along the road between Arlanda and Kimsta and a southern profile between Arlanda and Västerbytorp (Fig. 9). Sampling of the bedrock followed the SGU's routine for sampling for geochemical and petrographic analysis. The bedrock in the study area is in places lithologically heterogeneous, particularly in the As-enriched supracrustal complex. Multiple samples were taken at lithologically heterogeneous outcrops including also layers, xenoliths, veins, dykes and inclusions of other rock types. The central Arlanda metasupracrustal belt constitutes a high proportion of mica rich schists that have been more easily eroded than the metaintrusive rocks. While there are extensive exposures of metaintrusive rocks, the metasedimentary rocks are poorly exposed and occur in small outcrops along the margins of the plains, close to the contacts of resistant metaintrusives, or at construction sites, along infrastructure and in quarries within the plains. Access to quarries and blasted outcrops at the air side at Arlanda airport enabled access and sampling of fresh exposures of the otherwise deeply eroded and weathered metasupracrustal belt.

The sampling was guided by handheld X-ray fluorescence (HXRF) analysis of the bedrock surface in the field. HXRF measurements indicating potentially elevated concentrations of potentially acidifying or toxic elements led to targeted sampling for whole rock geochemical analysis. Where significantly elevated concentrations of potentially harmful elements, primarily As, were confirmed by total geochemical analysis, renewed sampling was performed to further investigate the reproducibility of the analytical results and the extent of enrichment of the element in the rock.

Figure 10. *Examples of documentation and reference material available from the sample localities for whole rock geochemical analysis. A) Photo documentation in the field. B) Archived reference material from whole rock analysis, to the right including crushed fraction at size <2 mm, (70% of the fraction) and to the left approximately 250-gram subsample of grinded rock powder used for analysis (<75 µm in grain size). C) Reference rock sample. D) Photo documentation of the area used for preparation of microscope preparate (thin section). E) Photomicrograph (optical microscopy, parallel nicols) of thin section.*

3.1.1 Detailed geochemical mapping in transects

Detailed lithological and geochemical mapping of lithologies at the dm-scale in pealed outcrops was carried out in two ca 3 km long east-west trending transects across the metasupracrustal belt (Fig. 9, cf. Evins et al. in press). The methodology of the mapping is described in detail by Evins et al. (in press) and summarized here only. The poor exposure of the metasedimentary belt hindered continuous mapping along the profiles. After substantial peeling of outcrops (removing moss, lichen and thin soil cover) about a sixth of the profile (ca 600 m) of outcrop could be recovered along the profiles. Detailed documentation of the lithological variation of the cleaned outcrops was accompanied by HXRF analyses at rock surfaces in the field, petrographical analyses (microscopy, including ore microscopy), and followed up by total geochemical analyses of selected samples and SEM mineral chemistry analyses of lithologies enriched in As.

3.2 Petrography

Petrographical analyses were made to document mineral assemblages, microtextures (grain size, grain size distribution, grain shape) and mineral chemistry. Petrographical analyses were made for all samples analysed for total geochemical composition (N=192) and some additional samples showing enrichments of rare earth element or radioactive minerals. The petrographic analyses were done using optical and electron microscopy of polished thin sections made from 2x3 cm large and 30 μm thick rock slides mounted on glass (cf. Fig. 10D and E). Ore microscopy was done on selected polished rock specimens, ca 2-5 cm in size.

3.2.1 Optical microscopy

The major and accessory mineralogy and the microtexture of the samples were documented using transmitted light optical microscopy. Ore minerals (oxides, sulphides, and arsenides) were documented by reflective light ore microscopy.

3.2.2 Electron microscopy

Electron microscopy was performed at Uppsala University using a Field Emission Electron Probe Microanalyzer (JXA-8530F JEOL SuperProbe), equipped with both wavelength dispersive (WDS) and energy dispersive (EDS) detectors, as well as secondary (SE) and backscattered electron detectors (BSE). BSE-imaging was used to document the occurrence and textural relations between minerals and for guidance of objects for EDS and WDS mineral chemical analyses. EDS and WDS analyses were also used to map the mineral chemistry of major and accessory phases in the samples, in places supported by WDS point analysis. Two samples highly enriched in As (samples JAN230010A and E) were selected for additional micro-petrographic analysis using

Quantitative Target Mineralogy (QanTmin) EDS mapping at the Luleå University of Technology using a ZEISS Sigma 300 VP scanning electron microscope. Sulphides (pyrite, arsenopyrite and pyrrhotite) in three As-rich samples (JAN220010D and E, and JAN210052B) were handpicked from heavy mineral separates obtained from processing at a Wilfley panning table and moulded in epoxy. The mineral chemistry of the sulphides in the mounts was analysed by a FEI Quanta 650 FEG SEM equipped with an 80 mm^2 Oxford instruments XMAX EDS detector at the Swedish Museum of Natural History in Stockholm.

3.3 Hand held X-ray fluorescence analysis

Hand held X-ray fluorescence (HXRF) on outcrop surfaces in the field was used for reconnaissance geochemical mapping of the bedrock. The results were used for the selection of sampling sites for bulk rock geochemistry. Two different HXRF instruments were used: a Bruker S1 Titan with an 8 mm collimator, using Geoexploration calibration and a Scantec Sci-Aps X-200 with a 10 mm collimator and calibrated for soil. An analytical time of 60 seconds was applied for both instruments. There was a good agreement in analytical results for As between the instruments. As a rule, three repetitions of measurements were done at each analysed spot, each covering ca 0.6-1.0 cm^2 of the outcrop surface (instrument collimators 8-10 mm).

3.4 Whole rock geochemistry

Only bedrock without rust staining or other weathering-related discolorations was sampled for geochemical analysis. Samples were recovered with hammers. The sample size was adjusted according to grain size and texture. Samples taken with hammer and sledgehammer in the field ranged in size from approximately 0.5 kg (fine-grained, homogeneous rocks) to over 20 kg (heterogeneous and coarse-grained rocks). The detailed sampling in the two transects across the arsenic-rich fine-grained metasupracrustal complex was done in some sections by drilling, recovering approximately 5–10 cm long drill plugs with a diameter of 2 cm. The drill plugs were cut in half and one part was used for bulk rock geochemical analyses. The other part was also cut in half, where one part was used to prepare a thin section, and the other part archived as reference material. Drill plugs used for geochemical analyses ranged in sample size between 0.03 and 0.20 kg. Sample quantities for all specimens have been reported along with the analytical data.

All sample preparation and whole rock geochemical analyses were done by ALS Scandinavia in Piteå. The sample preparation involved fine crushing of the samples (<2 mm, 70% of the fraction) followed by subdivision and fine grinding of an approximately 250-gram subsample into a rock powder with a grain size <75 µm. Dissolution of the samples was done in three different modes, producing three different aliquots. Complete dissolution of the sample, including robust silicate minerals, was done in a lithium borate fusion. Acid leaching (partial dissolution) included dissolution in four acids (nitric acid, perchloric acid, hydrochloric acid, and hydrofluoric acid), and

in aqua regia (sulfuric acid and hydrochloric acid). Analyses of volatile elements such as arsenic must be done by acid dissolution, as they vaporize during fusion of the sample. The analyses included ALS analysis packages CCP-PKG01, ME-MS41, and PGM-ICP23. These include analysis of all major elements, trace elements, and volatile trace elements (i.e., arsenic, cadmium), as well as carbon, sulphur, and base metals. The major elements are reported as oxides and include SiO_2, Al_2O_3, TiO_2, $FeO+Fe_2O_3$ (total FeO), MgO, MnO, Na_2O, K_2O, and P_2O_5.

The evaluation of the geochemical composition of the bedrock in the Arlanda-Rosersberg area performed in this study includes 192 whole rock whole rock geochemical analyses. All geochemical data used or produced in this study are available in the SGU map viewer for lithogeochemical analyses available at https://apps.sgu.se/kartvisare/kartvisare-bergartskemi.html . This data base includes information on analytical techniques, laboratory, sample size, sample ID, sample location and type of rock analysed. The data can also be directly downloaded from the SGU website in the file formats: OGC GeoPackage, ARC GIS Pro Layer Definition file or as a QGIS Layer definition file (cf. https://resource.sgu.se/dokument/produkter/lithogeochemistry-description.pdf).

