# ARSENIC IN THE WATER, SOIL BEDROCK AND PLANTS OF THE ESTER DOME AREA OF ALASKA

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## OF THE ESTER DOME AREA OF ALASKA

Arsenic in the water, soil, bedrock adn plants of the Ester Dome area of Alaska

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# ABSTRACT

Concentrations of arsenic as large as 10 ppm (200 times the safe limit for drinking water) occur in the groundwater of a mineralized residential area near Fairbanks. Bedrock of the area contains 750 ppm As, primarily as arsenopyrite and scorodite. The oxygen-poor groundwater is enriched in As(III) and ferrous iron while the surface waters are iron free and contain less than 50 ppb As(V). Arsenic is removed from the water by coprecipitation with ferric hydroxide. Some iron-rich stream sediments contain as much as 1,400 ppm arsenic.

The distribution of arsenic in the groundwater is controlled by the distribution of arsenic in the bedrock. The arsenic content of the B soil horizon over mineralized veins is about 150 ppm, while that over barren rock is 30 ppm. The vegetation over the veins is not significantly enriched in arsenic.

Lettuce, radishes and tomatoes grown with arsenic-rich water (5 ppm) contain 16, 8 and 1 ppm As, respectively; these amounts are significantly greater than plants not treated with arsenic.

Preliminary studies by state and federal health agencies show no detrimental effects on the health of persons drinking these arsenic-rich waters.

## ACKNOWLEDGMENTS

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# AN INTRODUCTION TO THE ARSENIC PROBLEM

Arsenic (As) is a rare element with an average concentration in crustal rocks of about 5 parts per million (ppm = 1 mg/kg). Its abundance in various terrestrial materials is shown in Table 1 (Whiteacre and Pearse, 1974).

TABLE 1:  ARSENIC CONCENTRATION IN VARIOUS ROCKS.

| Material | As (ppm) |
| --- | --- |
| Igneous rocks | 1.5 |
| Sedimentary rocks | |
| Shales | 13 |
| Sandstones | 1 |
| Limestones and dolomites | 1 |
| Pelagic clays | 11 |
| Coal | 25 |
| Metamorphic rocks | 0.4-18 |

Arsenic occurs in more than 100 minerals, but it is a major constituent in only a few of these. Arsenic is present naturally in soils at concentrations ranging from 1 to 100 ppm (Whiteacre and Pearse, 1974; Shacklette and Connors, 1975), with an average concentration of about 5-10 ppm.

In waters free from arsenic pollution, the arsenic concentration is about one part per billion (ppb = 0.001 mg/kg). In waters contaminated by natural or man-made sources of arsenic, the arsenic concentration can range as high as hundreds to several thousands of parts per million (National Academy of Sciences, 1977).

Arsenic is a common, naturally occurring, trace constituent in plants. The concentrations of arsenic in plants depend upon the kind of plant and the quantity of arsenic to which the plant was exposed. On the average, the arsenic content of plants varies from less than 0.01 ppm to about 5 ppm (dry-weight basis), with some plants containing as much as 94 ppm (National Academy of Sciences, 1977).

Arsenic is present in animals and man. Marine fishes may contain up to 10 ppm, while mollusks and some crustaceans contain as much as 128 ppm. Freshwater fishes usually contain less than 1 ppm, although values may reach 3 ppm. Domestic animals and man generally contain less than 0.3 ppm on a wet-weight basis. The total human-body content of arsenic is between 3 and 4 mg, and tends to increase with age (National Academy of Sciences, 1977).

Arsenic is widely used in insecticides, herbicides, dessicants, wood preservatives, feed additives, drugs, war gases and riot-control agents, and in various metallurgical applications. As a result, man contributes to the quantity and distribution of arsenic in the environment. The general cycle of arsenic shown in Figure 1 summarizes the different sources of arsenic and the transfer processes.

Arsenic has long been used as a pharmaceutical but a specific nutritional role of inorganic arsenic has only recently been revealed. A curious feature of arsenic biochemistry is the ability of arsenic to counteract partially the ill effects of another toxic element, selenium. Suffice it to say that the biochemistry of arsenic is complex and differs with different organisms (National Academy of Sciences, 1977).

The toxicological effects on humans from drinking arsenic-rich water is particularly relevant to the present study in which arsenic concentrations ranging from 400 to 21,000 ppb (0.4 to 21 ppm) have been found. Such concentrations, comparable to those in the waters of Ester Dome (near Fairbanks, Alaska) have been encountered elsewhere in the world and their toxicological effects documented. For example, acute and subacute arsenic intoxication occurred in Minnesota from drinking well waters contaminated by pesticides containing 11 to 21 ppm As. In Antefogasta, Chile, skin pigmentation changes and squamous cell carcinoma resulted from long-term consumption of well waters containing around 580 ppb arsenic. Similarly in Taiwan, peripheral vascular disease and skin carcinoma resulted from long-term ingestion of well water containing from 400 to 600 ppb arsenic (Harrington et al., 1978).

Although similar concentrations of arsenic are present in some well waters of the hillside areas around Fairbanks, no clinical abnormalities
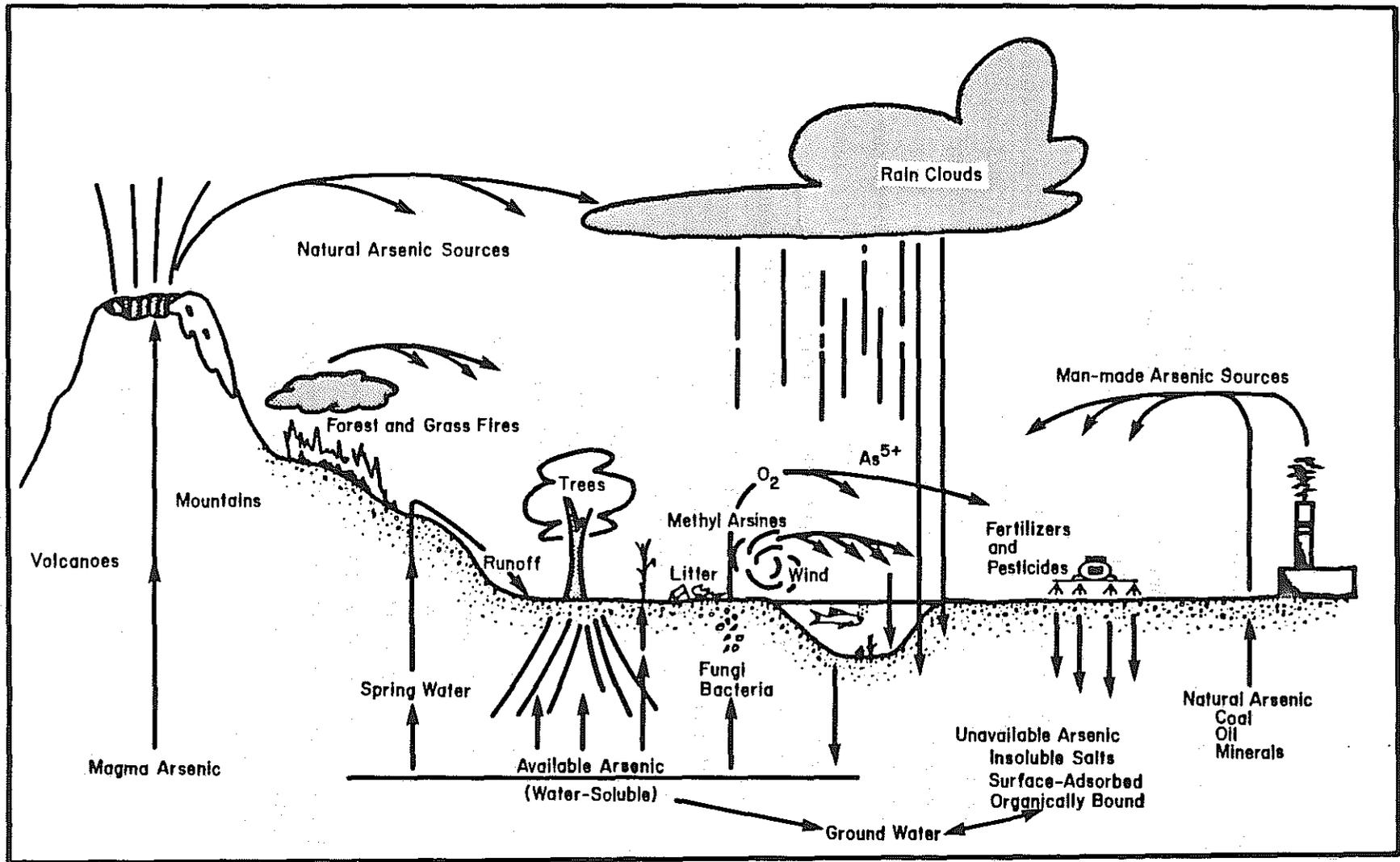
Figure 1.   The general arsenic cycle (adapted from National Academy of Sciences, 1977).

in the human population drinking these waters have been found. Harrington et al. suggest that "higher concentrations or longer times of exposure are required to produce overt clinical effects in a well-nourished U.S. population."

A number of studies bearing on different aspects of arsenic in the environment have been published recently. The list of references at the end of this section is not complete but serves as an introduction to the extensive literature on arsenic.

<div align="center">PREVIOUS WORK</div>

The earliest work on arsenic in waters of the Fairbanks area began in 1974 as an outgrowth of a joint University of Alaska and U.S. Geological Survey Heavy-Metals Project carried out from 1967-1970 (see, for example, Stevens et al., 1969). This project studied the gold mineralization in the Fairbanks District, where gold is accompanied by arsenic mineralization in soils and bedrock. The extent of arsenic enrichment in the area revealed by the study led one of us (DBH) to postulate that arsenic may be present in water of the Fairbanks area.

As a result, a study was begun (Wilson and Hawkins, 1978) to investigate arsenic concentrations in the surface and groundwaters of the Cleary Summit -- Pedro Dome area near Fairbanks. This study showed that surface waters and particularly stream sediments of certain streams were enriched in arsenic. A limited number of wells were sampled and analyzed for arsenic, three of which showed arsenic concentrations slightly in excess of 50 ppb -- the U.S. Public Health Service (USPHS) recommended concentration limit for arsenic in domestic waters.

A subsequent study of the waters of the Ester Dome area was undertaken by Wilcox (Johnson et al., 1978). This area has gold-arsenic mineralization and also is a developing residential area where the water supply is drawn from domestic wells. She showed that concentrations of arsenic as large as 3 ppm were present in domestic well waters. State and federal health authorities were notified, resulting in an extensive water-testing program (Johnson et al., 1978). As a result, a number of domestic wells in the Ester Dome area and other hillside residential areas around Fairbanks were found to contain arsenic in excess of the USPHS concentration limits.

These findings sparked a joint investigation by state and federal health authorities (Harrington et al., 1978). This study showed that persons drinking arsenic-enriched waters were themselves enriched in arsenic. No detrimental effects to the population were evident but, because the effects of chronic exposure to small concentrations of arsenic are not well known, a subsequent study by the Federal Center for Disease Control and the University of Boston's Medical School was undertaken in the summer of 1978. In this study, numerous people were examined for possible neurological damage resulting from ingesting arsenic. The results of this study are being evaluated.

Since 1976, geological and geochemical studies have been continued by the Institute of Water Resources and the Geology-Geophysics Program, at the University of Alaska. A Student-Initiated National Science Foundation grant was obtained by Burton et al. (1978) to investigate arsenic in soils, plants and waters of the Ester Dome area and, in particular, to examine the effects of mining activities on the arsenic concentrations of surface waters and groundwaters. Burton et al. suggested that lode-gold mining activities may contribute to the arsenic burden of streams draining the area where mining is being carried out. They also studied the uptake of arsenic by various plants native to the region. They concluded that Labrador tea (*Ledum* spp.) took up arsenic from the soil and that the arsenic content of the ash from this plant might be indicative of arsenic-bearing mineral assemblages in the bedrock.

THIS STUDY

The present study attempts to answer questions of geological and geochemical nature that were raised as a result of preceding work.

o    What are the sources of arsenic in the waters of the Ester Dome area?

o    What governs the behavior of arsenic in the groundwater?

o    Why are there such strong, lateral concentration gradients present in the groundwater?

o  What processes govern the behavior of arsenic in the surface
   waters?
o  Can areas of probable arsenic contamination of the groundwater
   be predicted on the basis of geology?
o  Does native vegetation become enriched in arsenic as a result
   of growing in arsenic-rich soil?
o  Do garden vegetables become enriched in arsenic as a result of
   being irrigated with arsenic-rich water?
o  Are there methods that can be used by homeowners to remove
   arsenic from their water supplies?

This study is divided into two segments:  the geologic aspects of
arsenic in the Ester Dome area; and the geochemistry of arsenic.


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# GEOLOGY AND LODE DEPOSITS OF ESTER DOME

# INTRODUCTION

To understand the geology of a small area such as Ester Dome, it is necessary to see how the geologic features of the small area fit into the general geologic fabric of a much larger region. This section presents an overview of the regional geology of the Yukon-Tanana Uplands before focussing on the geology of Ester Dome. This permits an interpretation of the geology of Ester Dome that is compatible with the geology of the surrounding region.

# REGIONAL GEOLOGY

## Petrology and Structure of the Crystalline Basement Rocks

The Yukon-Tanana Uplands are underlain by crystalline rocks of the Yukon-Tanana Complex (Foster et al., 1973). The oldest rocks in the complex are schists and gneisses which were formerly known as the Birch Creek Schist Formation (Spurr, 1898). Previously, this sequence of metamorphic rocks was believed to be of Precambrian age (Mertie, 1937), but recent work has shown that the Yukon-Tanana metamorphic terrane is composed of at least two sequences of metamorphic rocks, which include parental rocks of differing ages.

The metamorphics have been intruded by granitic rocks ranging in composition from granodiorite to granite. Some of these masses, including the Charley River and Mt. Harper plutons are of batholithic dimensions. The schists and gneisses have also been intruded by small mafic and ultramafic plutons, including diorite, gabbro, hornblendite, and periodotite.

Outcrop patterns of the metamorphic rocks in the Fairbanks and Big Delta districts form several northeast-trending belts (Figure 2). One belt of highly deformed greenschist-facies rocks characterized by slate and calc-phyllite is traceable through the central part of the Fairbanks district. To the southeast, a zone of cataclastic rocks separates two amphibolite-facies schist-terranes. The amphibolite-facies rocks include biotite schists and various gneisses. The fabrics indicate that
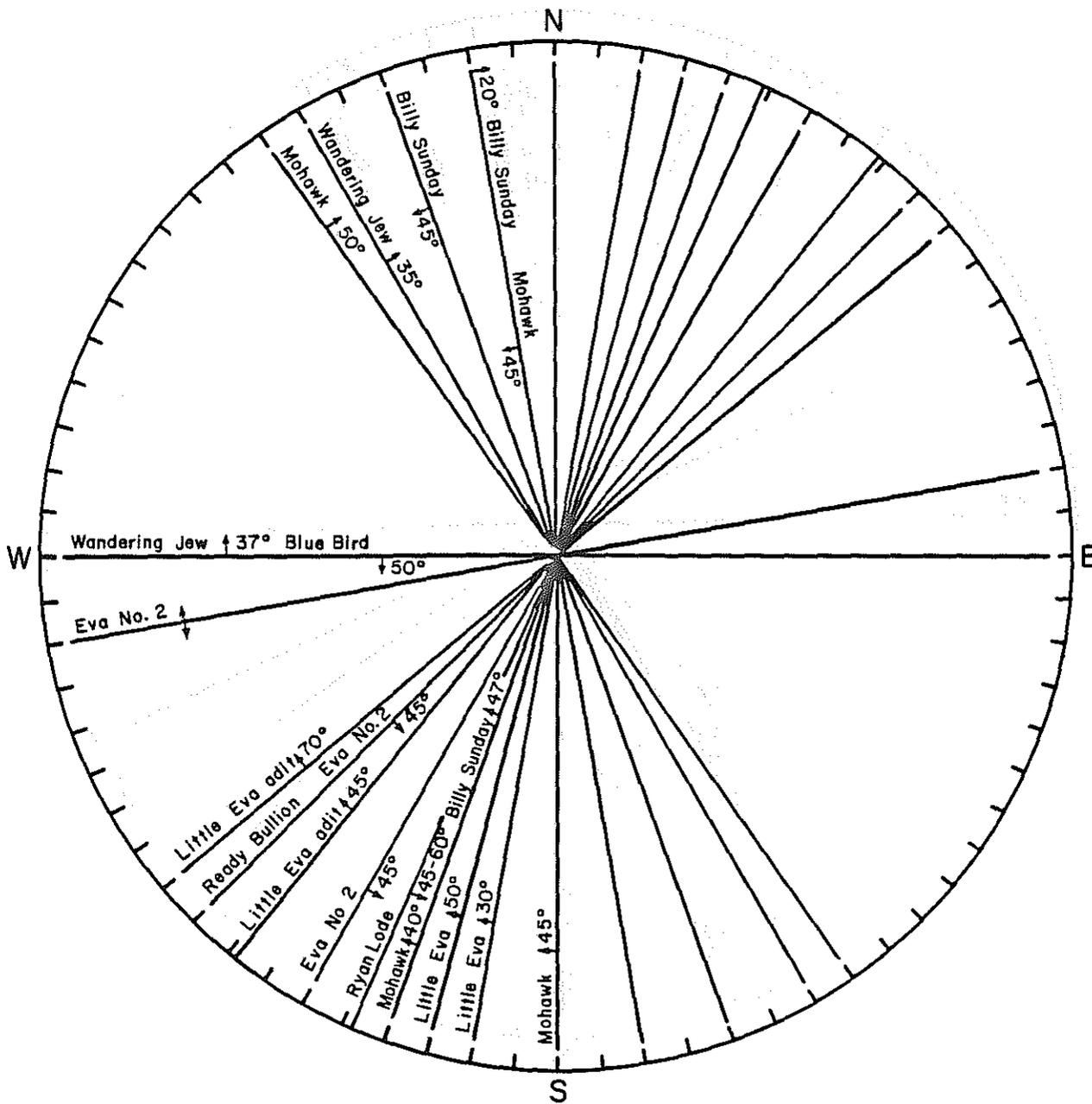
Figure 2A.  Orientation of veins in the Ester Dome area (adapted from
Hill, 1933).
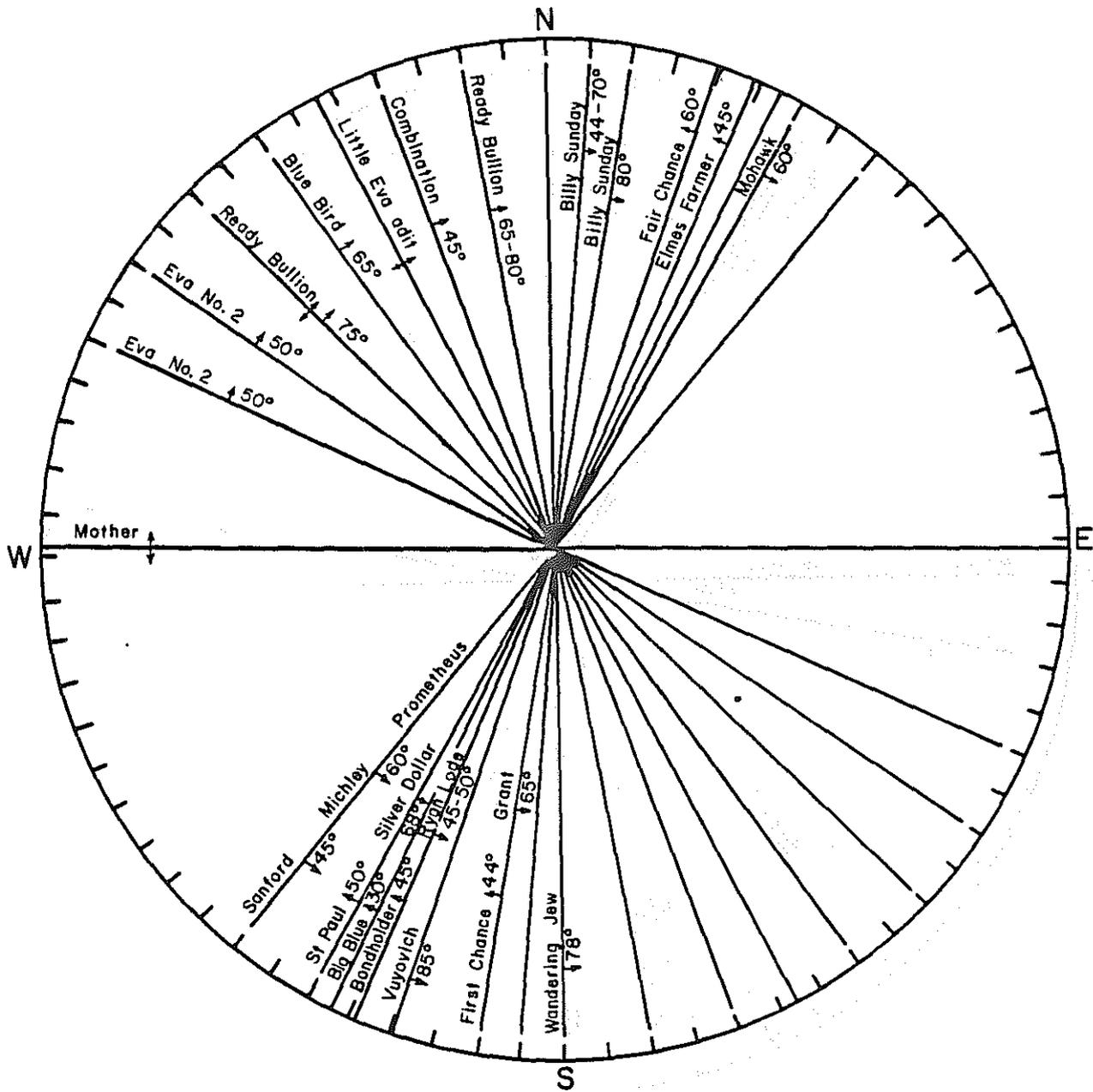
Figure 2B. Orientation of faults in the Ester Dome area (adapted from Hill, 1933).

these rocks have a polymetamorphic history, and that amphibolite-facies metamorphism was followed by a retrograde event that resulted in the formation of assemblages belonging to the greenschist facies in some areas.

The north margin of the greenschist-facies belt grades into mica schists transitional between greenschist and epidot-amphibolite facies. In this zone of metamorphic rocks (Figure 2), Prindle (1973) discovered an eclogite-bearing terrane about 15 mi (24 km) north of Fairbanks. Recent studies of these rocks by Forbes et al. (1968), and Swainbank and Forbes (1975), indicate that younger pelitic schists and subordinate greenschists have been thrust over the older and more highly deformed eclogitic rocks.

The thrust zone, where exposed along the Elliott Highway, is defined by coarse-grained garnetiferous calc-mylonites, which appear to be cataclastically deformed, and by retrograded calc-silicate rocks. Eclogitic rocks intercalated with calcium-magnesium schists are characterized by the assemblage clinopyroxene-garnet-carbonate-sphene (with or without quartz). The structural style is isoclinal-overturned, with northwesterly trending fold axes. This is a trend which is discordant to the northeasterly trending fold axes of the schists that compose the upper plate. The northwesterly trending folds of the lower plate were subsequently refolded into open synclines and anticlines along the younger northeasterly trend.

## Metamorphic and Intrusive History

To date, none of the potassium-argon (K-Ar) dating done on metamorphic rocks from the Uplands has produced a valid Precambrian crystallization age. Hornblende K-Ar data indicate an initial cooling age of 180-190 million years for amphibolite facies rocks from the Fairbanks, Fortymile, Delta and Eagle quadrangles. This earlier apparent age has been perturbed in some areas by a younger thermal event which is reflected in potassium-argon mica ages ranging between 90 and 120 million years.

The later thermal event is probably related to the widespread emplacement of granitic plutons during this same time period.

The eclogite-bearing schist terrane, which is exposed as a structural window north of Fairbanks, has yielded an amphibole potassium-argon age of 470 ± 35 million years (MY).

Based on radiometric age data and the above evidence, the metamorphic terranes of the Yukon-Tanana Complex were derived from parental rocks ranging in age from late Precambrian to late Paleozoic.

## MESOZOIC SEDIMENTARY AND VOLCANIC ROCKS

### Sedimentary Rocks

Fossiliferous Mesozoic sedimentary rocks have not been found in the southeastern part of the Yukon-Tanana Uplands, although a re-entrant of Cretaceous argillaceous sediments has been mapped in the Livengood District, northwest of Fairbanks.

### Volcanic Rocks

Small areas of tholeiitic basaltic rocks, including pillow basalts, subaerial flows and breccias occur in the Fairbanks District. These rocks are characteristically preserved in downthrown fault blocks. Preliminary potassium-argon age determinations indicate that these basalts are of late Cretaceous age. Silicified mata-sequoia trunks and branches have been discovered in a fossil soil zone between the schists and the overlying basalts, supporting an early Cretaceous age for the basalts.

Basalt also occurs as cross-cutting dikes throughout the Uplands. Based on compositional similarities to those in the Fairbanks District and minimal alteration, they are probably of late Cretaceous or early Tertiary age.

# TERTIARY SEDIMENTARY AND VOLCANIC ROCKS

## Sedimentary Rocks

Tertiary sediments including siltstones, sandstones and conglom-
erates occur as small, isolated outcrop areas in the Yukon-Tanana
Uplands. All of the known occurrences are of continental origin, and
some of the preserved sections contain coal measures and/or basal ba-
salts. The lithology and stratigraphy of these sections are similar to
that of the coal-bearing Tertiary on the north flank of the Alaska
Range.

## Volcanic Rocks

A large Tertiary volcanic field dominated by felsic volcanic rocks
including welded tuffs has been mapped in the Tanacross Quadrangle by
Foster (1970). The volcanic suite is similar to that of the Tertiary
volcanic fields of the Great Basin, or the circum-Pacific island arcs.
Possible caldera structures have also been described by Foster (1970).

# GLACIAL HISTORY

Although there is fragmentary evidence which suggests that glacia-
tion in interior Alaska may have been initiated in late Miocene or
Pliocene time, moraines and outwash of the Delta (Illinoian) Glaciation
mark the onset of glaciation in the northeastern part of the Alaska
Range (Pewe and Holmes, 1964; Holmes and Foster, 1968). Delta Glacia-
tion, following the Sangamon interglacial interval, was followed by the
Donnelly (Wisconsin) Glaciation.

Both Illinoian and Wisconsin glaciations have left a record in the
northeastern Alaska Range and the Yukon-Tanana Uplands. Glaciation in
the Uplands was restricted to local alpine glacial systems which devel-
oped in the higher hills, and included relatively small distributary

valley glaciers; however, valley glaciers descending the north slope of
the Alaska Range reached the flood plains of the Tanana, Delta and Tok
rivers, as documented by moraines and glacio-fluvial deposits which have
been identified and mapped by Pewe and Holmes (1964), and others.

Glacial advances were accompanied by a great increase in the amount
of suspended sediment brought into the master streams by tributaries
descending the north slope of the Alaska Range. As a result, the
Tanana, Delta and Tok rivers aggraded their channels, causing lateral
migration and meandering across the valley floor at a rate which was too
rapid for the development of vegetation on the valley floor. Therefore,
southerly winds were able to pick up a large quantity of silt from the
floodplains, and deposit it in the uplands to the north as "loess."

## Alluvial and Colluvial Deposits

During Holocene time (beginning 10,000 years ago), tributary streams
in the uplands have initiated a new cycle of downcutting. The master
streams are no longer overladen with glacial debris, and geodetic mea-
surements indicate that the Tanana Valley is subsiding, and that the
Alaska Range is undergoing relative uplift. Under these conditions,
glacial deposits are being reworked by the master streams in the flood-
plains, and colluvial deposits are developing in the uplands.

## Permafrost

During Wisconsin time, the $0^{o}C$ isotherm may have reached depths
down to 1,200 ft in the Yukon-Tanana District. During the last two or
three thousand years, a warming trend has initiated a long-term thawing
cycle. Currently, the permafrost table is deepest on north-facing
slopes, and in alluvial fill covered by vegetation in steep-walled
valleys. Broad valleys, floored by floodplain deposits are underlain by
discontinuous permafrost. South-facing slopes are generally free of
permafrost.

# THE GEOLOGY AND STRUCTURE OF ESTER DOME

## Crystalline Basement Rocks

Ester Dome is underlain by metamorphic and igneous rocks of the Yukon-Tanana Complex, including micaceous quartzites, quartz-mica schists (with and without garnet), calc-schists and subordinate green-schists, amphibolites and impure marbles. The metamorphic rocks have been intruded by dikes and small plutons of biotite-granodiorite-quartz monzonite. However, there is no proved association between these particular intrusives and mineralization on Ester Dome.

## Structure of the Crystalline Basement Rocks

Structural measurements including the attitudes of bedding, folia-tion, fold axes and mineral lineations indicate that the metamorphic rocks of Ester Dome have been deformed into a structural high with the geometry of an asymmetric anticlinorium (Figure 3).

Divergent plunge patterns of small fold axes, with an apex near the topographic summit of Ester Dome, suggest that the structure may actu-ally be an asymmetric dome. Additional evidence for a domal structure is offered by the break in continuity of both rock type and structural axes, between Ester Dome and the hills northeast of the Goldstream Valley. An alternate explanation could be supplied by offset due to faulting along the trace of the valley, or a combination of doming and faulting.

The amphibolites, coarse-grained garnet-mica schists and marbles that are exposed in the Murphy Dome road cuts (on the southwest slope of Goldstream Valley), have not been found in outcrop or in the subsurface mine workings on Ester Dome. However, this sequence of rocks does occur along the crest of Chena Ridge, south of the Ester-Cripple Creek Valley.
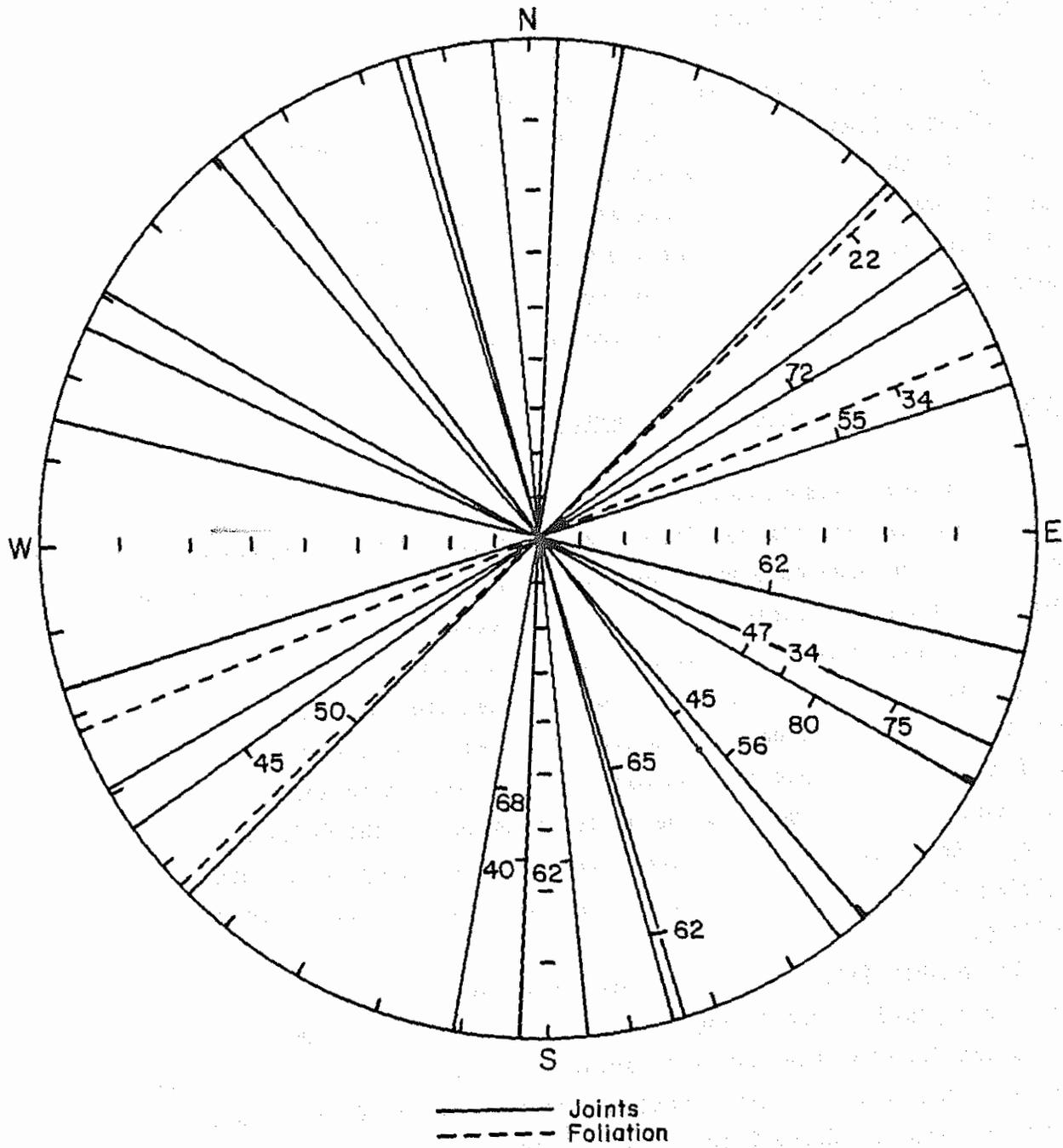
Figure 3.  Orientation of joints and the attitude of the foliation of host micaceous quartzites on the southeastern flank of Ester Dome.

## Surficial Deposits

The valleys descending the flanks of Ester Dome contain alluvial sands and gravels deposited by ancestral streams which were responsible for valley downcutting during Quaternary time. Most of the steep valley sides are underlain by colluvium which interfingers with the alluvial sands and gravels on the valley floors.

Wind-blown silt (loess) blankets the older colluvial and alluvial deposits, and it is mixed with organic material in the valley fill. Valley fill is perennially frozen on all exposures around the Dome. The loess blanket is thicker on the south slopes, and much thinner on the north (lee) flank of the Dome.

Surficial deposits on south-facing ridges are usually thawed, but loess and colluvium on north exposures are perennially frozen.

## Groundwater Geology

The loess blanket that overlies bedrock, colluvium and alluvium is relatively impermeable. Therefore, water wells driven into the upper slopes of Ester Dome obtain water from fracture systems in bedrock, as porosity is practically nil in the highly recrystallized metamorphic rocks. A few producing wells may obtain water from colluvial fans on valley sides, but the dominant aquifer at the higher elevations is fractured bedrock. Fracture systems are more highly developed in the more competent and brittle rock units (e.g. quartzites) rather than the more highly schistose rock types (e.g. mica schists).

In recent years, dwellings have been constructed on placer tailings in the Ester-Cripple and Sheep Creek valleys, and water is also being obtained from wells driven down to the contact zone between the base of the tailings and underlying bedrock.

Artesian systems are unknown in any of the above settings, and it is reasonable to conclude that groundwater recharge is dependent on the downward percolation of water from local rain and snowmelt into the aquifers. Currently, we do not have hard data on the recharge rate and draw-down characteristics of these aquifers.

# MINERALIZED ZONES AND VEIN SYSTEMS

## Lode Classification and Distribution

The gold-bearing quartz lodes of Ester Dome are similar to those that occur elsewhere in the Fairbanks gold belt, which extends northeast from Ester Dome to Pedro Dome and the Cleary Summit area. Although contact metasomatic deposits are known to occur around the margins of the quartz monzonite plutons on Gilmore Dome and Pedro Dome, and fissure sulfide veins were encountered in the Busty Belle adit on Pedro Dome, lode deposits in the Fairbanks gold belt are dominantly fissure-controlled or fault-controlled metalliferous quartz veins in micaceous quartzites and quartz-mica schists. Recent work also suggests that strata-bound sulfide concentrations also occur in the Pedro Dome -- Cleary Summit area, which were formerly interpreted as replacement deposits (P. Metz, personal communication). The existence of this type of deposit may be relevant to the arsenic problem, and the implications are discussed in more detail in a later section.

Although the lode deposits appear to be spatially related to granitic stocks and dikes in the Pedro Dome -- Cleary Summit area, this association is not obvious on Ester Dome, unless the plutons are unde-- tected in the subsurface.

## Mineralogy of the Lodes

Sandvik (1967), progressing beyond the earlier work of Hill (1936), determined that the "introduction of vein minerals took place in four phases, each preceded by tectonic activity which prepared the ground for the passage of hydrothermal solutions." Sandvik's proposed sequence of mineralization is shown in Table 2.

TABLE 2:  SEQUENCE OF MINERALIZATION IN FAIRBANKS
DISTRICT GOLD-BEARING LODES, AS PROPOSED BY SANDVIK (1967).

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Phase 1 - Quartz (barren of metallic minerals)

Phase 2 - Quartz, loellingite, arsenopyrite, pyrite, and
          gold

Phase 3 - Quartz, pyrite, gold, sphalerite, chalcopyrite,
          freibergite, jamesonite, galena, boulangerite,
          robinsonite, zinkenite, and stibnite

Phase 4 - Quartz and stibnite

Following Lindgren's depth-temperature classification, Phases 1 and 2
are high-temperature mesothermal, Phase 3 is low-temperature mesother-
mal, and Phase 4 is epithermal.