3.5 Documentation and reference samples

All sample localities have been documented with coordinates (Sweref99), description of rock type and a petrographic description from the field together with photographs of sample localities. For all rocks that have been analysed for their whole rock geochemical composition, a reference sample (fist size) and a thin section representative for the rock analysed have been archived at the SGU together with the leftover material from the geochemical analyses including the crushed sand-gravel fraction material and the analysed finely ground rock powder (Fig. 10). This material can be requested from SGU for further investigations.

4. RESULTS

4.1 Concentrations of arsenic in the bedrock

Evaluation of 192 geochemical analyses of the bedrock in the Arlanda-Rosersberg area shows that enrichment of As is restricted to the metasupracrustal complexes dominated by metasedimentary rocks (Fig. 11A; micaschists, psammites, pelites). 71 out of 109 analyses (65%) of the analysed metasedimentary rocks had an As content ≥ 10 ppm, and just over 30% had an As content ≥ of 100 ppm. Following the bedrock map of Stålhös (1972), the study area covers two principal metasupracrustal belts, a northern belt extending from Vidbo in the north to Odensala in the south-east, and a southwestern belt, from Lunda in the northeast to Norrsunda in the southwest (Fig. 3). Prevailing high concentrations of As (> 10 ppm) occurs in a ca 1.3 km wide zone within a NNE-trending steeply dipping metasupracrustal complex in the southeastern Arlanda area (Västerbytorp-Kimsta Zone; Fig. 3). Within this zone, several different lithologies are highly enriched in As, with concentrations > 100 ppm, in places exceeding 1000 ppm. Similar enrichment of As was rare in analogous lithologies outside the zone (Fig. 11B), and not observed outside the Arlanda-Rosersberg area. With the exception for a foliated metagranite (Uppsala granite) sampled close to the contact of the northeastern metasupracrustal belt (JAN230032A), none of the silica-rich intrusive rocks (granites and granitoids and reworked equivalents) showed any enrichments of As (Fig. 11A). A coherent 0.5 km wide and 6.5 km long metavolcanic unit dominated by sub-volcanic intrusions of andesitic to dacitic composition also show prevailing low concentrations of As (Fig. 3 and 11). However, ca 0.5-5 meters wide andesitic to dacitic plagioclase +/- amphibole porphyritic coherent volcanic sills and dykes in the southwestern supracrustal complex in places show high- (>10 ppm) to very high (>100 ppm) concentrations of As (Fig. 5A). Repeated sampling of the same sills/dyke showed low concentrations of As indicating local enrichment of As in this lithology only.

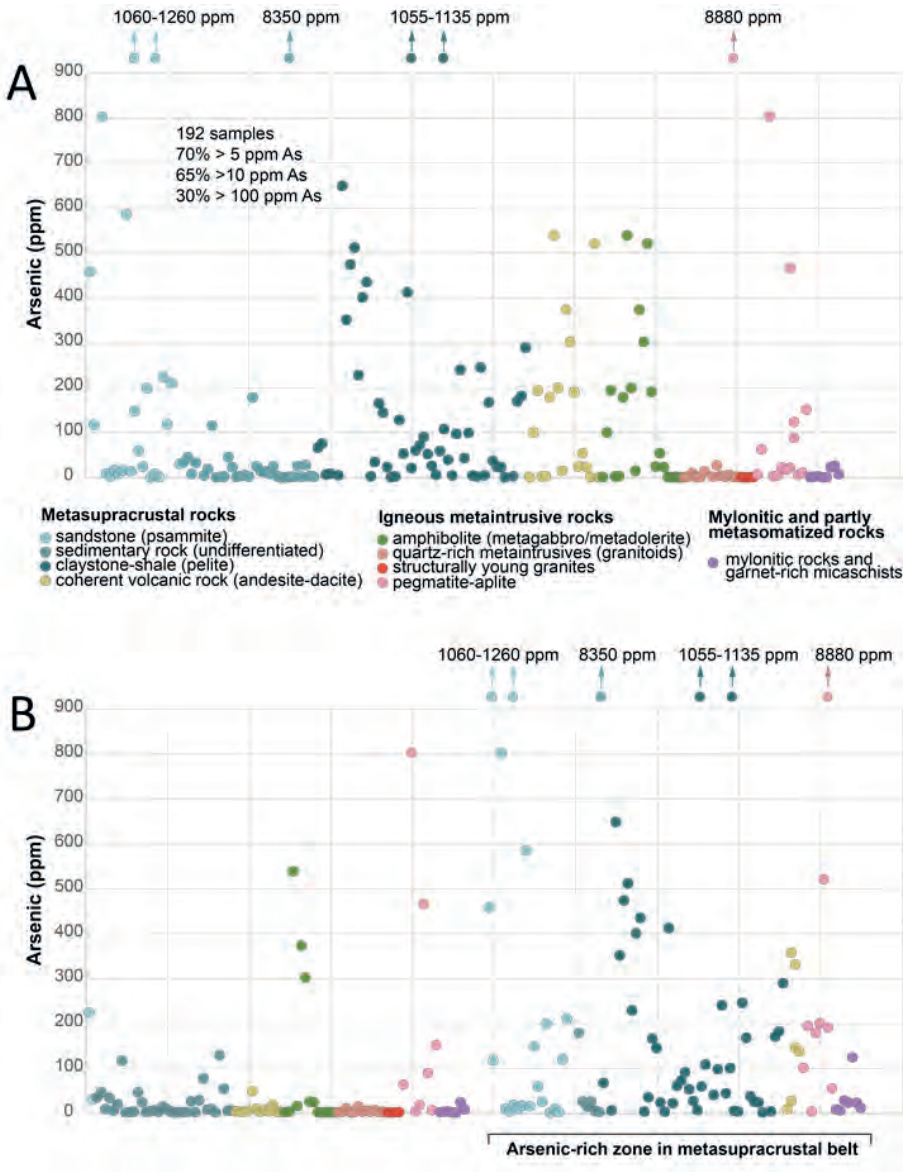

Figure 11. *Diagrams showing total content of As (ppm) in the bedrock in the Arlanda-Rosersberg area. Different colours denote different lithologies. Note that analysis > 900 ppm As are plotted outside (above) the diagrams with total concentrations indicated. A) Analytical data grouped according to lithology. B) Analytical data grouped according to lithology and location inside or outside a 3 km wide zone enriched in As (cf. Fig. 9).*

4.1.1 Enrichments of arsenic in the Västerbytorp-Kimsta Zone

Significant enrichments and in places highly elevated concentrations of As (≥ 100 ppm), locally up to near 1 wt.% As, were discovered in an approximately 1.3 km wide NNE-trending zone in the south-eastern metasupracrustal belt exposed east and south-east of the runways at Arlanda, here referred to as the Västerbytorp-Kimsta Zone (Fig. 9). The results from a layer-by-layer mapping campaign across the zone, complemented by handheld XRF analyses in each lithological layer, and whole rock geochemical analyses of selected samples is summarized here while presented in detail by Evins et al. (in press).

In the Västerbytorp-Kimsta Zone, high-As concentrations occur in metasedimentary units as well as in intermediate- to mafic sills and dykes, and fractionated pegmatites. The zone is dominated by metasedimentary rocks with a complex banding defined by variations in texture, the relative amounts of muscovite and biotite, and grain size. The individual bands range in thickness from less than a 1 cm to more than 1 meter. Occasionally, a relict sedimentary layering is observed, with individual layers varying between 1 and 5 mm in thickness (Fig. 4B). Arsenic enrichment is more common in the mica-rich bands. There is, however, no distinct correlation between silica content (linked to quartz content) and concentration of As.

The metasedimentary rocks are intercalated by bands and layers of mafic, quartz-poor intrusive rocks (amphibolites, <53% wt% SiO_2). Fine grained amphibolite typically occurs as 1–5 decimetre-wide deformed, folded, and disrupted layers (Fig. 6A). Over two meters wide disrupted layers of medium-grained amphibolite (metagabbro) are present in places. The amphibolites in the zone are all enriched in As. In the northern transect (Kimsta road), all amphibolitic layers show significant arsenic enrichment at between 100 and 500 ppm.

The arsenic rich zone includes pegmatites and aplites with arsenic levels between 20 and 800 ppm (Fig. 11A). In the coarse-grained pegmatites, medium- and coarse-grained arsenic minerals can be observed with the naked eye in the field (Fig. 8D, 12A-B). The arsenic-enriched pegmatites are characterized by greyish white to white feldspar, abundant white mica (muscovite), and typically with small amounts of blue-green apatite and pink garnet. Dark mica (biotite) is absent or rare in the As-rich pegmatites. The boron-bearing silicate tourmaline, occasionally in abundant amounts, also appears to be a characteristic mineral of the As-enriched pegmatites (Fig. 8E). The enriched pegmatite dykes are on average between 0.5 and 2 meters in width and generally run in a north-north-easterly direction, roughly parallel to the tectonic layering in the surrounding schist belt. In many places, enrichment of As in the bedrock was spatially associated with tourmaline bearing pegmatitic and aplitic rocks (Fig. 8F).

The highest concentration of As, up to near 1 wt% (8350 ppm) was measured in a small road cut in the northern transect, in the contact zone between a mica-rich banded rock of a presumable sedimentary origin and a medium grained amphibolite. The road cut exposes dark grey fine- to finely medium grained rocks without rusty staining or discolouring (Fig. 13). At this locality, layers enriched with arsenic minerals could be

identified in the field in non-oxidized freshly cut rock surfaces with the naked eye (Fig. 12C-D). Elevated levels of As in mica-rich metasedimentary rocks in places also occur along contacts and in the vicinity of tourmaline bearing pegmatites (Nilsson 2024).

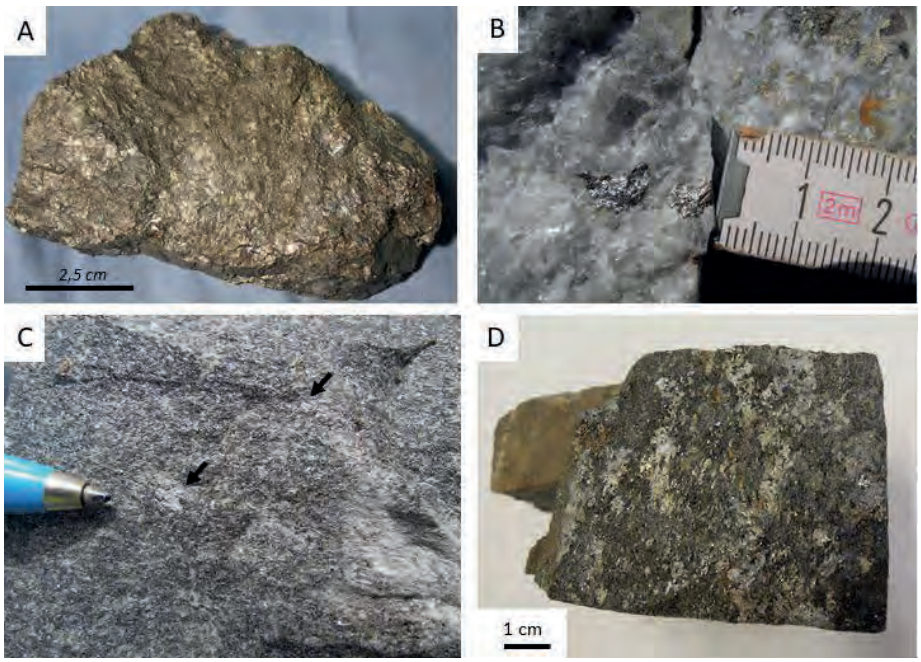

Figure 12. *Arsenic minerals in the* study *area. A) Large chunk of arsenopyrite from a fractionated pegmatite dyke collected during the construction of runway 3 at Arlanda airport. B) arsenopyrite/löllingite from garnet-tourmaline-beryl-bearing light grey K-feldspar pegmatite dyke at Långåsen (sample JAN210052C). C) Arsenopyrite/löllingite (silvery minerals marked with arrows) in the matrix of a strongly As-enriched fine-grained metasedimentary rock at Kimsta (8830 ppm As, sample JAN220010A). D) Same rock as in C) exposing a discrete shear zone with arsenopyrite, löllingite, pyrrhotite, and pyrite in the shear plane.*

4.2 Arsenic mineralogy and textural context of arsenic minerals

In rocks with an As concentration exceeding 500 ppm, the arsenic minerals arsenopyrite or löllingite could, as a rule, be identified with a hand lens directly in the field (Fig. 8D and 12). In coarse-grained pegmatitic rocks, medium- to coarse-grained arsenopyrite or löllingite, could be easily identified already with the naked eye (Fig. 12A and B). By using a hand-lens, medium- to fine grained enrichments of arsenopyrite-löllingite could also be detected in finer-grained rocks, such as medium-grained amphibolites. In the fine-grained sedimentary rocks, however, arsenic minerals could only be identified in the field in highly enriched layers (>5000 ppm; cf. Fig. 12C). In most cases, however, identification of the arsenic minerals had to be done by ore microscopy and the distinction between arsenopyrite, which is an arsenic sulphide (FeAsS), and löllingite, which is an arsenide ($FeAs_2$) could only be done by EDS electron microscopy.

In all investigated samples, As occurs as both arsenopyrite and löllingite. Arsenic was not found in pyrite. The arsenic minerals were located at grain boundaries rather than as inclusions within other silicates (Fig. 14). In rocks of an igneous origin (amphibolite), the arsenic minerals were evenly distributed in the rock matrix. With few exceptions, in non-weathered fresh rocks, there were no microtextural indications of enrichment of arsenic in alteration or deformation zones. High concentrations of As (>1000 ppm) were in places detected with HXRF measurements at rusty weathered fracture surfaces. Further investigations of weathered rock materials with WR analysis were beyond the scope of this study, but is a target for future studies.

In lithologically banded rocks of a sedimentary origin, the distribution of arsenic minerals differed between the different bands, with enrichment of arsenic minerals in some bands. In many places, the enrichment was found in mica rich bands.

Late re-mobilization of arsenic was also documented in a metasedimentary rock where arsenopyrite partly decomposed to scorodite was spatially associated with a structurally late discrete micro-shear zone. Detailed petrological mapping of the most As-rich metasedimentary rock (sample JAN220010A), by EDS quantitative mineralogical mapping showed that also the occurrence and proportion of arsenopyrite and löllingite differed at the microscale between the different lithological bands. Some bands were dominated by löllingite while others were dominated by arsenopyrite. The uneven distribution of arsenic minerals was accompanied by the uneven distribution also of minerals composed of immobile elements such as ilmenite.

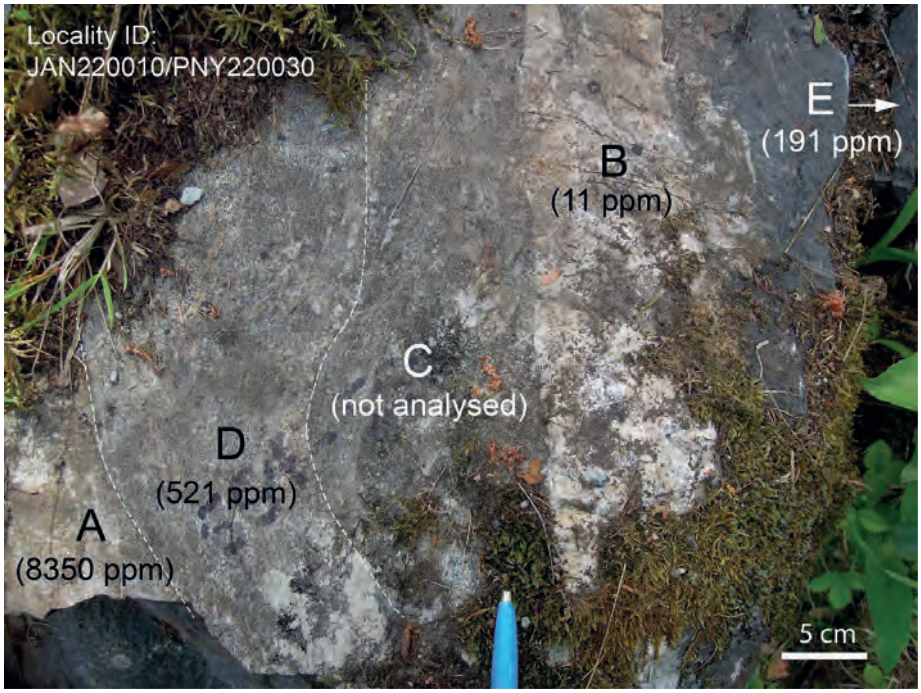

Figure 13. *Field photo of a lithologically heterogeneous outcrop in the supracrustal belt (ID: JAN220010 and PNY220030), strongly enriched in As. Whole rock As concentrations for each lithological unit given in parathesis. Labelling A to E refers to sample ID-extension for samples JAN220010A-E. Sample JAN220010A is a metasedimentary rock. Samples JAN220010C-E are amphibolitized metagabbro sills showing variation in texture, composition of amphiboles and plagioclase content. Sample JAN220010B is a tourmaline-garnet-apatite rich aplitic dyke.*