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High grade (bonanza) gold-quartz lodes on Ester Dome are usually
associated with arsenopyrite (FeAsS) and/or loellingite (FeAs$_2$), pyrite
(FeS$_2$) and one or more of the sulfosalts such as jamesonite (Pb$_4$FeSb$_6$S$_{14}$),
freibergite (Cu, Fe, Ag)($_{12}$Sb$_4$S$_{13}$), boulangerite (5PbS·2Sb$_2$S$_3$), robisonite
(7PbS·6Sb$_2$S$_3$) and zinkenite (PbS·Sb$_2$S$_3$).  Stibnite (Sb$_2$S$_3$) is also
associated with gold-quartz lodes, but it usually occurs as lens-shaped
masses along the hanging or foot walls of the lodes, and it appears to
represent the latest phase of mineralization.  Galena (PbS), sphalerite
(ZnS) and chalcopyrite (CuFeS$_2$) have also been reported in Ester Dome
lode assemblages, but they are relatively rare.

Stibnite-quartz lodes also occur on Ester Dome; they contain
little gold and accessory sulfosalts.  These lodes (e.g. McQueen Mine;
Stibnite Lode Mine) are highly brecciated and sheared, and appear to
have been deposited along fault or shear zones.

Arsenic Content of Stibnite and Secondary Alteration Products

Sandvik (1964), in his investigation of the trace metal content of
stibnite in the Fairbanks District, found that Pb, As, Bi, Ag and Cu are
frequently present in relatively high total concentrations (up to 3,000
ppm).  Stibnite from the Ester Dome lodes contains an average of

1,900 ppm arsenic, and is the probable source of scorodite ($FeAsO_4 \cdot 2H_2O$),
a secondary arsenic mineral which also occurs as an alteration product
of arsenopyrite (FeAsS) and loellingite ($FeAs_2$). Stibnite in some lode
deposits is almost completely replaced by secondary minerals including
cervantite ($Sb_2O_4$), stibiconite ($Sb_3O_6 \cdot OH$), senarmontite ($Sb_2O_3$), berme-
site ($Sb_2S_2$) and scorodite.


## STRUCTURAL CONTROL OF MINERALIZED LODES

### Distribution and Orientation of Lodes

As previously noted by earlier workers, most of the Ester Dome
lodes are located on the southeast flank of the Dome. The most inten-
sive concentration of lodes defines a northeast-trending mineralized
zone extending from the divide between Moose and Ready Bullion Creeks to
the hills forming the Happy Creek watershed (see Map 1). The lodes in
this belt have a dominant northeasterly strike, dipping to both the
northwest and southeast.

A second trend of mineralized lodes is characterized by northerly
and northwesterly trending vein systems, which can be traced from the
Eva Creek area over the west ridge of Ester Dome and down into the head-
waters of Nugget Creek. Northerly and northwesterly trending fractures
and fault zones seem to have favored stibnite deposition, and appear to
have formed subsequent to the northeasterly trending fractures.

### Large-Scale Structural Controls

The lodes of the northeasterly trending Ester-Happy zone have been
emplaced on the southwest and descending limb of an asymmetric anti-
clinorium (Figure 3). Although a local reversal occurs around a major
fold axis, layering and foliation strike consistently to the northeast,
and the dip is dominantly to the southeast.

There is a rather consistent discordance between the strike of
minor fold axes and the dip of foliation, indicating that the episode
which produced the northeasterly trending structures was preceded by an

earlier deformation producing folds around more westerly trending axes. Mineralization appears to have followed the more recent deformation.

The lodes of the Ester-Happy zone (including the Ryan Lode system) are dominated by northeasterly trending veins, which parallel the strike, but are usually discordant to the dip of layering and foliation.

Large-scale structural controls of the northwesterly trending Eva Creek -- Nugget Creek zone are more elusive, since the lodes appear to transect various bedrock attitudes, and the terrane appears to be dominated by northwesterly rather than northeasterly trending fractures and faults.


## Fracture, Joint and Fault Control

Hill (1933) was the first to investigate the possible correlation between faults and lode orientation on Ester Dome. Figure 2, taken from Hill, shows an apparent correlation between fault and vein orientation.

However, Figure 3 (a diagram which plots the orientations of unmineralized and open joints, and the strike and dip of foliation of the host quartzites and micaceous quartzites) presents a strong case for joint control of lode orientation. The diagram clearly defines a strike-joint set, and cross joints that are both normal and inclined to the northeasterly strike of foliation and fold axes. In fact, the joint orientation diagram reaffirms the validity of the "strain ellipsoid," including the presence of tensional joints in a zone approximately $45\frac{1}{4}$ degrees to the fold axes.

The lodes of the Eva-Nugget trend appear to be related to tensional joints, while some of the lodes along the Ester-Happy zone may be strike joint and/or fault controlled.

The joint system illustrated in Figure 3 shows that the more competent rock units are pervasively fractured and that such units in thawed terranes will have excellent fracture permeability, which will accelerate the down-dip movement of groundwater. Highly schistose units will have lower down-dip permeability (Figure 4).
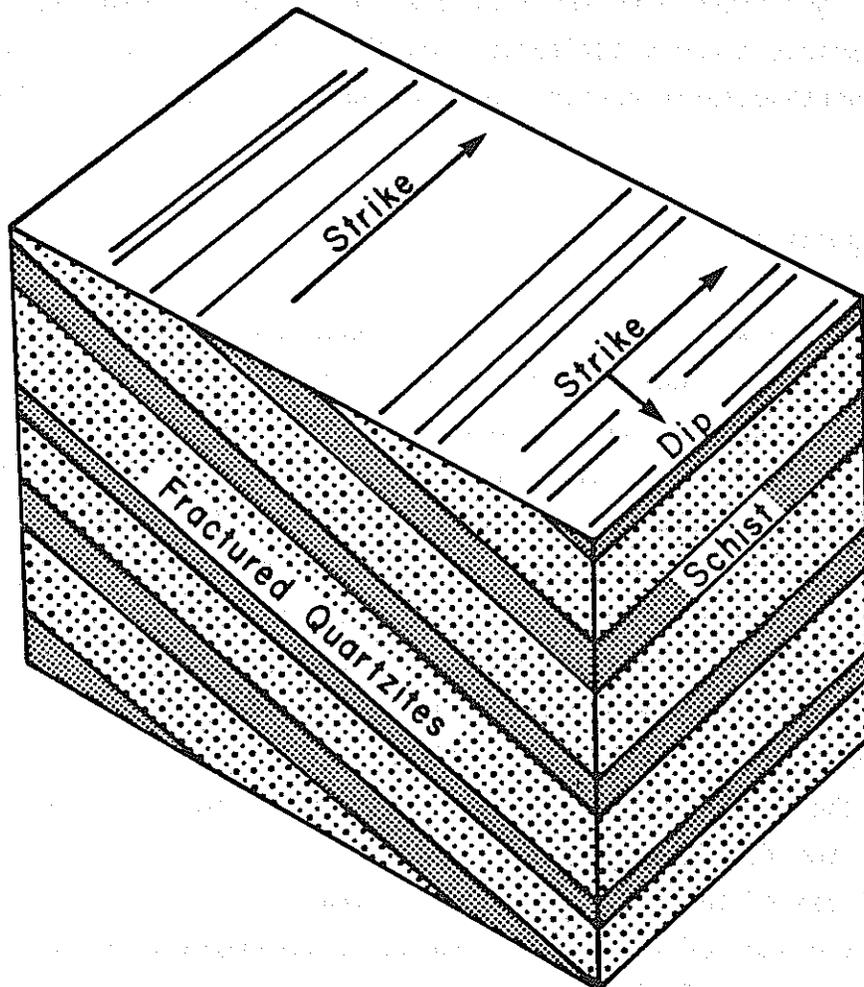
Figure 4.   Idealized block diagram illustrating
down-dip fracture permeability controlled
by joint sets.

## Lode and Auger Sample Concentrations

Map 2 identifies Ester Dome lode deposits and outcrop and auger samples that contain anomalous concentrations of arsenic-bearing mineral phases, including stibnite (Sandvik, 1967). The map patterns clearly show that over 90 percent of all of the known lode deposits on the southeastern slope of Ester Dome contain arsenic-bearing mineral phases, and that many grab and subsurface auger samples from the extension of the Ester-Happy zone (taken from ridge traverses and the Ester Dome road; Stevens et al., 1969) produce anomalous gold and covariant arsenic values (Table 3).

The covariant arsenic/gold anomalies documented in Table 3 and plotted in Map 2, show that the Ester-Happy lode zone is signalled by an arsenic-gold signature that can be traced to the northeast beyond the recognized occurrence of mineralized lodes.

## Influence of Arsenic-bearing Lodes and Host Rocks on Groundwater

The maps and data discussed above clearly show that the Ester-Happy lode zone is enriched in arsenic-bearing mineral phases, and that the surrounding host rocks carry anomalous concentrations of arsenic. This very wide zone of metallic enrichment, which includes lodes and inter-calated mineralized schists and micaceous quartzites, carries highly anomalous concentrations of arsenic. Considering the down-dip struc-tural setting (see Figure 4), and the well-developed fracture perme-ability of the more competent rock units, waters moving through this zone could be expected to develop high concentrations of arsenic miti-gated by chemical constraints including water temperature, Eh and pH.

TABLE 3: ARSENIC AND GOLD ASSAYS OF SURFACE (GRAB) AND DOWNHOLE SAMPLES TAKEN FROM ESTER DOME (ONLY ANOMALOUS ANALYSES ARE INCLUDED).

| Sample No. | Sample Interval (ft) | Arsenic (ppm) | Gold (ppm) | Sample No. | Sample Interval (ft) | Arsenic (ppm) | Gold (ppm) |
|---|---|---|---|---|---|---|---|
| ESTER DOME ROAD | | | | 107 | Surface | 1,750 | 0.045 |
| | | | | 108 | " | 1,730 | <0.02 |
| 27A | 3-6 | 1,430 | <0.05 | 109 | " | 2,060 | <0.02 |
| | 6-9 | 1,050 | <0.02 | 112 | " | 1,830 | 0.60 |
| | 9-10 | 950 | <0.05 | | | | |
| | | | | 113A West | Surface-2.5 | 1,400 | 0.26 |
| 32A | 3-6 | 1,150 | 0.11 | | 2.5-4 | 840 | 0.03 |
| | 6-11 | -- | <0.02 | | | | |
| | >11 | 950 | 0.11 | 113A East | 0-2.5 | 1,550 | 0.21 |
| | | | | | 2.5-5 | 1,350 | 0.26 |
| 42A | 2.5-3.5 | 2,250 | 0.77 | | | | |
| | 3.5-4.0 | 1,900 | 0.17 | 118 | Surface | 1,575 | <0.02 |
| | | | | 130 | Surface | 1,380 | 0.11 |
| 46A | 1.5-7 | 2,150 | 0.21 | 142A | 3 | 1,820 | 0.23 |
| | | | | 144 | Surface | 500 | 0.89 |
| 49A | 2.5-4.5 | 1,150 | 0.60 | 157A | 11-12.5 | 940 | 0.35 |
| | 4.5-10 | 940 | 0.12 | 166A | Surface | 1,050 | <0.05 |
| | 10-14 | 1,050 | 0.33 | 168A | 42-50 | 1,240 | 0.09 |
| | 14-16 | 1,600 | 1.35 | 172A | 30-32 | 1,430 | <0.02 |
| | | | | 176A | 48-49 | 1,350 | <0.02 |
| 86A | 2-6 | 3,170 | 2.80 | | | | |
| | 6-7 | 4,500 | 0.08 | HENDERSON ROAD | | | |
| 90A | 4-5.5 | 1,050 | <0.08 | 1A | 18-23 | 1,270 | 0.20 |
| | 5.5-7.5 | 1,140 | 0.25 | | 23-25 | 1,175 | 0.12 |
| | 7.5-9 | -- | 0.15 | | | | |
| | 9-13 | 940 | 0.15 | 7A | 15-17 | 1,200 | 0.14 |
| | 13-15.5 | 945 | <0.02 | 10A | 10-13 | 1,160 | 0.08 |
| | 15.5-17 | -- | <0.02 | | | | |
| | 17-19 | -- | 0.11 | GRAB SAMPLES FROM ESTER DOME OUTCROPS | | | |
| | 19-23 | 1,230 | 0.14 | | | | |
| | 23-24.5 | -- | 0.23 | ED9-1-1 | Surface | 940 | 0.06 |
| | 24.5-27 | -- | <0.02 | ED9-1-3 | " | 1,150 | 0.64 |
| | 27-29 | -- | <0.02 | ED9-1-4 | " | 1,220 | 0.11 |
| | | | | ED9-1-6 | " | 970 | 0.20 |
| 95 | Surface | 3,250 | 0.15 | ED9-1-12 | " | 960 | 0.20 |
| 97A | 2-5.5 | 1,030 | 0.07 | ED9-1-17 | " | 800 | <0.02 |
| | | | | ED9-1-21 | " | 2,620 | 1.46 |
| 101A | 4-14 | 1,700 | 0.35 | ED9-1-22 | " | 2,560 | 162.0 |
| | 10-14 | 2,050 | 0.22 | | | | |

a. From Stevens et al. (1969).

b. Emission spectrography. Detection limit: 500 ppm. Mean value estimated assuming samples with arsenic values less than the detection limit contained 250 ppm As.

c. Emission spectrography. Values reported only for those samples with arsenic values greater than 500 ppm.

d. X-ray fluorescence analysis of well cuttings. See Appendix D for a description of the analytical method. Analytical detection limit: 40 ppm.

e. Metamorphic rocks, unspecified.

# GEOCHEMISTRY OF ARSENIC

## ARSENIC IN SURFACE WATERS AND STREAM SEDIMENTS

Several streams on the eastern side of Ester Dome were sampled during August 1978. Sample localities are shown in Map 2 and the arsenic content of surface-water and stream-sediment samples are given in Table 4.

Data for pH, temperature, dissolved oxygen and specific conductance were obtained with a HORIBA Water Quality Meter, the probe of which was immersed in the flowing stream at the sample locality. The arsenic concentration of the water was determine by atomic absorption spectrometry (see Appendix A for method) following filtration through a 0.45 μ Millipore filter and acidification of the sample in the field. The arsenic content of the sediment samples was determined by x-ray fluorescence analysis of the total sample (see Appendix A).

A stepwise multiple-regression analysis (BMDP-2R; Dixon & Brown, 1977) was performed on the data of Table 4 to determine if the arsenic content of the stream water was related to the other variables measured. Sample 21 (from a seep draining a mineralized vein) was not included in this analysis because this is an exotic sample, not typical of the surface waters as a whole.

Based on multiple regression analysis, there was no significant relationship among the arsenic content of stream water, the arsenic content of the bottom sediments, pH, dissolved-oxygen content and specific conductance of the stream water.

Wilson and Hawkins (1978) observed a significant positive correlation between the arsenic content of stream water and that of the bottom sediments. The lack of agreement between these two studies may stem from the fact that Wilson and Hawkins studied streams which were very muddy while those of the present study were clear. Also -- and perhaps more important -- because of the heavy suspended sediment load, Wilson and Hawkins were unable to filter the samples in the field. Their samples were acidified and then filtered in the laboratory. This treatment led to some enrichment in arsenic of the waters by leaching from the suspended sediments, as was discussed by them.

TABLE 4: ARSENIC CONCENTRATION IN BOTTOM SEDIMENTS AND WATERS OF ESTER DOME.

| Sample | Date | pH | Water | | | | | Sediment |
|---|---|---|---|---|---|---|---|---|
| | | | Temp. $^{o}C$ | D.O. | Sp. Cond. | As (ppb) | | As (ppm) |
| Nugget Cr. 1 | 7/25/78 | 6.7 | 1.8 | 13.1 | 47 | <10 | | 123 |
| Nugget Cr. 2a | 7/25/78 | 6.8 | 4.9 | 11.5 | 93 | <10 | | 147 |
| Nugget Cr. 2b | 7/25/78 | 6.7 | 6.3 | 8.4 | 192 | --- | | 888 |
| Ready Ballion Cr. 8 | 7/25/78 | 7.3 | 10.0 | 12.9 | 81 | 14 | | 876 |
| Ready Ballion Cr. 9 | 7/25/78 | 7.5 | 3.6 | 13.1 | 47 | 14 | | 1,304 |
| Ready Ballion Cr. 11 | 7/25/78 | 7.2 | 17.9 | 9.2 | 440 | 20.37 | | 1,451 |
| Sheep Creek 12 | 7/27/78 | 6.5 | 4.9 | 11.5 | 64 | <10 | | 350 |
| Sheep Creek 13 | 7/27/78 | 6.1 | 8.1 | 10.3 | 157 | 30 | | 695 |
| Eva Creek 15 | 8/8/78 | 6.6 | 9.1 | 11.6 | 208 | 57 | | <40 |
| Eva Creek 16 | 8/8/78 | 6.6 | 7.9 | 9.9 | 202 | 74 | | 363 |
| Eva Spring at Clipper Mine 17 | 8/8/78 | 6.4 | 5.9 | 5.1 | 47 | <10 | | 228 |
| Eva Creek 18 | 8/8/78 | 6.5 | 8.4 | 3.2 | 62 | 38 | | 1,172 |
| Eva Creek 19 | 8/8/78 | 6.6 | 3.6 | 10.6 | 85 | 42 | | 570 |
| Eva Creek 20 | 8/8/78 | 6.7 | 10.6 | 8.2 | 47 | 10 | | 260 |
| Eva Creek 20a | 8/8/78 | 6.8 | 10.6 | 9.7 | 44 | --- | | --- |
| Eva Seep 21 | 8/8/78 | 8.7 | 23.2 | 10.3 | 85 | 229 | | 1,723 |
| Mill Stream 22 | 8/8/78 | 8.7 | dry | dry | dry | dry | | 1,120 |
| West Mill Pond 23 | 8/8/78 | 7.2 | 26.5 | 8.8 | 32 | 45 | | 1,389 |
| East Mill Pond 24 | 8/8/78 | 6.3 | 23.9 | 6.6 | 67 | 127 | | --- |

In clear streams, even those draining heavily mineralized areas, the arsenic concentration of the stream water is about 50 ppb. For those streams with a large suspended-sediment load, the arsenic concentration can be estimated roughly by the regression equation of Wilson and Hawk ins

$$\log As_{(ppb)}water = 0.352 \log As_{(ppm)}sediment + 0.992.$$

The data in Map 2 suggest that arsenic analysis of stream-sediment samples might be useful in geochemical exploration for arsenic-bearing mineral assemblages (e.g., certain types of gold deposits). Care must be taken in interpreting the results of such a study because the scavenging effect of the iron oxides produces many false anomalies. For example, samples 2A and 2B (Nugget Creek) were taken about 3 feet apart, but one is iron-oxide rich and contains 888 ppm As, while the other is normal sediment and contains 147 ppm As. Similarly, samples 12 and 13 from Sheep Creek contain 695 ppm As (iron rich) versus 350 ppm As (iron poor), respectively.

These samples serve as good examples of the self-cleansing property of surface waters as a result of sorption of dissolved arsenic on iron oxides and hydroxides. This process is shown schematically in Figures 5 and 6.