As a rule, arsenopyrite was the dominant arsenic mineral, but löllingite was easily recognised in all samples (Fig. 14). In rocks of both sedimentary and magmatic origin (amphibolites), löllingite occurred both as a single mineral phase (Fig. 14A), in places as euhedral crystals, and associated with arsenopyrite (Fig. 14-C). Löllingite commonly formed a core mantled by arsenopyrite and with the contact between löllingite and arsenopyrite typically including droplets of native bismuth and/or tellurium (Fig. 14B). In many places, the arsenopyrite mantle was in turn overgrown by pyrrhotite ($Fe_{(1-x)}S$; Fig. 14B). Löllingite cores overgrown by arsenopyrite was a common texture, but the proportions between arsenopyrite and löllingite varied widely within the same rock (cf. Fig. 134 and C). Intergrowths of euhedral löllingite and arsenopyrite were also observed (Fig. 14D). The content of As in pyrite was below detection limits for the applied EDS and WDS analyses, i.e. below 150-200 ppm. Consequently, pyrite is not a primary source for As in the studied area. Nickel-bearing löllingite [$(Fe_{0.7}\ Ni_{0.3})As_2$)] was in

places found in amphibolite together with arsenopyrite with zones enriched in nickel and in places also cobalt [(FeNiCo)AsS].

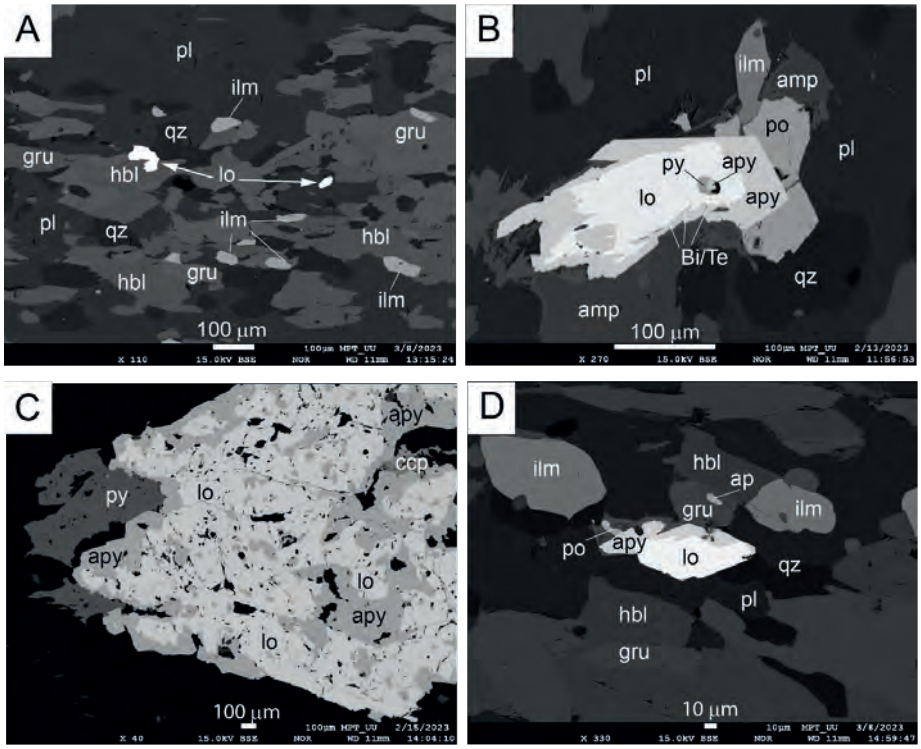

Figure 14. *Electron back scattered (BSE) images of arsenic minerals in rocks in the high-As zone at Arlanda (Kimsta road profile). A) Single löllingite crystals growing along grain boundaries in the rock matrix of a metagabbro (sample JAN220010C); B) Löllingite overgrown by arsenopyrite and pyrrhotite; a characteristic texture in the high-As rocks at Arlanda. Note the BSE-bright dissolution of Bi/Te at the contact between löllingite and arsenopyrite (meta-gabbro sill, sample JAN220010C); C) Löllingite megacryst intergrown with arsenopyrite and with pyrite at the left tip (metasedimentary rock; sample PNY220030A1 at contact to metagabbro). D) Euhedral löllingite growing together with euhedral pyrrhotite in a high-As amphibolite (same sample as in A). Abbreviations: lo-löllingite; apy-arsenopyrite; po-pyrrhotite; py-pyrite; ccp-chalcopyrite; pl-plagioclase; qz-quartz; amp-amphibolite; gru-grunerite; hbl-hornblende; ilm-ilmenite; ap-apatite; Bi/Te-bismuth/tellurium.*

4.3 Sulphur

The bulk rock geochemical analyses showed that 41% of the samples had a total S content < 0.1 wt% (N=79) and 68% <0.5 wt% (N=130). Both groups include all rock types sampled in the area. The quartz-rich intrusive rocks (granitoids and metamorphic equivalents) were overall low in S (< 0.1 wt%; N=17). A slight enrichment was noted in three samples from close to the contacts to the metasedimentary belts. The metasupracrustal rocks and quartz-poor metaintrusive rocks (amphibolites) vary in total S from below the detection limit up to ca 1 wt%. Rocks with a concentration exceeding 1 wt% were metasedimentary rocks and coherent volcanic layers in the metasedimentary belt (N=9). Only one sample of a biotite-rich quartz-feldspathic schist (psammite) had a higher S concentration of 2 wt % (sample IKL230015C). This is a common lithology in the supracrustal belt, and similar rocks ranged in total S from 1 wt% to below the detection limit. Values between 1.0 and 0.5 wt% was detected in amphibolites and metasedimentary rocks (N=24), but also these lithologies showed a wide range in S content varying from 1 to below 0.1 wt%.

4.4 Non-arsenic sulphides

Pyrrhotite ($Fe_{(1-x)}S$) was the dominating sulphide in the investigated samples and typically occurs as fine-grained crystals in the rock matrix. In many places, pyrrhotite occurs as an overgrowth on arsenopyrite (Fig. 13B). Pyrite was rare in amphibolites but more common in the felsic metaintrusions and the metasupracrustal rocks. It occurs both as fine-grained crystals in the rock matrix and as coarser grains in structurally late alterations zones spatially associated with low temperature minerals such as chlorite, albite, K-feldspar (cf. Fig. 5D). Pyrite is the dominating sulphide in discrete foliation planes developed oblique to the early deformation fabrics (early foliation, S1). In places, single crystals of pyrite are seen to grow across the mineral fabric also outside micro-shear zones. Other sulphides are subordinate and include chalcopyrite ($CuFeS_2$) and sphalerite (ZnS). Chalcopyrite in places occurs as inclusions inside pyrrhotite. Sphalerite is rare. Both chalcopyrite and sphalerite in places occur in late alteration zones together with chlorite and pyrite. Both amphibolites and metasedimentary rocks in places have pyrrhotite with intergrowths of pentlandite [$(Fe,Ni)_9S_8$].

5. DISCUSSION

Prediction and risk management of high concentrations of contaminating elements in the bedrock depend on an in-depth understanding of *where* and *how* these elements are sitting in the bedrock. At present, this knowledge is lacking for large parts of the Swedish bedrock outside metallogenetic domains of economic interest. Bedrock mapping in the construction intensive urban parts of the Mälardalen region, including the densely populated Stockholm area and the infrastructure centre of Arlanda airport was carried out in the years 1964-1966. As a consequence, there is a lack of up-to-date geological information critical for risk assessment such as trace element geochemistry and modern geological models for the build-up of the bedrock (c.f., Andersson et al. 2022). Later complementary mapping of the bedrock for assessments of crushed rock aggregate functionality and modelling of the structural build up the bedrock for construction projects has not involved surveying of bulk rock or mineral chemistry (Persson et al. 2002; Schoning and Wahlgren 2019).
In this study, reconnaissance geochemical mapping of the bedrock in the Arlanda-Rosersberg area accompanied by detailed geochemical mapping in traverses across a metasupracrustal belt enriched in As has contributed to two principal discoveries:

- Bulk rock contents of As in metasupracrustal belts at between 10-100 ppm, and in a 1.3 km wide zone, exceedingly high concentrations between 100 to 8000 ppm.
- Widespread occurrence of arsenide (löllingite) in the basement rocks.

These discoveries have a bearing on the management of rock masses from the area and the leaching properties of the rocks. The implications of these discoveries are discussed below.

5.1 Unexpected high concentrations and source of arsenic in the bedrock in the Arlanda-Rosersberg area

The concentration of As in the crust varies widely, the upper crust being more enriched (average 2-6 ppm), than the lower crust (average <0.06 ppm Witt-Eickschen et al. 2009; Hu and Gao 2009, Masuda 2018); and with large differences between different rocks. Non-mineralised quartz-feldspathic rocks are generally low in As (<20 ppm; Matschullat 2000). At Arlanda, granitic rocks and reworked equivalents are as a rule low in As (<10 ppm). The highest concentrations of As in granitoid rocks (6-14 ppm) occurs in metagranites sampled closed to the contact to the As-rich metasedimentary belt and in metagranite with high-As xenoliths suggesting contamination of the granitic magma may by assimilation of As-rich side rocks.

Subaqueous sedimentary rocks are known to be enriched in As; the average As content in North American shale is ca 30 ppm (North American Shale, NASH, Rollinson 1993). Organic shales, may however, be highly enriched with As contents up to 1000 ppm, and up to 3000 ppm for iron-enriched strata (Smedley and Kinniburgh 2002). The strongly As-rich supracrustal belt at Arlanda does not contain black shales. Some units are graphite bearing but to a moderate extent and none of the samples had a total carbon

content >0.8 wt%, and there is no correlation between As-enrichment and the total carbon content. This demonstrates that also low-carbon metasediments can be highly enriched in As at levels previously not known in Sweden.

Mafic and intermediate volcanic rocks may also be enriched in As, although generally not exceeding contents of 200 ppm (Smedley and Kinniburgh 2002; Mukherjee et al. 2024). In the Arlanda area, however, sills and dykes of mafic and intermediate metaintrusive rocks in the As-rich parts of the metasupracrustal belt in places have highly elevated concentrations of As at between 100 and 500 ppm, well above values reported in the literature for these rocks. Highly elevated As-concentrations at 400-700 ppm occur in gabbroic rocks emplaced in a high As (ca 100 ppm) metasedimentary belt in the Åkerberg area, ca 20 km north-northeast of Boliden in the Skellefteå ore district. The bedrock is part of a metallogenetic domain well known for hosting high-As mineralisations. The bedrock in the Åkerberg area is partly strongly impregnated by sulphide mineralisations associated with extensive enrichments of arsenopyrite/löllingite. The setting of the Arlanda area is different from the high-As metallogenetic domains in the Skellefteå and Bergslagen ore districts as significant sulphide mineralisations are lacking. In fact, in the Arlanda area, As is the only element showing extreme enrichment.

Results from this study demonstrates As-concentrations in the bedrock at Arlanda, by far exceeding values in the literature for non-mineralised basement rocks (e.g., reviews by Smedley and Kinniburgh 2002; Bowell et al. 2014; Masuda 2018). Highly elevated As-levels at 100 ppm to above 8000 ppm such as those discovered in a ca 1.3 km wide zone in a non-mineralised supracrustal complex at Arlanda (cf. also Nilsson, 2024; Evins et al., in press) are otherwise reported in coals and As-contaminated deposits associated with industrial or mining activity (Craw and Bowell 2014).

There is a striking correlation between the occurrence of high-As rocks (> 10 ppm) and As-contamination of groundwater (> 10 µm/l As), a pattern particularly evident for tubed wells penetrating the bedrock (Fig. 2). This relationship demonstrates that As is leaching from the bedrock to the bedrock aquifers. The time scale at which the contamination occurs is, however, unclear.

5.2 How common is arsenide in the non-mineralised basement?

Understanding how As is extracted from rocks and the mechanisms steering this process requires an understanding of how and it what form As is sitting in the rocks. In the literature, oxidation of arsenopyrite (FeAsS) and pyrite (FeS_2) is assumed to be the primary source for As-contamination of the hydrosphere (e.g., Bowell et al. 2014; Mukherjee et al. 2024.). Although incorporating smaller amount of As only, due to its common occurrence, pyrite is assumed to the main source of As contamination (Bowell et al. 2014.). Löllingite ($FeAs_2$) is generally not considered as a relevant source as it is regarded as a rare mineral, known to occur primarily in hydrothermal ore deposits and fractionated pegmatites (Majzlan et al. 2014; Drahota et al. 2022). In reviews of primary sources of As, löllingite may not even be mentioned (e.g., Smedley and Kinniburgh

2002; Hug et al. 2020). In this study it is demonstrated that löllingite is a common As-mineral in non-mineralised basement rocks, including metasedimentary rocks (psammites to pelites), amphibolites (metadolerite and metagabbro) and felsic metavolcanic rocks (metadacite-metaandecite; Andersson et al. 2024a, b). Löllingite in common basement rocks were also discovered in a geochemical mapping campaign focused on As enrichments carried out in the Södertälje-Mariefred area in the southern parts of the Mälardalen region (Andersson et al. in press). In this area, löllingite was documented in mafic to intermediate intrusions and sedimentary rocks metamorphosed at high-temperature granulite facies conditions. In both the Arlanda-Rosersberg and the Södertälje-Mariefred areas, löllingite was discovered by detailed investigations of ore minerals by ore microscopy followed up by EDS/WDS analyses. Such detailed studies of ore minerals are commonly not applied in petrographic analysis of non-mineralised rocks. The distinction between arsenopyrite and löllingite, however, requires identification by electron microprobe or Raman spectroscopy. Consequently, it is reported in detailed studies of mineralisations (e.g., sulphide ores and fractionated pegmatites) where these techniques are applied, but otherwise foreseen. It is therefore likely that the occurrence of löllingite is foreseen in investigations of non-mineralised crust and that the occurrence of löllingite ordinary basement rocks has been hugely underestimated. If this is true, it is problematic as löllingite is more readily oxidized than arsenopyrite; it also results in different oxidation products and the relative proportion and textural relations between löllingite and arsenopyrite steers the mode of As-leaching (Zhou et al. 2021). A consequence of the assumption that löllingite is an uncommon As mineral, is that its oxidative dissolution has attracted little attention (c.f. Lengke et al. 2009). The literature on As-contamination from non-mineralised, low sulphide As-rich rocks is scarce (c.f., Bundschuh et al. 2013). The discoveries of löllingite as a significant primary source of As in the unmineralised central Swedish basement call for further research on the mechanism of extraction of arsenic from löllingite.

5.3 Source of arsenic in the Arlanda basement rocks

To understand the distribution of As in the bedrock, it is necessary to understand how As was concentrated in the rocks. In this study, extreme enrichments of As was documented in mica-rich metasedimentary units, in intermediate-mafic sills and dykes, and in fractionated pegmatites within a strongly deformed metasupracrustal belt. Similar lithologies outside the belt lack As enrichments in the bedrock and accompanying contamination of bedrock groundwater aquifers (Fig. 3).