Note that the surface waters draining the mineralized area of Ester Dome and in contact with sediment having 570 ppm As contain about 50 ppb As. The groundwaters draining the same area contain 5 ppm arsenic (100 times as much arsenic as in the surface water). This indicates that the coprecipitation and sorption process responsible for the removal of arsenic from surface waters is largely inoperative in the poorly oxygenated groundwater.

From Map 2 it is evident that the arsenic content of the stream sediments increases toward the headwaters of Eva and Ready Bullion creeks, an area of widespread arsenic mineralization. Several lode-gold mines are located in this area. No discernible effect on the arsenic content of the streams can be exclusively attributed to the mines. However, the arsenic-enriched sediments of the settling ponds by the mill (samples 22, 23, 24) are clearly the result of arsenic-enriched
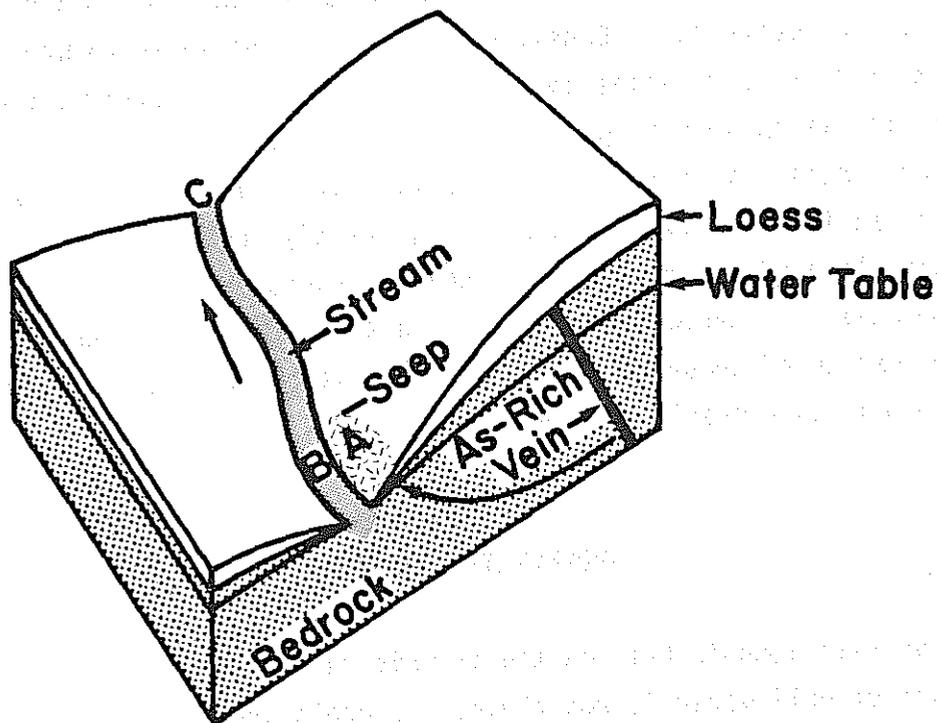
Figure 5.   Idealized cross section of stream on Ester Dome
showing the relation of arsenic-bearing vein to
groundwater and surface water.
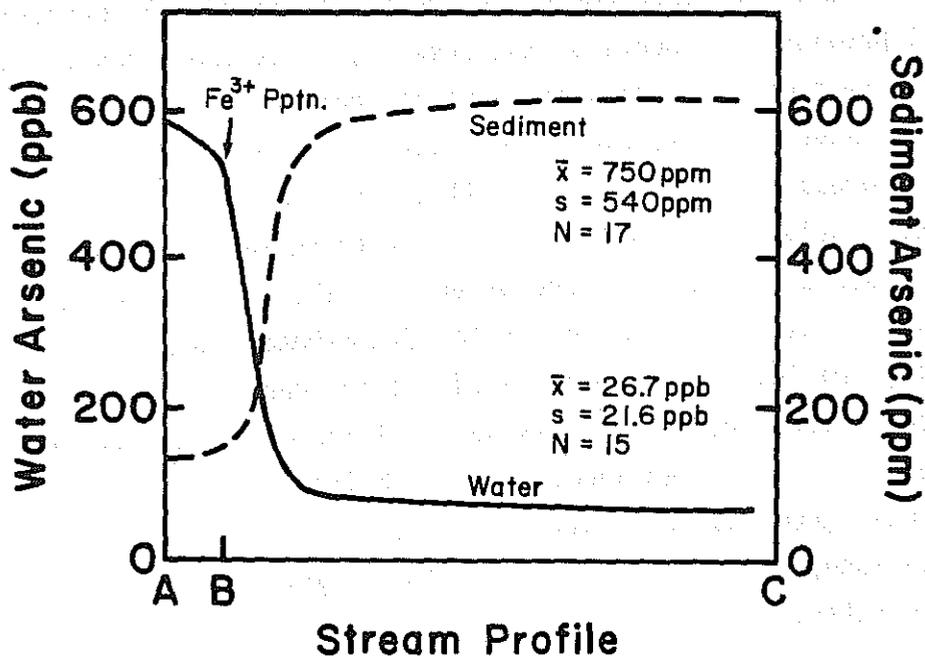


Figure 6.   Arsenic concentration in stream and groundwater
at localities A, B, and C (see Figure 5).  Note
the removal of arsenic by coprecipitation on
ferric hydroxide.

mill tailings. Streams could be adversely affected if subsequent mining and milling activities produce waste and tailings piles of arsenic-enriched materials. Consideration must be given to reduce the impact of these activities on the surface and, particularly, the groundwaters if extensive mining develops in the future.

We stress that stream sediments contain by far the greatest quantity of arsenic in a stream. Placer mining greatly increases the suspended-sediment load of a stream and thereby increases the quantity of arsenic transported. If these sediments are settled, the dissolved arsenic concentration of these streams will be about 50 ppb, assuming no changes in pH occur as a result of the placer mining.

## GROUNDWATER STUDIES

The relationship between the arsenic content of the country rock and that of well water is not clear. We don't know whether arsenic-rich well waters obtain their arsenic from sulfide-bearing veins somewhat distant from the wells in question, or whether the arsenic-contaminated wells penetrate arsenic-rich rocks, which are the source of arsenic in the well water. In other words, do the rocks immediately penetrated by the well provide the source of the arsenic, or is the arsenic source distant from the well? Does the distant source produce a zone of contaminated water which is then intercepted by a well?

To address these questions, well-cuttings and water from various wells were analyzed for arsenic to see if there was a relationship between arsenic in the rocks and arsenic in the water. Six wells were studied, only two of which were in the Ester-Dome area. These wells were being drilled in the upland subdivisions during the summer of 1978 by various homeowners. Few wells were drilled on Ester Dome because homeowners were concerned about the possibility of arsenic contamination of the local groundwater. They therefore chose not to spend the considerable money needed to drill wells that might produce arsenic-contaminated water.

With the cooperation of the various well drillers in the area, cuttings from 6 wells were sampled at various depth intervals. The number of intervals sampled and the distance between intervals depended upon the total depth of the well. Depth intervals usually ranged from 10 to 25 feet and cuttings from 6 to about 12 intervals were sampled. Special attention was paid to sample the rocks immediately above and below the water table, and to obtain at least several samples of cuttings from below the water table. A water sample was taken from each well. The sample was filtered through a 0.45 μ Millipore filter, acidified with 8 M nitric acid, and returned to the laboratory for analysis. Water from Brown's, Dean's and Gibson's wells were resampled in November 1979. The data are summarized in Table 5.

The results for Gibson's well are striking in that all rocks are rich in arsenic. The arsenic content of the rocks seems to be affected by the water table; samples above the water table contain more arsenic (mean value 1,200 ppm) than do those below the water table (mean value 400 ppm). The groundwater flowing through these rocks is much enriched in arsenic (850 ppb). These results are shown schematically in Figure 7.

The rocks in Brown's well (below the water table) contained an average of 200 ppm arsenic, yet the arsenic content of the water was less than 10 ppb. A resampling and reanalysis yielded results consistent with the initial values.

TABLE 5: AVERAGE CONCENTRATION OF ARSENIC IN ROCKS BELOW THE WATER TABLE AND IN THE GROUNDWATER FOR DIFFERENT WELLS.

| Well | Average As in Rock (ppm) | Average As in Water (ppb) |
|------|--------------------------|---------------------------|
| Brown | 100 | <10 |
| Dean | <40 | <10 |
| Gilbert | <40 | <10 |
| Koppelins | <40 | <10 |
| Maddocks | 40 | 24 |
| Gibbson | 407 | 728 |

Well

Average Arsenic Content
of Rock above Water Table:
## 1200 ppm

Fractured
Quartzite

Water Table

Arsenic Concentration
in Ground Water:
## 900 ppb

Mica
Schist

Average Arsenic Content
of Rock below Water Table:
## 400 ppm
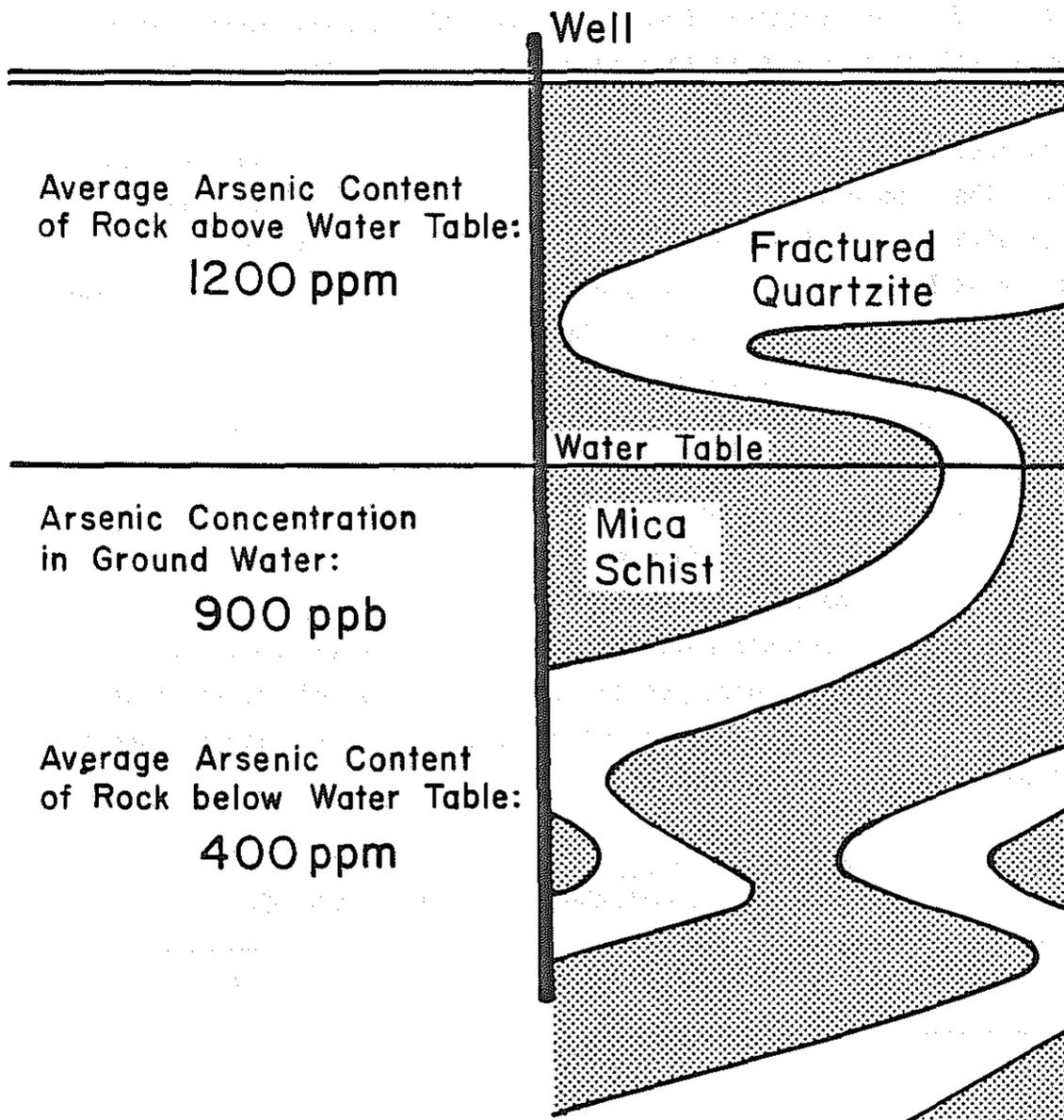
Figure 7.  Schematic representation of the arsenic content of rocks
and groundwater.

The data of Table 5 -- particularly the data for Gibson's well -- suggest that, in some instances, the arsenic in the water is being derived from the rocks in the immediate vicinity.

The data from Gibson's well suggest that, for large concentrations of arsenic to occur in the groundwater, there must also be heavy arsenic enrichment in all the rocks in the groundwater zone intercepted by the well.

The presence of a few arsenic-rich zones in the rocks at depth may, depending upon permeability, contribute arsenic-rich water to the well. The overall arsenic concentration in the well depends upon the arsenic concentration of the rocks, the number of horizons penetrated, and the amount of water flowing through the arsenic-rich zones relative to the amount of water flowing through arsenic-free zones. The zonal nature of arsenic contamination in the groundwater has been confirmed elsewhere in the Fairbanks area by the U.S. Geological Survey (Dorothy Wilcox, personal communication). The USGS investigators were able to seal off a zone of arsenic-rich water by means of hydraulic packers, and to pump water of low arsenic concentrations from the remaining zone. Unfortunately, this is not an economically feasible method for home owners to obtain good drinking water. A further problem (because of the erratic vertical distribution of arsenic-rich horizons) is that, upon deepening a well to obtain greater yield, a zone of arsenic-rich rocks and water may be intercepted, resulting in arsenic-rich water in a previously uncontaminated well.

## ARSENIC LEACHING EXPERIMENT

To determine if arsenic could be contributed to groundwater by leaching arsenic-rich rocks, the following experiment was performed. We placed 1.0 g of well cuttings (-2 mesh, Gibson well) into 30 ml centrifuge tubes with 25 ml of distilled water (pH 6.3). Nitrogen was bubbled through the solution in half the tubes for 2 minutes to dispel air. All the tubes were covered with Parafilm, capped, and then agitated on a mixing wheel for one week at room temperature. At the end of one week,

the pH of the solutions was measured and the solutions were filtered through 0.45 μ Millipore filters and analyzed for arsenic. The results of this experiment are shown in Table 6A. The quantities of arsenic leached in the nitrogen-saturated solutions were indistinguishable from those leached in the air-saturated solutions. Therefore, all results with the same final pH were pooled.

The average final pH values, the average arsenic values and values of arsenic predicted to be in equilibrium with $FeAsO_4$ based on Wagemann's calculations (1978) at the different pH values are shown in Table 6B. The concentrations calculated by Wagemann are for $FeAsO_4$ in a "typical freshwater." Because of the difference in ionic strength of the "freshwater" and the distilled water used in this experiment, concentrations calculated for this system should be somewhat lower than the calculated values in Table 6B.

Several features are evident in Tables 6A and 6B. Significant quantities of arsenic can be quickly leached from the country rock by distilled water. Furthermore, the arsenic concentration in solution follows a pH dependency like that for $FeAsO_4$. This suggests that the source of the arsenic in the rock is probably scorodite ($FeAsO_4 \cdot 2H_2O$) or possibly arsenic-bearing iron oxide. These results suggest that for well waters in the area, $FeAsO_4$ or a similar compound is the major component from which the groundwaters derive their arsenic concentrations. The arsenic concentration can be expected to vary as a function of the pH and $E_h$ of the groundwater, with higher arsenic concentrations favored at low pH and $E_h$. This implies that, as either pH or $E_h$ is increased, arsenic should be removed from solution first by coprecipitation on ferric hydroxide or by enhanced sorption on ferric-oxide coatings. Looking futher at the process of coprecipitation by $Fe(OH)_3$ and arsenic in an oxidizing environment, Figure 8 shows the decrease in total dissolved arsenic (TDA) of a groundwater sample originally containing 1 ppm arsenic, followed by standing undisturbed for 550 hours. It can be seen that coprecipitation in an oxidizing environment is capable of reducing arsenic concentrations from greater than 1 ppm to the U.S. Public Health Service drinking-water level of 50 ppb.

TABLE 6A: ARSENIC CONCENTRATIONS AND pH OF SOLUTIONS
IN CONTACT WITH CUTTINGS FROM GIBSON'S WELL.

| Sample Depth (ft) | As in Rock (ppm) | As in Solution (ppb) | | Final pH |
|---|---|---|---|---|
| | | $N_2$ Saturated | Air Saturated | |
| 96 | 1,304 | 388, 295 | 245, 433 | 6.3, 6.2, 6.3, 6.3 |
| 121 | 1,451 | 144 | 535, 192 | 6.3, 6.6, 6.7 |
| 125 (Water Table) | 1,730 | 172 | 228 | 7.3, 7.3 |
| 146 | 653 | 72, 84 | 70 | 7.7, 7.9, 7.9 |
| 171 | 876 | 74, 140 | 92, 74 | 8.1, 8.1, 8.2, 8.2 |

TABLE 6B: COMPARISON OF OBSERVED ARSENIC CONCENTRATIONS (TABLE 6A) WITH THOSE
EXPECTED FOR EQUILIBRIUM WITH SCORODITE (WAGEMANN, 1978).

| Final pH | Average As Concentration in Solution (ppb) | Total Dissolved As in Equilibrium with $FeAsO_4$ at Different pH Values[a] |
|---|---|---|
| 6.4 | 320 | >860 |
| 7.3 | 200 | >200 |
| 8.0 | 90 | 60 |

[a] From Wagemann (1978, p. 141).
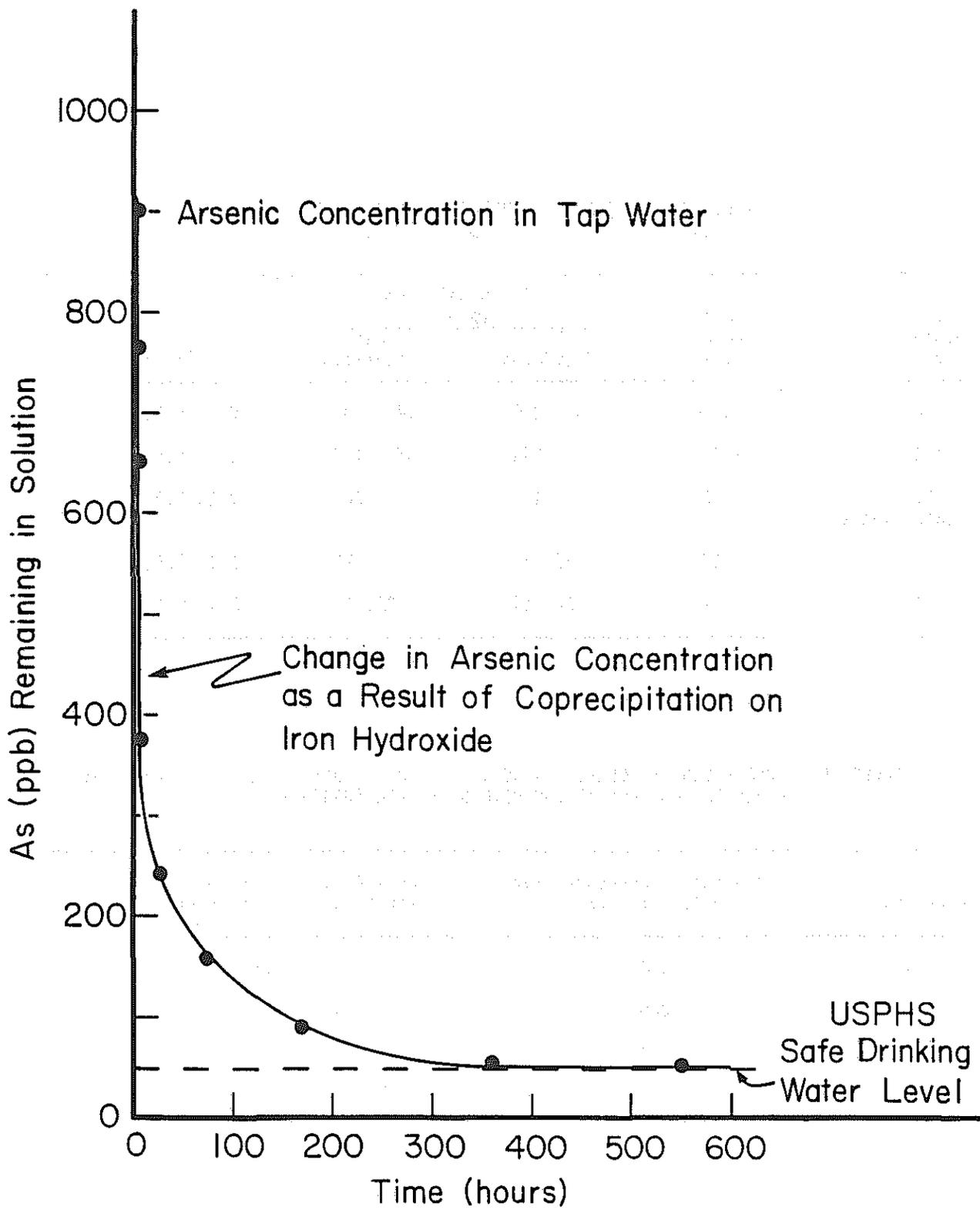
Figure 8.   Arsenic concentration versus time in a domestic water sample.