The high concentrations of As in the metasedimentary rocks are interpreted to reflect primary enrichments in the sedimentary deposits, likely at reducing conditions (cf., Mukherjee et al. 2014; 2024). A primary enrichment is supported by the strata bound distribution of As-minerals in the metasedimentary rocks. A spatial correlation with tourmaline in the high-As sedimentary rocks supports a marine boron rich environment. The difference in As enrichments in the sedimentary belts at Arlanda and seemingly

similar sedimentary belts immediately east and west thereof, may reflect differences in the redox conditions that once prevailed in the different sedimentary deposits.

The arsenic rich pegmatites at Arlanda may have formed from partial melting of the As-enriched metasediments whereby fractionation of the melt has led to successive enrichments of As (Smeds 1990). Analogous with the As-rich sedimentary rocks, the fractionated pegmatites are rich in tourmaline suggesting a boron rich source rock. Arsenic enrichments in the mafic to intermediate metaintrusions could be a primary feature reflecting As enrichment during fractionation of a gabbroic magma or assimilation of As-rich sedimentary side rocks (Alirezaei and Cameron 2002; Evans et al. 2022). The arsenic minerals are evenly distributed in the rock matrix of the metaintrusions which preserves a relict igneous texture, and are not concentrated along contacts to the side rock or along fluid driven alteration pathways. The mafic to intermediate metaintrusions are penetratively amphibolitized. Since As is easily carried by hydrous fluids, it could also be speculated that As-rich hydrous fluids infiltrated these rocks during recrystallization and hydration of these rocks at amphibolite facies metamorphic conditions.

5.4 Methodology

Low density reconnaissance geochemical mapping across the area using HXRF measurements on outcrops followed up by whole rock geochemical analysis of selected lithologies proved to be a fast and effective method for a first-hand distinction between high- and low As lithologies. It allowed early recognition of felsic igneous rocks and reworked equivalents as non-risk lithologies. For example, the early recognition of a ca 500 wide and 1.3 km long volcanic unit in the central part of the area as a non-risk lithology. Felsic volcanic rocks are otherwise a well-known source of As that may contribute to significant contamination of the hydrosphere (Bunschuh et al. 2012; Morales-Simfors et al. 2020). Felsic volcanism is also a main source of As in the Swedish metallogenetic Bergslagen and Skellefteå districts (Allen et al. 1996; Jansson et al. 2021). The results from the early reconnaissance mapping showed a close correlation between high concentrations of As in the bedrock and contamination of potable water in tubed wells penetrating the bedrock. These early results were later corroborated by detailed geochemical mapping (WR and HXRF) in two transects across a ca 3 km wide steeply dipping metasedimentary belt enriched in As, including layer mapping at scale <1m (Evins et al. in press). This work resulted in a narrowing down a high-As zone to ca 1.3 km in width with a high-frequency of lithologies with an As content >100 ppm. Further layer by layer mapping at scale < 1m, however, was not effective to further resolve the lithological and spatial resolution of high-As concentrations. The results from the detailed layer mapping and the effectiveness of the use of HXRF measurements in the field in the detailed mapping is further discussed in Evins et al. (in press). Strategies for sampling and the application of whole rock and HXRF analysis in geochemical mapping of the bedrock are further discussed below.

5.4.1 Strategies for sampling and whole rock geochemical analysis

Selection of samples for WR analysis was based on the lithological characteristics of the rocks, and with the aim to understand if and how As was enriched in the different lithological components. Application of grid sampling would have given information on the rough areal distribution of As and a statistical estimate of the rick to encounter high As rocks. It would, however, not account for the geological context of samples, information critical for the understanding of how As is distributed in the crust. In addition, about ¾ of the study area exposes felsic igneous rocks, known as a non-risk lithology. Grid sampling would have resulted in a large number of WR analyses of these mostly lithlogically homogeneous non-risk lithologies, and few, if any representative analyses of layers, sills and dykes of high-As igneous rocks in the banded sedimentary belt and xenoliths in the igneous rocks. In contrast, lithological basis for sampling in detailed transects help to get an understanding of how and where As is sitting in the bedrock. Successful targeted sampling, however, relies on geological expertise and proper documentation of samples.

When whole rock geochemical analyses are done on rock masses in construction projects the analytical work is commonly focused on a specific array of elements, for example common potentially contaminant trace elements such as Cu, Ni, Pb, and Zn. If the main concern is fear for acid rock drainage, only sulphur (S) may be analysed. The choice to analyse only one or a restricted range of elements is motivated by lower costs for the analytical work. It may, however, be more efficient, and in the long run, more cost efficient to analyse a full range of elements. For some laboratories, the difference in cost between a full geochemical characterisation and a selection of elements may be minor. A full geochemical characterisation, including also major elements, will significantly help to understand the lithological characteristics of rock masses enriched in the element of interest. For example, a high concentration of Ni is commonly correlated with low-silica igneous rocks with high contents of magnesium, titanium and phosphorous, and low contents of potassium (gabbroic rocks). Such established correlations between major and trace elements can be a powerful tool for coupling enrichments of contaminating elements with a specific rock type. Analysis of only one or a few elements may also lead to that high-concentrations of elements harmful to health and the environment is foreseen. Environmental impact potentially related to acid rock drainage from crushed rock aggregates at two sites in the southern Stockholm area has resulted in extensive analysis of the total S content of rock masses in construction projects in the Mälardalen region. Commonly, however, only S is analysed. This hinders modelling of the lithological and geological context of aggregates enriched in S and turn focus from other potentially contaminating elements not bound to S, such as for example, As. The rocks at Arlanda analysed in this study showed no simple correlation between high content of S and As. Likely due to the common occurrence of the As-mineral löllingite ($FeAs_2$). In fact, many samples with a high As-content, 100 > ppm, had a total S content below 0.1 wt% S, or even below the detection limit with the applied analytical method (<0.01 wt%). This exemplifies the drawback with a focus on only one element.

5.4.2 Tracing enrichments of elements harmful to health and the environment using handheld X-Ray fluorescence analysis

Handheld X-ray fluorescence analyses may be a powerful tool for reconnaissance mapping of trace elements in the bedrock. Although there are differences between different HXRF instruments in the analytical performance and the ability to correct for spectral interferences, the technique has proven to be highly suitable for analysis of elements like As, Pb and Cu (e.g., Hall et al. 2014). However, successful application of HXRF measurements on rock surfaces in the field also depend on the distribution and grain size of As-minerals in the rock. The collimators of HXRF devices are in the range of being 0.8-1.0 cm wide. Consequently, the analyses cover a small portion of the rock and only include analysis of the rock surface. In this study, HXRF analysis on outcrops in the field proved to be a powerful tool to trace high-As concentrations (>100 ppm) in the bedrock. The bulk of the investigated bedrock consisted of fine-grained to finely-medium grained rocks with As-minerals well distributed in the rocks and with few exceptions lithologies well suitable for HXRF analysis (Fig. 15A). If the As minerals are unevenly distributed, for example enriched in fractures and bands, or in other ways nuggety or clustered in the rock, HXRF analysis may generate highly variable results that may be difficult to interpret and enrichments may be foreseen (Fig. 15B). Rocks with the same bulk As-content but with differing grain size of the As-minerals may also give highly different results (Fig. 15C). Ergo, a rock with fine-grained As-minerals evenly distributed in the rock matrix is more prone to give reproducible analytical results across the rock surface while a similar rock with few coarse-grained As-minerals will give variable results due to a lower probability to hit the few, but larger As-minerals.

In this study, samples with As concentrations <100 ppm showed a lower agreement between the HRF and WR analyses than samples with As >100 ppm. Such scattered results from measurements across a seemingly homogeneous surface is interpreted to reflect a low density of As minerals coupled to a lower bulk rock content of As (Fig. 15A).