This particular sample was from a residence equipped with a home-made iron-removal system. The sample was from freshly pumped water before the water passed to the iron-removal system.

Samples of water that had passed through the iron remover were also analyzed. These showed a behavior consistent with coprecipitation on ferric hydroxide. The first sample was of milk prepared by mixing dried milk with tap water. This sample contained 160 ppb As (the same as that of the tap water). A sample of coffee made by means of an electric percolator was analyzed and found to contain 60 ppb As. The reduced arsenic concentration in the coffee results from aging; boiling reduces the concentration by flocculating ferric hydroxide, and the ferric hydroxide is removed by filtration through the coffee grounds. In other words, this process is a homely example of the classic Group III iron-separation scheme of qualitative analysis, and results in a decrease of both iron and arsenic in the water.

It is interesting to note that the final arsenic concentration achieved upon precipitation of the ferric hydroxide is close to that of the surface waters of Ester Dome.


ARSENIC SORPTION


To test the effect of sorption in oxidizing and reducing environments, the following experiment was performed. Duplicate samples weighing 0.25 g each of $Fe_2O_3$-coated quartz sand (Gent, 1977) (-35 to +60 mesh) and Fe-stained micaceous quartzite (Ester Dome) (-35 + 60 mesh) were placed in 30 ml thick-walled, screw-cap culture tubes. Then 25.0 ml of 1.0 ppm As standard solution in distilled $H_2O$ was added to each tube, and the pH was adjusted to 5.5, 6.5, or 7.5 with NaOH and $HNO_3$. Nitrogen was bubbled through half the solutions for 2 minutes. The tubes were covered with Parafilm, capped, and placed on a mixing wheel for 24 hours. The solutions were the filtered through 0.45 μ Millipore filters, and the pH of the solutions measured. The pH values of all solutions changed, with the final average pH = 7.1 ± 0.45.

The design for this experiment is shown in Table 7A; in effect, this was a modified partly replicated $2^3$ factorial design. Analysis of

the design showed that pH variations were not significantly different over the final pH range. Therefore, the data were pooled over the duplicates and the 3 pH values originally studied. Blanks were run to test the sorption of arsenic on glass or glass plus acid-washed quartz under nitrogen or air. The values in Table 7B are the observed arsenic concentrations (ppb) remaining in solution at the end of the experiment.

TABLE 7A: ARSENIC SORPTION EXPERIMENT.

| pH | Quartz + $Fe_2O_3$ | | Schist | |
|---|---|---|---|---|
| | Air | $N_2$ | Air | $N_2$ |
| 5.5 | 52 | 206 | 246 | 547 |
| 6.5 | 60 | --- | 185 | 487 |
| 7.5 | 77 | 267 | 280 | 469 |

TABLE 7B: CONTROLS FOR ARSENIC SORPTION EXPERIMENT.

| | Quartz & Glass Tube | Glass Tube | |
|---|---|---|---|
| $N_2$ | 750 | 796 | |
| Air | 964 | 848 | $\bar{x} = 840 \pm 140$ ppb |

With a coefficient of variation of 0.17, the above values for the blanks are indistinguishable statistically from each other, and the mean value is not significantly different from the original concentration. There may be a tendency for some sorption, (circa 15 percent) to occur on the vessel walls, but this cannot be shown statistically by the above data. As a consequence, no attempt was made to adjust the values of arsenic remaining in solution in contact with the schist or $Fe_2O_3$-coated quartz for sorption on the vessel walls.

It is evident from Table 7A that arsenic removal is enhanced under oxidizing, iron-rich conditions.

# ARSENIC SPECIATION

The conditions under which the different inorganic arsenic species are stable are summarized in the Eh-pH diagram (Figure 9), which also shows stability relations of some common iron species as well. Knowledge of the ionic state of arsenic in groundwater is important both in terms of understanding the chemical behavior of arsenic in groundwater and coping with the greater toxicity of As(III) relative to As(V). Unfortunately, the data in Table 8 do not answer the question of arsenic speciation unequivocally. Because of analytical difficulties, we were able to determine only total arsenic ($As_T$) concentrations on samples A, B, and C of Table 8. Maynard (1979) determined As(III) but not As(V) using polarographic methods. We could not determine As(V) by difference between these two values because the error in the individual determinations was too large. It seems, however, because of the good agreement between the As-total and As(III) values, that most of the arsenic is present in these samples in the As(III) state.

The data of Harrington et al. (1978; samples 1-5) show a wide range in the As(V) to As(III) ratio from 30/1 for sample 1, 1.5/1 for sample 2, and from 2/1 to 3.5/1 for the remaining samples. These data support our observations that most of the arsenic is present in the As(III) state. The samples analyzed by Harrington et al. were shipped to a laboratory in the conterminous United States for analysis. It is possible that because of the time elapsed between collection of the sample and analysis that some oxidation of the arsenic to the As(V) state occurred in spite of the precautions taken to minimize such oxidation. As a result, the relatively large concentrations of As(V) in some of the samples may be an artifact of the sample treatment. The data of Harrington et al. suggest that little arsenic occurs as an organic complex. This result is also speculative in that oxidation of the sample could destroy arseno-organic complexes prior to analysis.

These limited data suggest that most of the arsenic is present in the groundwater as As(III), and that As(V) and organic arsenic complexes are much less abundant. This conclusion is supported by Bottomley's
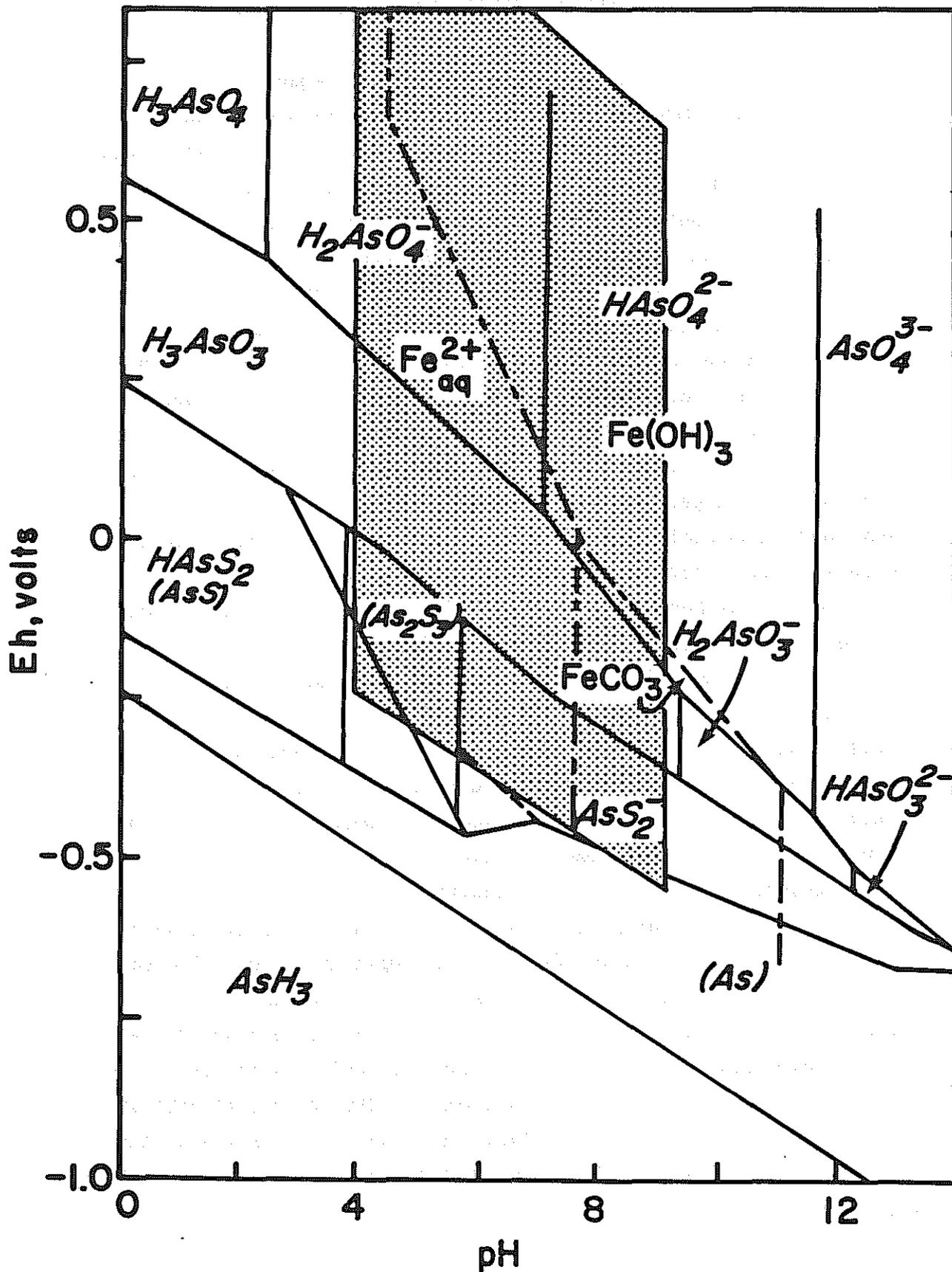
Figure 9. $E_h$ versus pH diagram, showing various arsenic ionic species and iron minerals (adapted from Wilson and Hawkins, 1978).

(1979) observation of arsenic in well waters of Nova Scotia. We stress that precise determination of relative quantities of different species of arsenic in groundwaters of the area remains to be done.

No data are available on the distribution of the different arsenic species in the surface waters of the Fairbanks area. Braman and Foreback (1973), in their studies of surface waters in Florida containing less than 5 ppb As, showed that about 40 percent of the arsenic was present as As(V), about 25 percent as As(III) and the remainder was present as methylarsonic and dimethyl arsonic acid. Presumably, most of the arsenic in the surface waters of the Fairbanks area is present as As(V).

TABLE 8: CONCENTRATION OF ARSENIC IN DIFFERENT IONIC STATES IN GROUNDWATERS OF FAIRBANKS AREA, EXPRESSED AS PARTS PER BILLION.

| Sample | As(Total) | As(III) | As(V) | MA[d] | DMAA[e] |
|--------|-----------|---------|-------|-------|---------|
| A | <10[a] | 9.67[b] | --- | --- | --- |
| B | <10[a] | 16.5[b] | --- | --- | --- |
| C | 851[a] | 854[b] | --- | --- | --- |
| 1 | --- | 35[c] | 1,065[c] | <1 | <1 |
| 2 | --- | 1,140[c] | 1,780[c] | <1 | <1 |
| 3 | --- | 1,170[c] | 2,470[c] | <1 | <1 |
| 4 | --- | 90[c] | 330[c] | <1 | <1 |
| 5 | --- | 110[c] | 410[c] | <1 | <1 |

a Determined by atomic absorption spectrometry.
b D. Maynard, (personal communication), determined by differential pulse polarography.
c Harrington et al. (1978), determined by differential pulse polarography.
d Methylansinic acid.
e Dimethylarsinic acid.

WATER-TREATMENT METHODS FOR REMOVING ARSENIC

Methods exist for large-scale removal of arsenic from domestic water supplies. Several of these are discussed by Whiteacre and Pearse (1974). A method using activated alumina for the removal of arsenic and fluoride from drinking water has been developed by the Alcoa Company

(see Commerce Bus. Daily, 4-18-78) under the auspices of the U.S.
Environmental Protection Agency. Several of these units are being
evaluated in Alaska for arsenic removal (Wm. Morgan, personal communica-
tion), but we have no information on their performance.

Until arsenic-removal systems suitable for single residences are
available, homeowners with arsenic-contaminated and iron-contaminated
waters may alleviate the problem through the use of a properly func-
tioning iron-removal system (for example, see Casper, 1975). The
arsenic concentration of the treated water should be carefully monitored,
and the water should not be consumed if the arsenic concentration
exceeds 50 ppb.


## SOIL AND VEGETATION STUDIES


Burton et al. (1978) investigated the arsenic content of different
types of vegetation on Ester Dome. Their results are shown in Appendix
C. Their work suggested that Labrador tea (*Ledum* spp.) might be useful
in geochemical exploration because the plant was ubiquitous, and because
its arsenic content seemed to vary in response to changes in the arsenic
content of the soil on which it was growing. Results of their geochemical
sampling across the Ryan Lode are shown in Figures C1-C4.

We examined further the arsenic content of Labrador tea and its
usefulness in biogeochemical prospecting for arsenic and related min-
erals. We analyzed different soil horizons as well as Labrador tea at
various sample sites. Our results are shown in Tables 8A, 8B, and 8C
and in Figures 10A, 10B, and 10C.

Our results, particularly for samples from the Ridge traverse
(Table 8B and Figure 10C) show that arsenic concentrations vary in the
different samples of Labrador tea, and the $A_o$- and A-soil horizons. The
agreement between our analytical results and those obtained by neutron
activation analysis indicate that the arsenic enrichment in the Labrador
tea samples is real.

The relationship between the arsenic content of the Labrador tea
and that of the different soil horizons is not clear. Our data for the
Ryan-Lode traverse show little response in these sample media to the

TABLE 8A: ARSENIC IN SOILS AND VEGETATION: RYAN LODE TRAVERSE
(VALUES IN PPM).[a]

| Locality | Soil Horizon | | | Vegetation |
| | $A_0$ | $A_1$ | B | Labrador Tea |
|---|---|---|---|---|
| 1 | 5.9 | 25 | 33 | 2.5 |
| 2 | 10 | 16 | 25 | <2.0 |
| 3 | 6.9 | 24 | 24 | 2.1 |
| 4 | 7.9 | 31 | 33 | <2.0 |
| 5 | 7.9 | 35 | 28 | <2.0 |
| 6 | 10 | 32 | 24 | --- |
| 7 | 10 | 20 | 25 | <2.0 |
| 8 | 22 | 23 | 27 | <2.0 |
| 9 | 93 | 31 | 25 | <2.0 |
| 10 | 28 | 28 | 29 | <2.0 |
| 11 | 25 | 24 | 151 | 2.1 |
| 12 | 6.9 | 22 | 26 | <2.0 |
| 13 | 7.9 | 30 | 33 | <2.0 |
| 14 | 7.9 | 18 | 34 | <2.0 |

a
Analytical precision for values reported under $A_0$, $A_1$ and Labrador
Tea, in Table 8A, 8B, 8C. Error range at 2 standard deviations.

| For | x <2.0 | Error ± ∞ |
| " | 2.0< x <3.0 | " ± 1.0x to ± 0.8x |
| | 3.0< x <5.0 | " ± 0.8x to ± 0.5x |
| " | 5.0< x <10 | " ± 0.5x to ± 0.3x |
| " | 10< x <20 | " ± 0.3x to ± 0.2x |
| " | 20< x <50 | " ± 0.2x to ± 0.17x |
| " | 50< x | " ± 0.17x |

TABLE 8B:   ARSENIC IN SOILS AND VEGETATION.

| Locality | Soil Horizon | | | Vegetation | |
|---|---|---|---|---|---|
| | $A_o$ | $A_1$ | B | Labrador Tea | |
| 1 | 18 | 35 | 33 | <2.0 | |
| 2 | 16 | 26 | 29 | <2.0 | |
| 3 | 70 | 110 | 52 | 3.8 | 4.91[a] |
| 4 | 22 | 80 | 36 | <2.0 | 1.15[a] |
| 5 | 5.0 | 19 | 49 | <2.0 | |
| 6 | 2.4 | 20 | 48 | 2.3 | |
| 7 | <2.0 | 3.9 | 67 | 3.4 | |
| 8 | 3.8 | 3.0 | 148 | <2.0 | |
| 9 | 2.8 | 11 | 60 | <2.0 | |
| 10 | 3.5 | 18 | 32 | <2.0 | |
| 11 | 3.4 | 24 | 18 | --- | |
| 12 | <2.0 | 15 | 23 | --- | |
| 13 | 5.9 | 14 | 21 | --- | |
| 14 | 4.9 | 14 | 19 | <2.0 | |

[a] Neutron activation analysis results.