In summary, WR analysis of homogenised rock powder gives a measure of the geochemistry of the sampled rock volume typically in the order of several kilos of rock, while HXRF analysis only measure the concentrations of elements of a rock surface (and only elements heavier than magnesium). These are two distinctly different analytical methods and comparison of data between the two methods is not straight forward. If the elements to be analysed are fine grained and well distributed in a fine-grained matrix the measurements of the elemental concentrations obtain by the two methods may be conformal (Fig. 15A). If the elements are heterogeneously distributed in the rock mass, the results are likely to diverge. Handheld X-ray analysis of homogenised rock powder is more likely to align with WR analytical data.

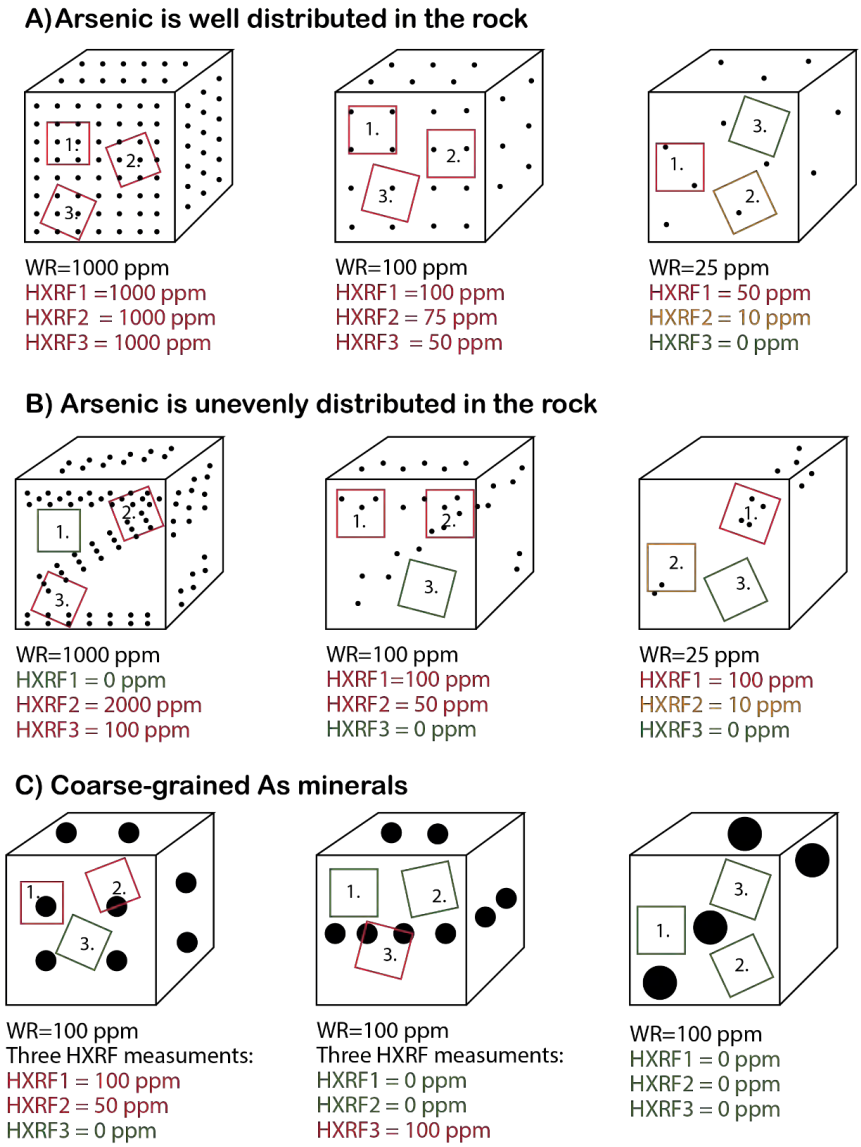

Figure 15. *Schematic illustration of how the grain size and distribution of As-minerals in a rock volume may affect the success of geochemical analysis using HXRF measurements on rock surfaces. Note that an uneven mineral distribution may also contribute to measurements giving higher values with HXRF than those obtained by WR analysis (the right box in B).*

The great advantage of the HXRF is the high number of analyses that can be obtained in a short time which allows time effective reconnaissance geochemical mapping of rock exposures. All geochemical mapping requires a sampling strategy, and HXRF is an effective tool to obtain preliminary data of enrichments of heavy elements to steer sampling of the rock. Therefore, although measurements by HXRF analyses in the field may be challenged by calibration issues, weather conditions and differences in operational mode between operators, it is a powerful tool for reconnaissance geochemical mapping and guidance of targeted sampling.

5.4.3 On the use of geophysical methods to trace high-As enrichments

Measurements of the magnetic and electrical properties of rocks are widely used methods for geophysical mapping of the bedrock. Application of these methods, however, rely on an understanding of the mineralogical properties that cause the geophysical signatures. The magnetic anomaly map of the Arlanda area shows a north-northeast trending positive magnetic anomalies roughly following the arsenic-rich zones (SGU 2024). No specific high magnetic lithology was encountered in the detailed mapping of the bedrock across the anomalies. None of the rocks examined were magnetite bearing, but all rocks contained the weakly magnetic mineral pyrrhotite. Granitoids, pegmatite granites and pegmatites had low magnetic susceptibilities (0-$10 \cdot 10^{-5}$ units) while the highest values were obtained in amphibolites (50-$100 \cdot 10^{-5}$ units). These differences in magnetic susceptibility likely reflect differences in pyrrhotite content. There was, however, no correlation between pyrrhotite content and concentration of As. Since pyrrhotite is the dominating sulphide in the area, the magnetic properties of the rocks could be measured to trace enrichments of sulphides.

The electrical conductivity of the rocks is enhanced by high contents of sulphides and graphite. Some of the rocks in the high-As supracrustal belt are graphite and pyrrhotite bearing and could perhaps be traced by measurements of the electrical resistivity. The dense infrastructure in the Arlanda area may be a challenge for electrical surveying at above outcrop scale.

5.5 Application of results from the Arlanda area to the bedrock in other parts of Sweden

This study shows that strong enrichments of As (>100 ppm) in the bedrock at Arlanda are restricted to a metasedimentary complex with intercalations of mafic and felsic metaigneous rocks, and in fractionated late kinematic tourmaline bearing pegmatites intruding the complex. Metasedimentary belts with tourmaline bearing pegmatites occur in a semi-continuous belt in the northern Mälardalen region, from the area of Arlanda in the north-east, across the Enköping area and the city of Västerås in the west. This belt is characterised by lithogeogenic As contamination of groundwater and is seen as a prominent anomaly in the map of arsenic contamination of groundwater in tubed wells penetrating the bedrock (Fig. 2). It is likely that the bedrock in the Enköping-Västerås

part of the belt also includes remnants of sedimentary deposits enriched in As. Screening the bedrock for As following the same method as applied at Arlanda would likely be an effective method to narrow down the extent of bedrock units with a high-As content in this region.

5.5.1 Metasedimentary belts in the Sundsvall-Örnsköldsvik region

Medium-metamorphic grade metasedimentary belts in the southern parts of the Bothnian basin (Hietanen 1975; Kumpulainen 2009) between Örnsköldsvik and Sundvall show similarities with the bedrock in the Arlanda-Västerås area. They are mainly composed of metasedimentary rocks originating from marine sediments, intercalated with intermediate to mafic igneous rocks (mainly volcanic; Lundqvist et al. 1997). They are metamorphosed at amphibolite facies conditions and intruded by late kinematic granitoids and pegmatites, the latter commonly tourmaline bearing (Lundqvist et al. 1997; Smeds 1990). The metasedimentary belts are not mineralised but similar to the Arlanda-Västerås area, the region is recognized for a high frequency of As-contaminated bedrock wells (cf. Fig. 1). However, the contamination patters vary and so does the As content of the bedrock (Kumpulainen 2009). Reconnaissance geochemical mapping following the method applied in this study should be an effective tool to identify sedimentary deposits enriched in As.

5.5.2 High-arsenic mafic rocks in a metasedimentary gneiss belt in Sörmland

The county of Sörmland is part of the lithotectonic domain of Bergslagen. Parts of the bedrock includes different sulphide and iron oxide deposits, some associated with enrichments of arsenic minerals (arsenopyrite, löllingite), but part of the bedrock is composed of non-mineralised metasedimentary belts without known mineral deposits (Stephens and Jansson, 2020). Reconnaissance geochemical mapping in search for enrichments of As in a non-mineralised metasedimentary belt between Södertälje and Mariefred was carried out parallel to the investigations at Arlanda (Andersson et al., in press). The mapping was initiated after alarms of high-As concentrations in crushed rock aggregates from a quarry and followed the same method applied at Arlanda. The bedrock has been metamorphosed at higher temperatures than at Arlanda, reaching upper amphibolite to granulite facies conditions (T=780-800 °C; Andersson et al., 2024b). It constitutes high-alumina migmatitic gneisses of a sedimentary origin (stromatic cordierite-garnet-sillimanite migmatite complex) with abundant layers and lenses of metamafic rocks and injections of structurally young granitoids and pegmatites (cf. Stålhös 1982; 1983). Tourmaline bearing pegmatites are not known from the area. Also in this area, reconnaissance geochemical mapping with HXRF followed by bulk rock geochemical analyses proved to be a fast and effective method to identify the main source of As, in this case, high-temperature metamorphic mafic intrusions (Andersson et al. in press). The metamafic intrusions occurred as amphibolites, garnet amphibolites

and mafic granulites. Among these rocks, however, high-As (300 to more than 800 ppm) was encountered in garnet amphibolites and mafic granulites only, and the As concentration varied widely in one and the same rock type. The dominating sedimentary gneisses were, with few exceptions, low in As. This demonstrates that the reconnaissance mapping method using HXRF was effective also when applied to a bedrock with As enriched in specific and subordinate lithological elements only. Similar to the Arlanda area, sampling and petrographic work was intensified in high-As units, but focused on specific lithological elements instead of enrichments in zones. In this case, successful application of the method relied on the ability to distinguish between As-free amphibolites and partly highly enriched garnet amphibolites and mafic granulites. Such work requires geological expertise. It should also be noted that, when harmful elements are enriched in subordinate lithologies only, it is still necessary to analyse all different rock units to avoid sampling of enriched units not representative for the bulk rock matrix.

5.6 Recommendations for surveying, documentation and risk assessment for the occurrence of high-As rocks

Prediction and risk management of high-As rocks depend on an understanding of *where* and *how* As is sitting in the bedrock and it is necessary to know if As enrichment is restricted to a specific lithology, and if so, how much of the rock matrix constitutes this lithology, or if it is zonal and includes several different lithologies. Such basic understanding of the bedrock is important for the sampling strategy, and necessary for the recognition of non-risk rock masses, at the scale of a quarry or a tunnel construction, as well as on a regional scale for physical planning. Hence, surveying of the bedrock must include assessments of where and how As is sitting in the bedrock and the surveying must be conducted by geological expertise.

Understanding the leaching properties of high-As rocks is a central issue for risk management. Leaching tests of As-rich rocks give contradicting results. For example, leaching tests of blasted rocks and drill cuttings during construction of the Rosersberg interchange just south of Arlanda indicated significant leaching (Bidros 2014) and the literature includes examples of release of up to 50% of the total As from rocks by weak acid leaching (e.g., Peters et al. 1999). Other studies point to low degree of leaching (e.g., Parviainen et al. 2015). Differences in the geological materials tested and design of leaching tests, however, makes comparison of results difficult.

It is well documented that the textural context, grain size and the type of As-mineralogy steers the rock leaching properties (Peters and Blum 2003; Bondu et al. 2018). Leaching experiments on pure and intergrown As-minerals also show that the extraction reactions vary widely between different mineral constellations and that they depend on matrix mineralogy (e.g., Igarashi et al. 2008; Lengke et al. 2009, Corkhill and Vaughan 2021).

In the literature on leaching tests of As rocks, however, petrographical and mineralogical information is commonly limited, or even lacking. The Geological Survey of Finland (GTK) has done important pioneering work for risk management in

aggregate production in high-As rocks (Lehtinen and Sorvari 2014; Parviainen et al. 2015). Their guidelines, however, include little information about the geological surveying and the petrography and mineralogy of rock materials investigated.

Application of lab results to natural settings is even more complex as it requires an understanding of both the geological and hydrological setting (Bretzler et al. 2017). Comparison of the mobilization of As from the bedrock in natural settings shows a wide variety in the bedrock geology, including the whole range of igneous, sedimentary, and metamorphic rocks, and different types of mineralisations, in addition to differences in the geochemical environment (e.g., Pili et al. 2013; Dummer et al. 2015; Bretzler et al. 2017; Mazuda 2018; Tabelin et al. 2018; Morales-Simfors et al. 2020; Russell et al. 2021; Maciag et al. 2023; Mukherjee et al. 2014; 2024). This illustrates that As is a highly versatile element and that the understanding of the mobilization of As is site dependent and requires in depth understanding of the local geological and hydrogeological setting (cf. Bundschuh et al. 2012).

In summary, surveying potentially high-As bedrock domains and assessments of risk to encounter high-As lithologies requires detailed lithological, mineralogical and geochemical mapping of the rock matrix. Interpretation of leaching tests and the implication of test results rely on solid documentation of the petrography, and the As- and matrix mineralogy of the material leached. This work requires geological expertise in bedrock mapping, petrology and mineralogy.

This study includes detailed documentation of the occurrence, textural and proportional relations of löllingite and arsenopyrite in different rocks from the Arlanda area, along with detailed documentation of other sulphides and the matrix mineralogy of the investigated rocks and their bulk rock geochemical composition. This information cannot be used to establish thresholds for concentrations of As in the bedrock in direct assessments of the risk for handling the rock masses in construction projects. It is, however, critical in data for leaching tests that will promote the application of results from leaching experiments to the natural environment.

6. CONCLUSIONS

Geochemical mapping of the bedrock in the Arlanda-Rosersberg area shows that significant enrichments of As of >100-1000 ppm, locally up to near 1 wt%, occurs in a ca 1.3 km wide zone inside a metasupracrustal belt in the south-eastern Arlanda area. Such high As concentrations are highly anomalous for a non-mineralised quartz-feldspathic bedrock.

Arsenic concentrations exceeding 100 ppm occurs in metasedimentary, preferentially mica-rich units, in intermediate- to mafic sills and dykes, and in fractionated tourmaline bearing pegmatites. Tourmaline bearing pegmatites appear to be spatially associated with As-enrichments of the country rocks and may be tracers for high-As units. Silica-rich intrusive rocks (granitoids and reworked equivalents) are low risk lithologies

The arsenic is hosted in arsenopyrite (FeAsS) and arsenide (löllingite, $FeAs_2$). Except for quartz-rich intrusive rocks, arsenide was encountered in all investigated lithologies, including mafic to intermediate igneous rocks and metasedimentary rocks. Such widespread occurrence of arsenide has not previously been documented and indicates that the presence of arsenide in the country rocks have been hugely underestimated.

Pyrrhotite ($Fe_{(1-x)}S$) is the most common sulphide in the bedrock in the Arlanda-Rosersberg area. Pyrite (FeS_2) occurs in granitic rocks and is common in structurally young discrete deformation alteration zones together with a low-temperature mineralogy (albite, K-feldspar, chlorite).

Handheld X-Ray fluorescence analysis on outcrops in the field is an effective method for reconnaissance geochemical mapping of As. The method is suitable for strategic selection of samples for targeted whole rock analysis but depends on geological expertise and high frequency of HXRF measurements across the rock exposures including all sub-lithologies.

The methods applied in the surveying of the Arlanda-Rosersberg area are applicable in other parts of Sweden, for example, in non-mineralised metasedimentary belts with tourmaline bearing pegmatites in the Enköping-Västerås and the Sundsvall-Kramfors areas. Arsenic is, however, a highly versatile element and the strategy for surveying the bedrock intimately depends on the geological setting.

In Sweden, there is a close correlation between enrichments of As in the bedrock and As contamination of the groundwater. The geochemistry of the groundwater can be a powerful predictive tool for the recognition of high-As bedrock domains.

Prediction of enrichments of elements harmful to health and the environment in rock masses requires knowledge of *where* and *how* (in what mineral) the elements are hosted in the bedrock. Effective surveying of the bedrock relies on geological expertise in the fields of petrography, mineralogy and structural geology. Such surveying should be performed early in the planning process of construction projects.

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