## TABLE 8C: ARSENIC IN LABRADOR TEA NEAR STREAM SAMPLE LOCALITIES (ALL VALUES IN PPM).

| Locality | $A_s$ |
|---|---|
| Nugget Creek 1 | <2.0 |
| Nugget Creek 4 | <2.0 |
| Nugget Creek 5 | <2.0 |
| Nugget Creek 6 | <2.0 |
| Nugget Creek 7 | <2.0 |
| Eva Creek 14 | <2.0 |
| Eva Creek 15 | 2.9 |
| Eva Creek 16 | <2.0 |
| Eva Creek 17 | <2.0 |
| Eva Creek 19 | <2.0 |
| Ready Bullion Creek 9 | <2.0 |
| Gilbert Weil Creek 4 | <2.0 |
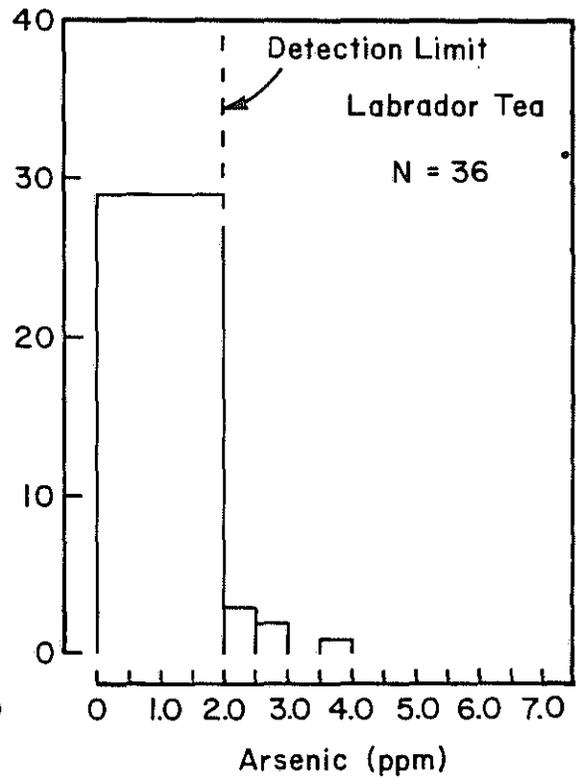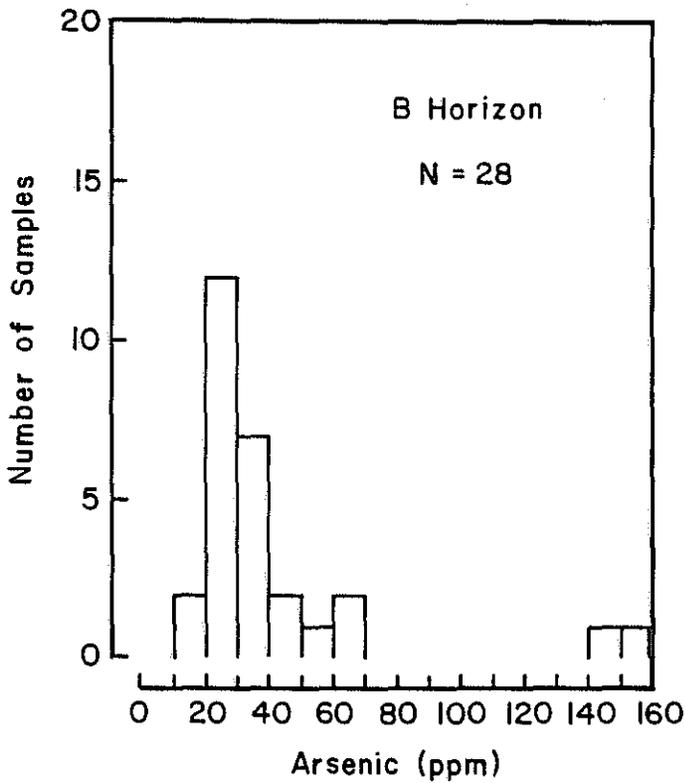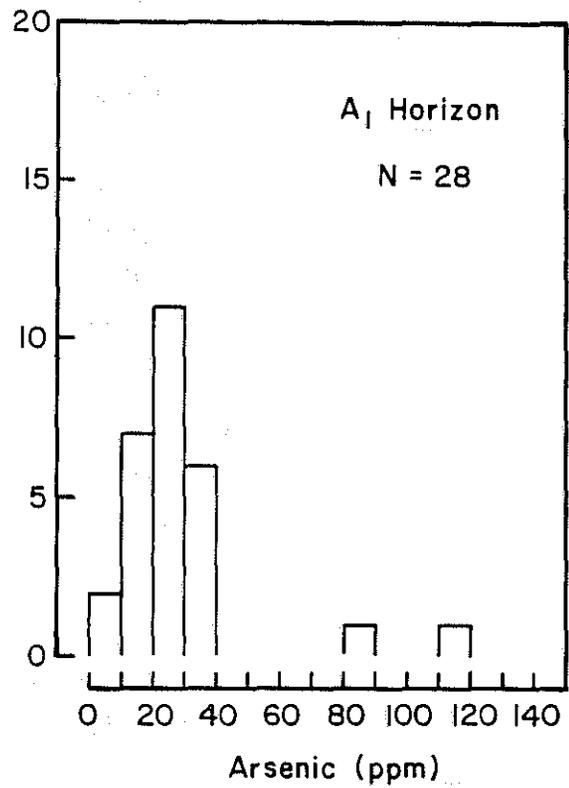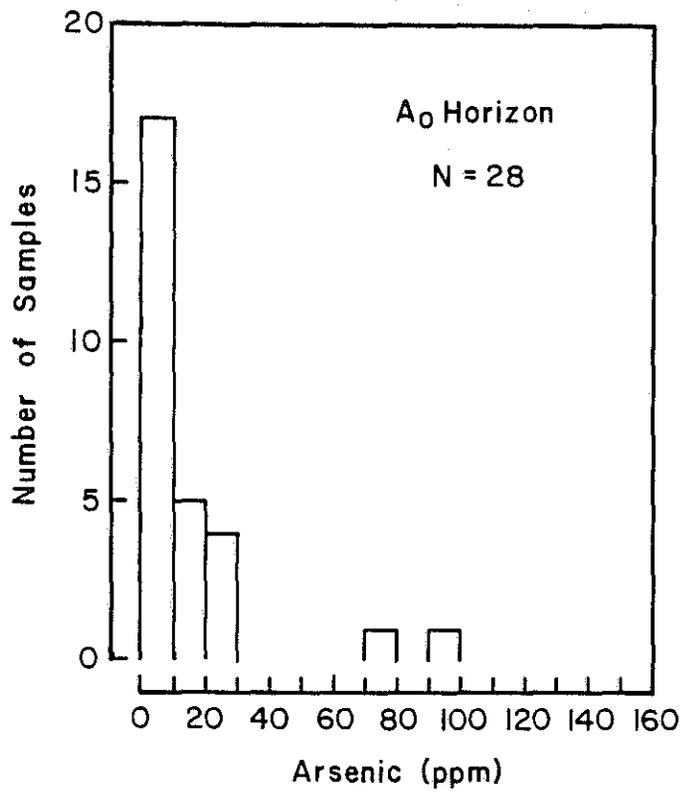
Figure 10A.  Arsenic content in soil horizons and vegetation from
            Ryan Lode traverse.
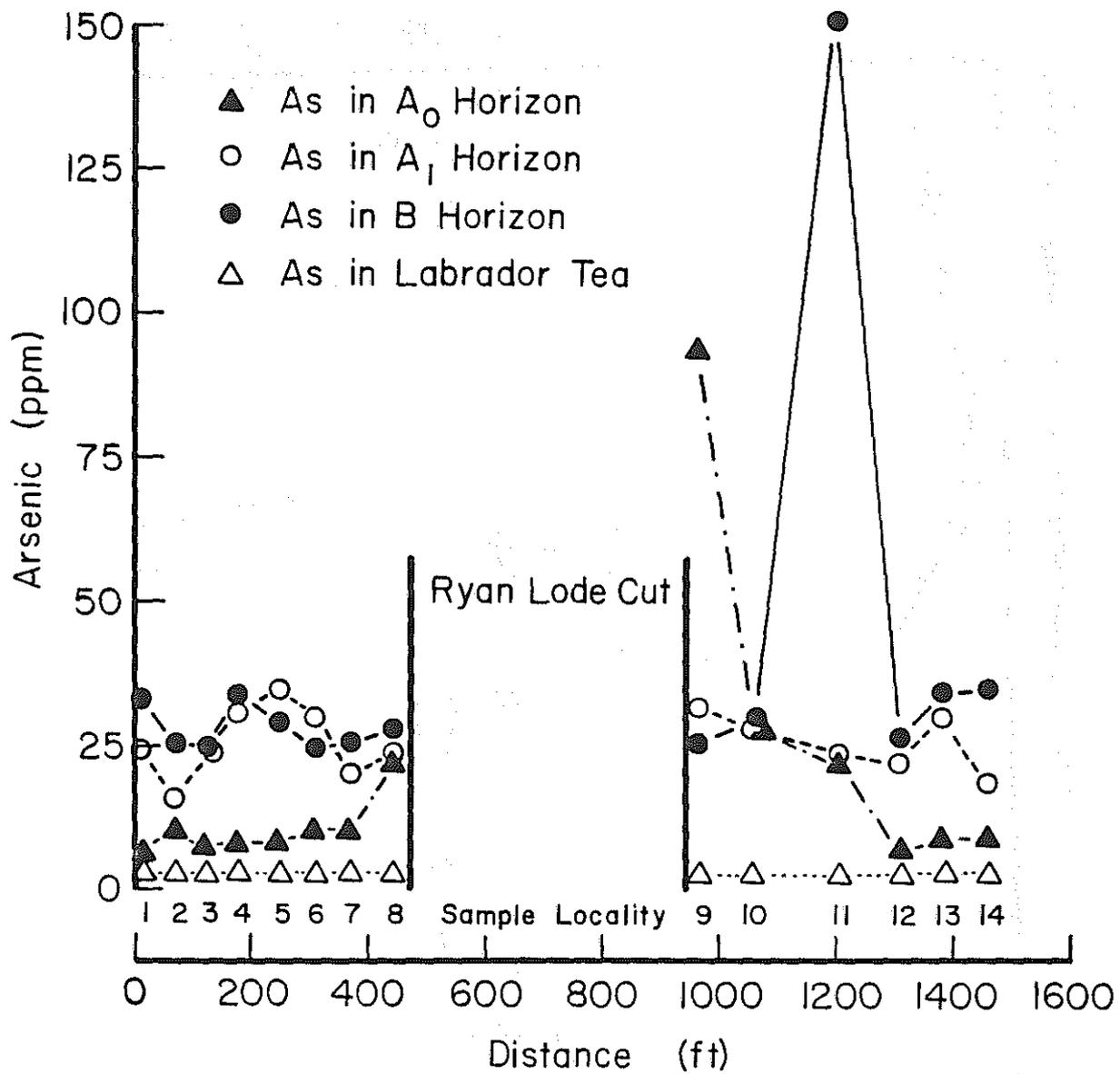
Figure 10B.   Arsenic in samples from Ryan Lode traverse.
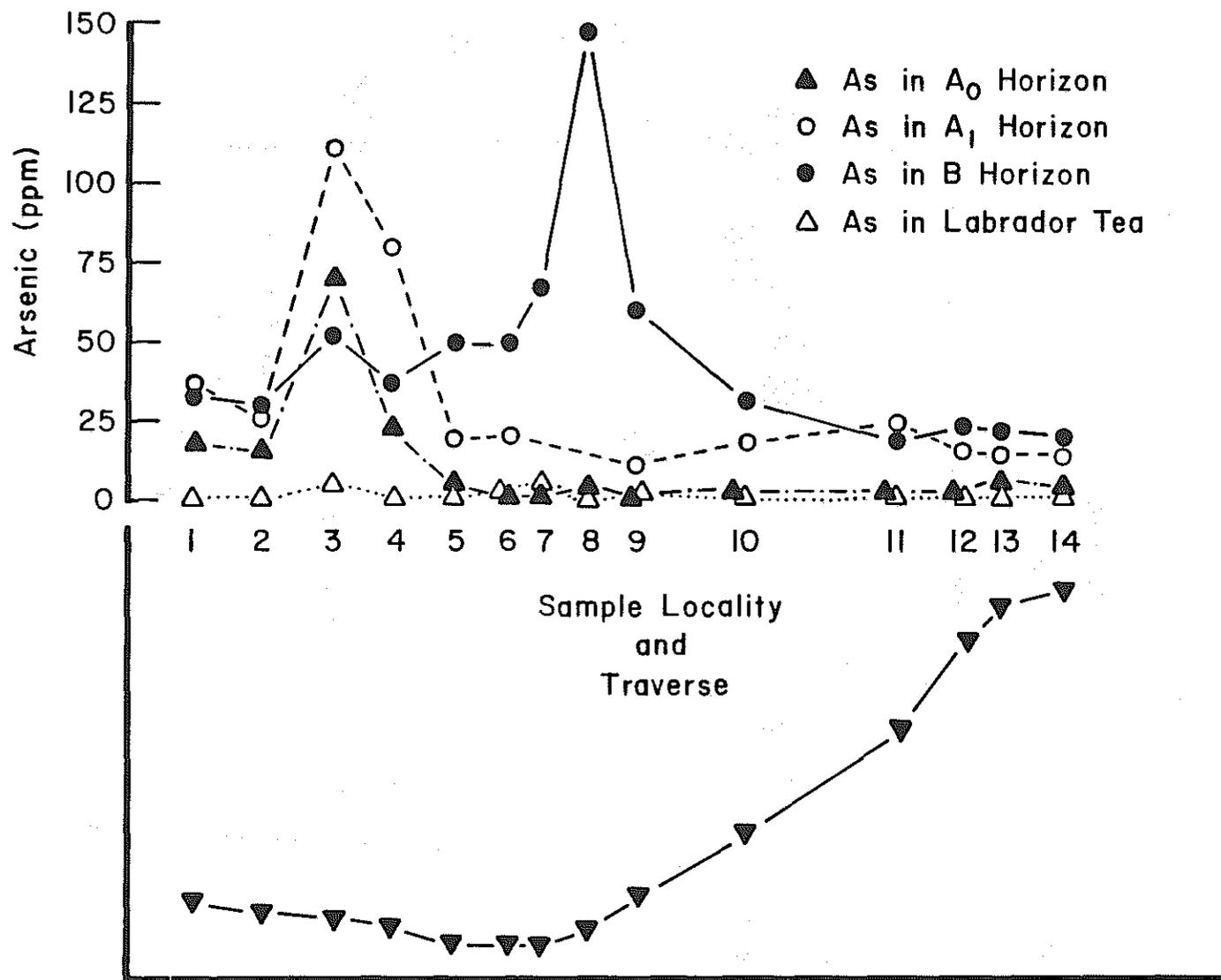
Figure 10C. Arsenic in samples from ridge traverse.

presence of arsenic mineralization. The single high value for arsenic in the $A_o$-horizon sample from locality 8 may be a result of contamination by arsenic-rich dust from the stripped mineralized zone adjacent to this locality. Samples from localities 1 through 4 of the Ridge traverse may be contaminated by dust from the Ester Dome road.

Arsenic enrichment in the B-horizon samples from both the Ryan-Lode and Ridge traverses is statistically significant ($\propto$ 0.05) and chances of contamination are slight. Arsenic should be enriched in the B-horizon because iron oxides accumulate in this zone. The affinity of iron oxides for arsenic should then lead to enrichment of this horizon in arsenic. The anomalously high values of arsenic in some of the B-horizon samples probably reflect arsenic mineralization in the bedrock. A comparison of these arsenic values with gold values obtained by Stevens et al. (1969) along the same (Ridge) traverse supports this interpretation. Of particular interest are the analytical results for samples 7, 8, and 9 of the Ridge traverse, which suggest a mineralized zone beyond and approximately parallel to the Ryan Lode.

We are unable to reproduce the arsenic values obtained by Burton et al. in vegetation samples taken across the Ryan Lode. We agree with them that arsenic is taken up from the soil by Labrador tea, but our results suggest that this enrichment is slight. It seems that for this reason, and due to ease of analysis, that sampling and analysis of the B-soil horizon is a more effective geochemical prospecting method to delineate arsenic mineralization than is vegetation sampling.

With regard to using plants for geochemical prospecting, Horler et al. (1980) present data on detecting arsenic-rich areas by remote-sensing methods. Their results show a highly significant negative correlation (>99 percent between the soil arsenic concentration and the 1.65 µm reflectance for oak trees growing in arsenic-rich soils). Other investigators have shown that plants under stress are most evident in the fall or in the spring immediately after snowmelt. These results suggest that a remote-sensing investigation using this spectral region might be useful in delineating arsenic-rich areas in the Fairbanks region.

# GREENHOUSE STUDIES

Various common garden vegetables were grown under greenhouse conditions to test the extent to which garden vegetables become enriched in arsenic when grown with arsenic-containing water. Six plants each of tomatoes (Tuckcross 533), cucumbers (LaReine), lettuce (Ostinata), and radishes (Burpee White) were grown. Three of each set were treated with arsenic-containing well water (Ester Dome area, 3.5 ppm As) and three were treated with water containing no arsenic. The tomatoes and cucumbers potted in 2 gallon nursery cans (1 plant per can) received 500 to 1,000 ml of water every 2 to 3 days per plant.

At the end of the experiment, the plants were harvested and the fruit, leaves and roots were dried and ground. The air-dried plant material was then analyzed for arsenic following the method described in Appendix A. Because of possible arsenic contamination from the analytic reagents and the possible loss of arsenic through volatilization during the ashing process, we checked our results using neutron activation analysis on selected samples of dried plant material. The results of both types of arsenic analysis are summarized in Table 9 and given in greater detail in Appendix B.

The soils in which the plants were grown were also analyzed and the results of these analyses are presented in Table 9.

The data of Table 9 clearly show that soils treated with arsenic-rich water are enriched in arsenic. The tomatoes received about twice as much water as did the other plants because of the longer growing period required. The soils in which the tomatoes were grown contained correspondingly more arsenic. Clearly then, the more arsenic-containing water added to the soil, the more arsenic there is in the soil. The water used in this study contained much iron, which precipitated in the jug upon standing, carrying down the arsenic. Such arsenic-rich iron precipitate becomes concentrated in the top part of the soil, where it sorbs arsenic from solution in subsequent waterings. As a result, the soil (particularly the top part, as can be seen in Table 9) becomes increasingly rich in arsenic with continued watering.

TABLE 9A: ARSENIC IN GREENHOUSE VEGETABLES EXPRESSED IN PPM.[a]

| Plants | Without Arsenic | | With 3.5 ppm Arsenic | |
| | Roots or Fruit | Stems & Leaves | Roots or Fruit | Stems & Leaves |
|---|---|---|---|---|
| Tomato | <2 ppm | <2 | <2 | 2.9 |
| Cucumber | <2 | 2.8 | <2 | 4.8 |
| Lettuce | -- | 2.9 | -- | 12 |
| Radish | <2 | <2 | 4.8 | 14 |

[a] Experiments were done in triplicate. Analysis was by atomic absorption spectrometry and neutron activation. Natural arsenic-containing well water was used.


TABLE 9B: ARSENIC IN GREENHOUSE SOIL EXPRESSED IN PPM.

| Soil | Top Half | Bottom Half | Total (ppm) | Liters Water Used |
|---|---|---|---|---|
| Untreated | -- | -- | 14.8 | -- |
| Treated | | | | |
| Tomato | 200 | 41.7 | 122 | 52 |
| Cucumber | 47.2 | 27.8 | 38.5 | 15 |
| Lettuce | 36.7 | 28.7 | 32.7 | 20 |
| Radish | 45.3 | 31.0 | 38.2 | 23 |

The fruit of tomatoes and cucumbers grown with arsenic-rich water are only slightly enriched in arsenic relative to the untreated plants. Tomato leaves and particularly cucumber leaves of the arsenic-treated plants contain more arsenic than do those of the untreated plants. The arsenic-treated lettuce and radishes showed significant arsenic enrichment compared with the untreated plants. The leaves of radishes are enriched in arsenic compared to the roots of the same plants. It appears from our data that for the different plants studied, the leaves tend to be richer in arsenic than do the roots or fruit. Furthermore, it appears that radishes and lettuce have higher arsenic concentrations than do tomatoes and cucumbers, even though the tomatoes in particular received a larger quantity of arsenic.

The results of our atomic absorption analysis are in reasonable agreement with those obtained by neutron activation analysis. Our data (Tables 9A and 9B) are comparable to similar data from other studies (Table 10).

The arsenic enrichment of the top, compared with the bottom, part of the soil suggests a possible explanation for why the lettuce and radishes contained more arsenic than did the tomatoes and cucumbers.

The lettuce and radishes are shallow rooted and thus grow in the top arsenic-rich zone, whereas the tomatoes and cucumbers are deeper rooted and draw water and nutrients from the lower arsenic-poor zone. Differences in the arsenic content of the water reaching the roots of the plants, as well as differences in the arsenic-assimilative behavior of the different plant types, may also contribute to the observed differences in arsenic content among the plants studied.

The tendency for arsenic to accumulate in the top part of soil treated with arsenic-rich water may be cause for concern among gardeners with long-established garden plots. The following calculation is an attempt to estimate (for the worst possible case) the quantity of arsenic that might be obtained by eating vegetables grown on such arsenic-rich plots. Assume a tilling depth of 1 foot and a 10-year period of watering the soil with water containing 5 ppm arsenic. Using the soil from the tomatoes as an example, 125 ppm arsenic accumulates in one

TABLE 10:  ARSENIC CONCENTRATIONS IN GREEN VEGETABLES GROWN
WITH AND WITHOUT ADDITION OF VARIOUS ARSENIC COMPOUNDS.

| Vegetable | Without arsenic (ppm dry wt.) | | With arsenic (ppm dry wt.) | | |
|---|---|---|---|---|---|
| | Root or Fruit | Stem & Leaves | Root or Fruit | Stem & Leaves | Arsenic[a] Treatment |
| Tomato | 0.01-2.95 | <.2 | 0.68-39.5 | 334 | 4S |
| Tomato | 0.08-0.09 | 6.75 | trace-0.09 | 11.4 | 1S |
| Tomato | trace | -- | 3.75-145 | --- | 3N |
| Tomato | trace | -- | trace-18.1 | --- | 4N |
| Cucumber | 0.02-2.4 | -- | 0.2 | --- | 1S |
| Lettuce | --- | 0.01-3.78 | --- | 0.0-2.1 | 3P |
| Lettuce | --- | -- | --- | 0.08-0.32 | 1S |
| Radish | 0.01-2.02 | -- | trace | --- | SP |
| Radish | --- | -- | 0.02-0.22 | --- | 1S |

a  Treatment codes, 1 = lead arsenate, 2 = calcium arsenate,
   3 = sodium arsenate, 4 = sodium arsenite, N = nutrient
   solution, S = soil, SP = smelter pollution.

year. If the arsenic content is linearly additive, then in 10 years there might be 1,250 ppm arsenic (as a ferric arsenate compound) in the soil. This value is comparable to that found in stream sediments of the area. If the arsenic content of the vegetables is proportional to the arsenic content of the soil in which they are grown, then a person could possibly ingest 625 µg arsenic by eating one normal serving of lettuce per day (see calculation in Appendix B). In comparison, the average daily water intake of an adult is about 1,000 ml. The quantity of arsenic ingested from such lettuce is then comparable to that obtained from drinking a normal daily quantity of water containing 625 ppb (µg/L) arsenic.

It is not known if such high arsenic concentrations in lettuce can result from growing lettuce on soil watered for prolonged periods with arsenic-rich waters. The data of Table 10 show that concentrations as large as this have been found in vegetables and that the resultant concentration in the plant appears to depend upon the form in which the arsenic is added.

As a result, the possibility exists that eating vegetables grown in gardens watered with arsenic-rich water can provide quantities of arsenic comparable to those obtained from drinking arsenic-rich waters. In other words, the arsenic dose received by eating such vegetables may not be insignificant in comparison to that from drinking arsenic-rich water.

We have no data on the actual arsenic content of garden vegetables, or of garden soils in the Ester Dome area. Such data should be obtained.

SUMMARY

This study was undertaken as a result of an earlier study in which arsenic concentrations as large as 10 parts per million (ppm) were found in domestic water supplies of the area. This earlier study triggered an intensive study by various state and federal agencies to evaluate the threat to human health posed by these arsenic-rich waters.

From a health standpoint, 70 mg arsenic has proven to be toxic to humans, while arsenic in low concentrations (less than 1 ppm) appears to be a carcinogen. The U.S. Public Health Service (USPHS) recommended guide limit for arsenic in potable water supplies is 10 parts per billion (ppb). An arsenic level above 50 ppb constitutes grounds for rejecting the water as a public water supply.

From the work of the various health agencies in the Fairbanks area, it was found that people drinking arsenic-rich waters are themselves enriched in arsenic. It is not known at this time if such arsenic-enrichment affects the health of the individuals involved. It was also found that the arsenic contamination of the groundwaters of the Fairbanks area was more extensive than first thought. The Ester Dome area has a high proportion of wells containing large concentrations of arsenic.

Our previous work and subsequent studies by the U.S. Geological Survey and the Alaska Department of Environmental Conservation suggest that arsenic-containing wells in the area are confined to the hillside residential subdivisions where water is obtained from bedrock aquifers. The arsenic concentrations in these areas show large horizontal concentration gradients, so that while one well might have 5 ppm arsenic, another well 100 yards away might contain only 50 ppb arsenic. The waters from these areas are used for domestic purposes as well as for irrigating vegetable gardens.

OBJECTIVES

1. To determine the geological and geochemical factors affecting the distribution of arsenic in the waters of the Ester Dome area.

2. To evaluate the arsenic content of plants grown on arsenic-rich soils or with arsenic-rich water.

# RESULTS AND CONCLUSIONS

Geologic mapping of the Ester Dome area has more clearly defined the previously recognized mineralized region there. A very wide zone of metallic enrichment exists which includes lodes and interlayered mineralized schists and micaceous quartzites typified by high concentrations of arsenic. For example, rocks from this zone contain an average of 750 ppm arsenic, which is about 100 times higher than the arsenic content of average unmineralized metamorphic rocks. The arsenic-rich zone is situated on the southeastern flank of Ester Dome and the rocks in this zone have a predominantly southeasterly dip. The structural dip and the well-developed fractures in the more competent rock units provide channels for the down-dip movement of arsenic-rich waters. As a result of this geologic setting, it is not surprising that many of the domestic wells with the highest concentrations of arsenic are situated on the southeast flank of Ester Dome, down dip from the arsenic-bearing mineralized zone.

Analyses of well cuttings from domestic wells show that some wells are drilled through rocks enriched in arsenic. These rocks serve as an immediate source of arsenic to the groundwater flowing into the well. The primary source of arsenic in the waters of the area is the arsenic-bearing sulfide mineralization in the mineralized zone. However, equilibration of the groundwater with the iron-arsenate mineral, scorodite, seems to be the main factor governing the concentration of arsenic in the groundwater. None of the cuttings from these arsenic-rich wells contained arsenic sulfides visible under the microscope. The cuttings did have arsenic-containing iron oxides coating veinlets and fissures. Laboratory experiments showed that arsenic in concentrations comparable to those found in groundwater (500 ppb) could be obtained from these cuttings by leaching the rock chips in distilled water under oxygen-poor conditions at pH values near 6.5. Such solutions are rich in arsenic and iron, and oxygen poor.

A similar situation obtains in the groundwaters of the area where it appears that most of the arsenic in the groundwater is present in the reduced (As III) rather than oxidized (As V) form. When these waters

are aerated, arsenic is coprecipitated with ferric hydroxide. Also, arsenic is removed by sorption (especially under oxidizing conditions) on previously precipitated ferric hydroxides and oxides. This process is operative in the surface waters of the area and is the means whereby the arsenic concentration of the surface waters is naturally reduced.

Stream sediments in the area tend to be enriched in arsenic particularly in or near the mineralized zone. This suggest that arsenic analysis of stream sediments might be useful in geochemical prospecting for sulfide and accompanying gold mineralization in the area. Our earlier work showed a significant correlation between the arsenic content of the bottom sediments and the dissolved arsenic concentration in the stream water. This relationship was especially evident in streams containing large amounts of suspended sediments, such as would result from placer mining. It was suggested that removal of the suspended sediments would significantly reduce the dissolved arsenic concentration.

Results from the present study on samples taken under conditions in which the suspended-solids content was low showed no relation between the arsenic content of the bottom sediments and the dissolved arsenic concentration. Furthermore, the arsenic concentrations were all less than 100 ppb, even for those samples in which the bottom sediments contained as much as 1,400 ppm arsenic. This tends to confirm our earlier observations that, although placer mining contributes arsenic to stream waters through the sediment burden produced, the dissolved arsenic concentration is brought to near or below the USPHS concentration guide limits when these sediments are removed.

Garden vegetables, grown under greenhouse conditions using water containing 3 ppm arsenic tend to be enriched in arsenic relative to those vegetables grown with arsenic-free water. The leaves of lettuce and radishes grown with arsenic-rich water contained 12-24 ppm arsenic per gram of dry plant, whereas the leaves plants treated with arsenic-free water contained less than 2 ppm arsenic. (Two ppm was the analytical detection limit using atomic absorption spectrometry. These results of vegetation analysis were confirmed by neutron activation analysis of selected samples.) Radish roots of arsenic-treated plants

contained 5 ppm arsenic, whereas the roots of untreated plants contained less than 2 ppm. Fruits of tomatoes and cucumbers grown with arsenic-rich waters were not significantly enriched in arsenic relative to the untreated plants. Because the soil becomes enriched in arsenic upon prolonged watering with arsenic-rich water, and because the arsenic content of the plants may increase with increasing arsenic content of the soil, the quantity of arsenic a person might ingest by eating garden vegetables grown with arsenic-rich water may not be insignificant relative to that obtained by drinking the water. Further study of this seems warranted.

In an earlier and related study, students of the Geology-Geophysics Program of the University of Alaska obtained results suggesting that alder and Labrador tea growing in arsenic-rich soils were enriched in arsenic. Our study showed only a slight increase in the arsenic content of Labrador tea growing in heavily mineralized areas. Furthermore, there was no clear-cut relationship between the slight enrichment found and location of mineralized veins. This suggests that the vegetation studied does not become particularly enriched in arsenic and that biogeochemical prospecting using these plants may not be very successful. Similarly, there was a tendency toward slight arsenic enrichment in the organic-rich $A_o$ and A-soil horizons. The B-soil horizon, on the other hand, showed significant arsenic enrichment that seemed to parallel known arsenic mineralization. This suggests that the B horizon might be much more useful as a sampling medium in geochemical prospecting for arsenic and accompanying gold mineralization.

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# APPENDIX A
## ANALYTICAL METHODS

Water

The procedure used here is the same as that used by Wilson and Hawkins (1978) which was modified from Fernandez and Manning (1971). The instrument used was a Perkin-Elmer Model 303 Atomic Absorption Spectrophotometer equipped with an HGA-2600 graphite furnace.

Twenty-five microliters (25 µL) of filtered and acidified water sample was transferred to the furnace by means of a pipette. Next, 25 µL of an aqueous 0.1 percent $Ni(NO_3)_2 \cdot 6H_2O$ solution was added to reduce volatility of the arsenic during the drying and charring stages. Instrumental conditions for the spectrophotometer were the same as those listed for arsenic in the Perkin-Elmer manual. The furnace settings were as follows:

| | | |
|---|---|---|
| dry | $115^\circ C$ | 60 seconds |
| char | $1,200^\circ C$ | 30 seconds |
| atomize | $2,500^\circ C$ | 15 seconds |
| gas | argon | 300 $cm^3$/minute |

Standards containing 10, 20, 30, 50 and 100 ppb arsenic were prepared by serial dilution with 0.2 m $HNO_3$ of a 1,000 ppm arsenic standard solution. Standards and samples were analyzed in triplicate under the above instrumental conditions. The quantity of arsenic in the sample was obtained from a standard curve. Because of the number of arsenic analyses performed and the fact that the standard curve changed somewhat during the lifetime of a graphite tube, the arsenic standard curve was fit by means of a computer program. This program (called ASSTD; written by D. Hawkins) constructs a least-squares fit to the arsenic-standard curve data and calculates the concentration of arsenic in the samples giving a mean value plus or minus 2 standard deviations. The estimated precision of the analysis was obtained by taking into account both the variance of the standard curve data and that for the samples. A copy of the program can be obtained from us on request.

## Vegetation Analysis

The method used was slightly modified from that of Ward (1975) for the dry ignition of vegetation prior to the arsenic determination. This method is similar to that used by Burton et al. (1978) which was based on the method of Friend, Smith and Wishart (1977).

Following Ward:

1. Weigh 2.0 g of ground, air-dried vegetation into an evaporating dish and wet with a small amount of ethanol.

2. Add 10 ml of magnesium oxide-magnesium nitrate-nickelous nitrate slurry and mix well.

3. Place dish on a hot steam bath and dry.

4. Transfer dish to preheated muffle furnace and ignite vegetation at 550-600°C for 2 to 4 hours.

5. Remove dish from the muffle furnace, cool, moisten ash with a minimum amount of water and cover dish with a cover glass. Add about 15 ml of 6 M HCl.

6. Heat dish and contents on water bath until sample dissolves.

7. Filter the hot acid solution through Whatman 41 filter paper into 100 ml volumetric flask. Rinse dish and filter with several portions of hot distilled, arsenic-free water until filtrate volume is about 60 ml. Cool the flask to room temperature and dilute the flask to volume with water.

8. Analyze using atomic absorption spectrometry following the method described above. It is not necessary to add additional nickelous nitrate.

The furnace settings as follows:

| | | |
|---|---|---|
| dry | 115°C | 2 minutes |
| char | 1,200°C | 1 minute |
| atomize | 2,500°C | 15 seconds |

Magnesium oxide-magnesium nitrate-nickelous nitrate slurry:

Suspend 37.5 g magnesium oxide, 52.5 g magnesium nitrate hexahydrate and 24.8 g nickelous nitrate hexahydrate in enough arsenic-free water to make 500 ml of solution. Shake slurry vigorously before using.

This solution serves as an ashing aid with the nickel acting to reduce the volatility of arsenic both during ashing and during analysis.

## Soils

In this method, arsenic bound with iron oxides dissolves, but only partial dissolution of arsenic-bearing sulfides is achieved.

1. Weigh 1.0 g of -100 mesh air-dried soil into a 125 ml beaker.
2. Add 50 ml of 1.0 N HCl. Cover beaker with a watch glass, place beaker on hot plate at medium heat and boil gently for 1 hour.
3. Filter the hot acid solution through Whatman 41 filter paper into a 100 ml volumetric flask, washing the cover glass, beaker and filter several times with hot, arsenic-free water until the filtrate has a volume of about 60 ml.
4. Cool the flask to room temperature and dilute to volume with arsenic-free water.
5. Analyze for arsenic following the procedure used for the water analysis.

## Rocks and Sediments

The rocks (well cuttings) and stream sediments were analyzed by x-ray fluorescence. Because iron is the only abundant element affecting the $K_\alpha$ spectra in the sample, standards were prepared by serial addition of $As_2O_3$ to finely ground mixtures of quartz and $Fe_2O_3$. The iron content of the standards were 10 percent by weight, which is about equal to the sum of $Fe_2O_3$, FeO, MgO and MnO in the samples (quartz-mica schists). The standards were mixed for three minutes in a Pitchford blender. Next, 1.50 g of the standard was mixed with 3.0 g stearic acid, and this was mixed for 1 minute in the blender. A pellet was prepared from this mixture by pressing in a pellet press for 20 seconds at 20,000 psi. The standards ranged from 0 to 5,000 ppm arsenic.

W X-ray tube

40 Kv

20 mA

Air path

LiF Crystal

Arsenic $K_\alpha$, 33.98° 2 ⊖

Pulse Height Analysis

Scintillation detector                 950 v

Base level                           17.6 v

Window width                  28.0 v

Counting time               50 seconds

     Three 50-second counts were taken at 33.0, 33.98 and 35.0 degrees
to determine background and peak intensities. The background at 33.98
degrees was estimated by interpolation from the background at 33.0 and
35.0 degrees. A standard curve of net counts per 50 seconds versus
arsenic concentration was prepared over the range 0 to 5,000 ppm. Under
these conditions, the detection limit was 40 ppm arsenic.

APPENDIX B

ARSENIC CONTENT OF GREENHOUSE SAMPLES

# CALCULATION OF POSSIBLE ARSENIC CONCENTRATION OF
# LETTUCE GROWN ON ARSENIC-RICH SOILS.

Assumptions:

1. As content of soils increases with long-term exposure to arsenic-rich waters, achieving a value of 1,250 ppm (like that observed for rocks and sediments of Ester Dome).

2. The As content of lettuce is directly proportional to the As content of the soil on which it is grown. A probable ratio for As/g dried plant to As/g soil is 0.1 (see Table B1).

3. Lettuce contains 90% water by weight.

4. Average serving of lettuce weighs 50 g (Marguerite Stetson, personal communication).

Calculations:

From Assumptions 1 and 2, we find

$$1{,}250 \; \frac{\mu g \; As}{g \; soil} \times \frac{0.1 \; \mu g \; As/g \; dry \; lettuce}{1 \; \mu g \; As/g \; soil}$$

= 125 µg As/g dried lettuce.

From Assumption 3, 10 g of wet lettuce corresponding to a dry weight of 1 g.

Thus a person <u>could</u> ingest 125 µg As/10 g wet lettuce.

If the average lettuce serving/person is 50 g (Assumption 4), then an average daily serving of wet lettuce would yield <u>625</u> µg As.

TABLE B1:  ARSENIC CONTENT OF GREENHOUSE SAMPLES.

| Plant | As(AA) | As(NAA) | Soil (-100 mesh) Ave. As(mg/g) | | Total Added As (mg) |
|---|---|---|---|---|---|
| | | | Top Half | Bottom Half | |
| Lettuce Leaves | <2 | ≤1.7 | 14.8 | 14.8 | 0.0 |
| "          " | 3.8 | | " | " | " |
| "          " | 3.4 | | " | " | " |
| "          " | 17 | 16.4 | 36 | 29 | 70,000 |
| "          " | 4.4 | | " | " | " |
| "          " | 14 | | " | " | " |
| Radish Leaves | <2 | | 14.8 | 14.8 | 0.0 |
| "          " | <2 | | " | " | " |
| "          " | <2 | | " | " | " |
| "          " | 2.1 | | 45 | 30 | 80,500 |
| "          " | 17 | | " | " | " |
| "          " | 23 | | " | " | " |
| "      Roots | <2 | ≤0.60 | 14.8 | 14.8 | 0.0 |
| "          " | <2 | | " | " | " |
| "          " | <2 | | " | " | " |
| "          " | 5.3 | | 45 | 30 | 80,500 |
| "          " | 3.3 | 8.6 | " | " | " |
| "          " | 5.7 | | " | " | " |
| Cucumber Leaves | <2 | | 14.8 | 14.8 | 0.0 |
| "          " | 2.9 | | " | " | " |
| "          " | 4.1 | | " | " | " |
| "          " | 4.1 | | 47 | 30 | 52,500 |
| "          " | 6.4 | | " | " | " |
| "          " | 4.0 | | " | " | " |
| Cucumber Fruit | <2 | ≤0.49 | 14.8 | 14.8 | |
| "          " | <2 | | " | " | |
| "          " | <2 | | " | " | |
| "          " | <2 | | 47 | 30 | 52,500 |
| "          " | <2 | 1.43 | " | " | " |
| "          " | <2 | | " | " | " |
| Tomato Leaves | <2 | | 14.8 | 14.8 | 0.0 |
| "          " | <2 | ≤1.1 | " | " | " |
| "          " | <2 | | " | " | " |
| "          " | 2.8 | | 200 | 42 | 182,000 |
| "          " | 2.9 | 1.64 | " | " | " |
| "          " | 2.8 | | " | " | " |
| "      Fruit | <2 | | 14.8 | 14.8 | 0.0 |
| "          " | <2 | | " | " | " |
| "          " | <2 | | " | " | " |
| "          " | <2 | | 200 | 42 | 182,000 |
| "          " | <2 | | " | " | " |
| "          " | <2 | | " | " | " |

APPENDIX C

DATA FROM BURTON ET AL. (1978)

Because the results of this study were not published and because they bear on this work, the data from Burton et al. (1978) are included here.

Stream sediments: The data from our study and those of Burton et al. are not strictly comparable because the entire stream-sediment sample was analyzed in our study while only the -100 mesh fraction was analyzed in the Burton et al. study. Our study yields a value for the total arsenic in the sample while that of Burton et al. is for the acid-soluble fraction of the -100 mesh part of the sample.

Surface Waters: The Burton et al. data are for May 1977 during a period of high flow and high suspended-sediment load. Our data are for August 1978 during a period of low flow and low suspended-sediment load. The most significant difference is in the treatment of the samples. Unfortunately, because of the high suspended-sediment load, Burton et al. found it very difficult to filter the samples in the field. To avoid loss of arsenic from solution by coprecipitation with iron, it was necessary to acidify the samples immediately. Thus, the samples were acidified in the field and then filtered in the laboratory. This treatment resulted in arsenic being dissolved from the suspended material, yielding arsenic concentrations in the water that are too high.

In our study, we were able to filter the sample in the field prior to acidification because of the light sediment load. This resulted in significantly lower observed concentrations of arsenic.

Vegetation: There was only a slight difference in the two methods used to ash vegetation in our two studies. The results of our study and Burton et al. should be comparable.

TABLE C1: STREAM DATA.

| Sample Number | Water (ppb) | Bottom Sediments (ppm) | Suspended Lode (L=low, H=high) | pH of Water | Temp. (°F) | Collection Date | Distance Downstream From 003 (feet) | Comments |
|---|---|---|---|---|---|---|---|---|
| A | 16.6 | 511 | L | | | 5/5/77 | Short tributary off map | Roadside creek leaves road. Creek absorbed into vegetation. |
| B | -- | 345 | L | | | " | | |
| 1 | 60.9 | a | L | 5.1 | 34 | " | Start right fork | Water on snow |
| 2 | 41.5 | 7,184 | L | 5.1 | 33 | " | 211 | " |
| 3 | 27.9 | 1,824 | L | 5.1 | 34 | " | 475 | " |
| 4 | 751.0 | 10,240 | H | 5.1 | 33 | " | 713 | " |
| 5 | 658.4 | 6,824 | L | 5.1 | 33 | " | 950 | " |
| 6 | 569.4 | a | L | 5.1 | 34 | " | 1,161 | " |
| 7 | 1,219.0 | a | L | 5.2 | 34 | " | 1,293 | " |
| 8 | 979.6 | 2,790 | L | 5.2 | 34 | " | 1,531 | " |
| 9 | 1,169.0 | 1,764 | L | 5.2 | 34 | " | 1,779 | " |
| 10 | 646.9 | 2,010 | L | 5.2 | 34 | " | 2,054 | " |
| 11 | 664.0 | 1,360 | L | 5.2 | 35 | " | 2,307 | Small meadow. |
| 12 | 789.0 | 2,719 | L | 5.2 | 35 | " | 2,492 | |
| 13 | 593.8 | 1,680 | L | 5.2 | 35 | " | 2,740 | |
| 14 | 978.6 | 1,088 | L | 5.2 | 34 | " | 2,962 | |
| 15 | 1,004.6 | a | L | 5.2 | 33 | " | 3,147 | East fork joins. |
| 16 | 815.0 | a | L | 5.2 | 32 | " | 3,405 | |
| 17 | 1,170.4 | a | L | 5.2 | 32 | " | 3,659 | |
| 18 | 983.2 | 1,196 | L | 5.2 | 33 | " | 3,907 | Beginning of valley bottom meadow. |
| 19 | 72.1 | a | | | | 5/16/77 | 4,166 | Small tributary joins from east. |
| 20 | 82.8 | a | | 5.2 | 32 | " | 4,351 | |
| 21 | 74.5 | a | L | 5.2 | 32 | " | 4,593 | |
| 22 | 75.4 | a | H | 5.2 | 32 | " | 4,757 | |
| 23 | 68.5 | 12,008 | H | 5.3 | 32 | " | 4,942 | High water sediment sample. |
| 24 | 171.6 | a | L | 5.3 | 32 | " | 5,185 | |
| 25 | 144.2 | 112 | L | 5.4 | 33 | " | 5,443 | |
| 26 | 153.9 | a | L | 5.4 | 33 | " | 5,665 | |
| 27 | 112.1 | 16,808 | L | 5.5 | 34 | " | 5,813 | |
| 28 | 119.6 | 4,372 | L | 5.5 | 34 | " | 6,077 | |
| 29 | 103.2 | 5,960 | L | 5.5 | 34 | " | 6,288 | Pond, creek partially diverted. |
| 30 | 103.2 | 6,180 | L | 5.5 | 39 | " | 6,531 | |
| 31 | 73.5 | 10,636 | L | 5.5 | 36 | " | 6,795 | |
| 32 | 97.2 | 6,416 | L | 5.5 | 37 | " | 7,006 | |
| 33 | 81.6 | 2,114 | L | 5.6 | 37 | " | 7,191 | Top of waterfall into eastern dredge pit. |
| 34 | 141.2 | a | H | 5.4 | 34 | 5/19/77 | 7,376 | Base of falls. |
| 35 | 200.1 | a | H | 5.3 | 34 | " | 7,587 | Beginning of eastern dredge pit. |
| 36 | 180.7 | a | | 5.4 | 34 | " | 7,709 | Creek running on snow |
| 37 | 161.9 | a | | 5.4 | 34 | " | 7,915 | " |
| 38 | 223.5 | 1,003 | | 5.4 | 34 | " | 8,020 | Dredge tailings begin. |
| 39 | 294.7 | 5,040 | | 5.3 | 34 | " | 8,142 | |
| 40 | 97.2 | 5,484 | L | 5.3 | 34 | " | 8,321 | Dredge tailings end. |
| 41 | 129.6 | 205 | L | 5.3 | 34 | " | 8,453 | |
| 42 | 127.9 | 268 | L | 5.3 | 34 | " | 8,611 | |
| 43 | 228.7 | 246 | | 5.4 | 34 | " | 8,728 | |
| 44 | 322.5 | 280 | | 5.4 | 33 | " | 8,817 | |
| 45 | 222.6 | 321 | H | 5.4 | 34 | " | 8,934 | |
| 46 | 272.4 | 184 | L | 5.5 | 34 | " | 9,176 | |
| 47 | 95.8 | | L | | 33 | 5/12/77 | | Creek along Henderson Road |
| 48 | 50.0 | 272 | H | | 33 | " | | " |
| 49 | 26.7 | 312 | | | 33 | " | | " |
| 50 | 31.4 | 117 | H | | 33 | " | | " |
| 51 | 27.9 | 300 | H | | 33 | " | | " |
| 52 | 50.9 | 201 | H | | 33 | " | | " |
| 53 | 36.4 | 284 | L | 5.1 | 34 | " | | East fork, Eva Creek. Nearly stagnant. |
| 54 | 443.7 | 213 | L | 5.1 | 34 | " | | " |
| 55 | 15.5 | 299 | L | | 34 | " | | Below Clipper Mine. |
| 56 | 21.6 | 349 | L | | | " | | " |
| 57 | 22.2 | 419 | L | 5.2 | | " | | " |
| 58 | 11.6 | 238 | L | 5.2 | 45 | " | | Nearly stagnant. |
| 59 | 119.3 | 4,268 | L | 5.7 | 36 | 5/18/77 | | Pond at end of swampy area, beginning of Little Eva #1. |
| 60 | 66.0 | | H | 5.4 | 34 | " | | Little Eva #1 Just before Parks Hgwy. |
| 61 | 40.3 | | H | 5.4 | 34 | " | | " |
| 62 | 44.7 | | H | 5.4 | 34 | " | | " |
| 63 | 38.0 | | H | 5.4 | 34 | " | | " |
| 64 | 30.9 | | H | 5.4 | 34 | " | | Nearly stagnant. |
| 65 | 76.8 | | L | 5.5 | 34 | 5/19/77 | | " |
| 66 | N.D | | H | 5.5 | 34 | " | | Large tailings pile on hillside above creek. Opposite "Little Eva Mine". |
| 67 | 51.2 | | | 5.5 | 34 | " | | " |
| 68 | 49.2 | | H | 5.6 | 34 | " | | " |
| 69 | 33.9 | | H | 5.4 | 34 | 5/18/77 | | " |
| 70 | 31.2 | | H | 5.4 | 34 | " | | " |
| 71 | 32.2 | | H | 5.3 | 34 | " | | " |
| 72 | 32.7 | | H | 5.3 | 34 | " | | " |
| 73 | 22.9 | | H | 5.3 | 34 | " | | Nearly stagnant. |
| 74 | 54.9 | | H | 5.2 | 34 | " | | Just before Parks Hgwy. |
| 75 | 90.9 | | L | 5.1 | 34 | " | | " |
| 76 | N.D | 1,360 | L | 5.4 | 40 | " | | Little Eva #2 |
| 77 | 10.3 | 1,592 | L | 5.3 | 40 | " | | Beginning of Little Eva #2. |
| 78 | 11.0 | 1,558 | L | 5.5 | 40 | " | | Top of cascade into western dredge pit. |
| 79 | 41.7 | 2,208 | L | 5.5 | 40 | " | | " |
| 80 | 24.8 | 1,484 | L | 5.6 | 40 | " | | " |
| 81 | 26.6 | 1,732 | L | 5.6 | 40 | " | | " |
| 82 | 22.4 | 1,478 | L | 5.6 | 40 | " | | " |
| 83 | 20.5 | 1,512 | L | 5.5 | 40 | " | | " |
| 84 | 25.1 | 1,488 | L | 5.5 | 40 | " | | Nearly stagnant. |
| 85 | 13.3 | 1,460 | L | 5.5 | 40 | " | | Spring has much organic matter. |
| 86 | 313.8 | | | | 33 | 5/12/77 | | |
| 87 | 13.1 | | | | 33 | " | Spring | |
| 88 | 86.1 | | H | 5.5 | 32 | " | 3,094 | 10 ft below Fe-rich spring which flows into main creek at 017. |
| 89 | | | H | 5.6 | 32 | " | 3,090 | Source of spring, smells of H₂S, dry creekbed above spring. |
| 90 | 175.0 | a | L | 5.2 | 33 | 5/16/77 | Water on snow | Sample from tributary that joins at 020. |
| 91 | 11.6 | a | L | 5.2 | 42 | " | | Stagnant spring. |
| 92 | 36.8 | 6,328 | L | 5.6 | 34 | 5/18/77 | | Stagnant pond. |
| 93 | 32.0 | 4,612 | L | 5.4 | 38 | " | | Spring 100 ft west of 047 |
| 94 | 28.8 | a | H | 5.3 | 34 | " | | Stagnant pool midway down wonderloess fall, main creek. |

a Bottom sediment samples unobtainable.

TABLE C2: ARSENIC CONTENT OF PLANTS FROM ESTER DOME.

| Common Name | Scientific Name | Mean Arsenic (ppm) | High Value Arsenic (ppm) |
|---|---|---|---|
| Alder | Alnus spp. | 27.90 | 117.5 |
| Aspen | Populus fremuloides | 29.57 | 56.1 |
| Labrador Tea | Ledum spp. | 22.50 | 52.50 |
| White Spruce | Picea glavea | 13.52 | 38.52 |
| Willow | Salix spp. | 14.79 | 31.71 |
| Fireweed | Epilobium angustifolium | 12.01 | 26.49 |
| Bunchberry | Cornus canadensis | 11.97 | 21.12 |
| Rose | Rose acicularis | 13.03 | 18.97 |
| Cranberry | Vaccinium vitis-idaea | 13.74 | 17.66 |
| Birch | Betula papyrifera | 15.08 | 16.35 |
| Grass | Gramineae spp. | 11.33 | 13.66 |
| Juniper | Juniperus horizontalis | 12.20 | 12.62 |
| Moss | | 2.67 | 2.67 |
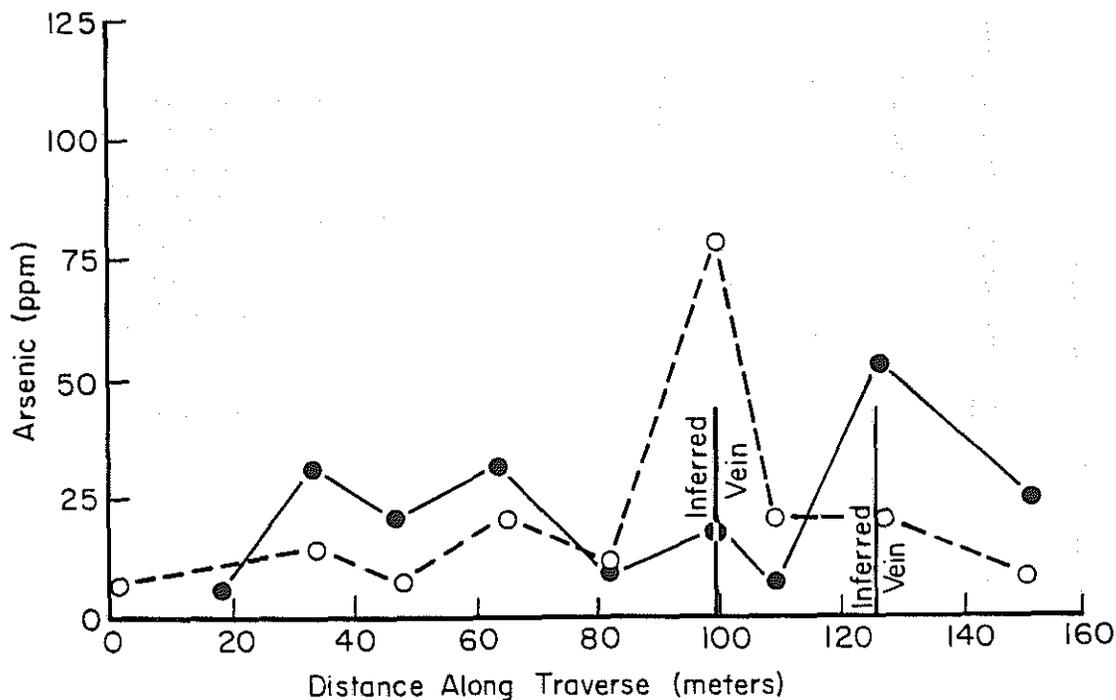


Figure C1. Arsenic concentrations in alder and Labrador tea along middle traverse of Ryan Lode.
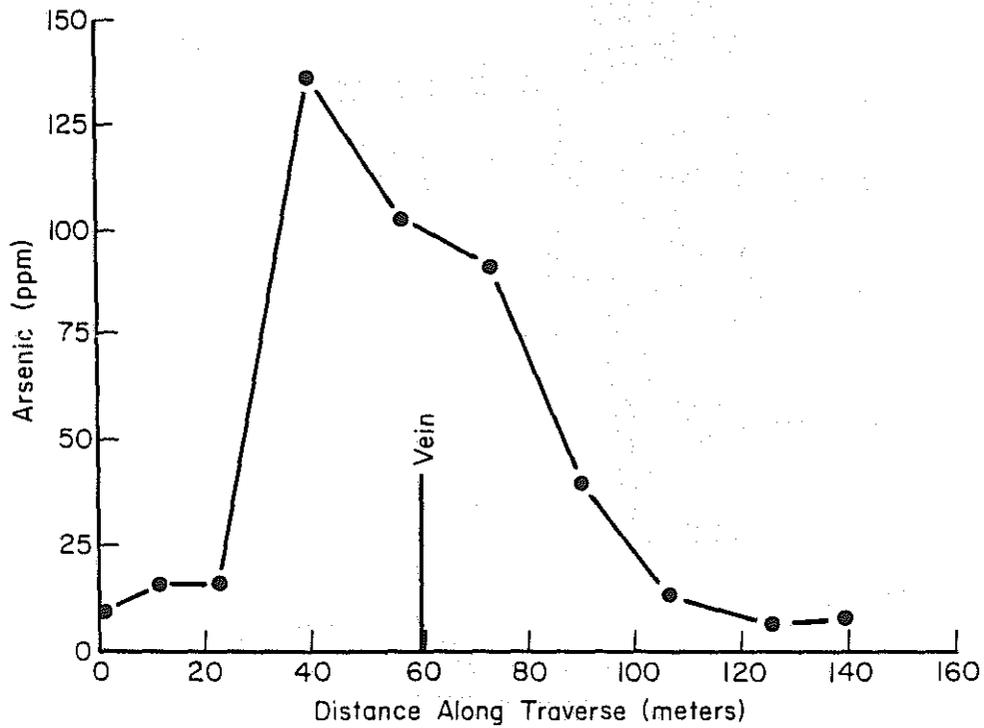
Figure C2.   Arsenic concentrations in alder along upper
traverse of Ryan Lode.
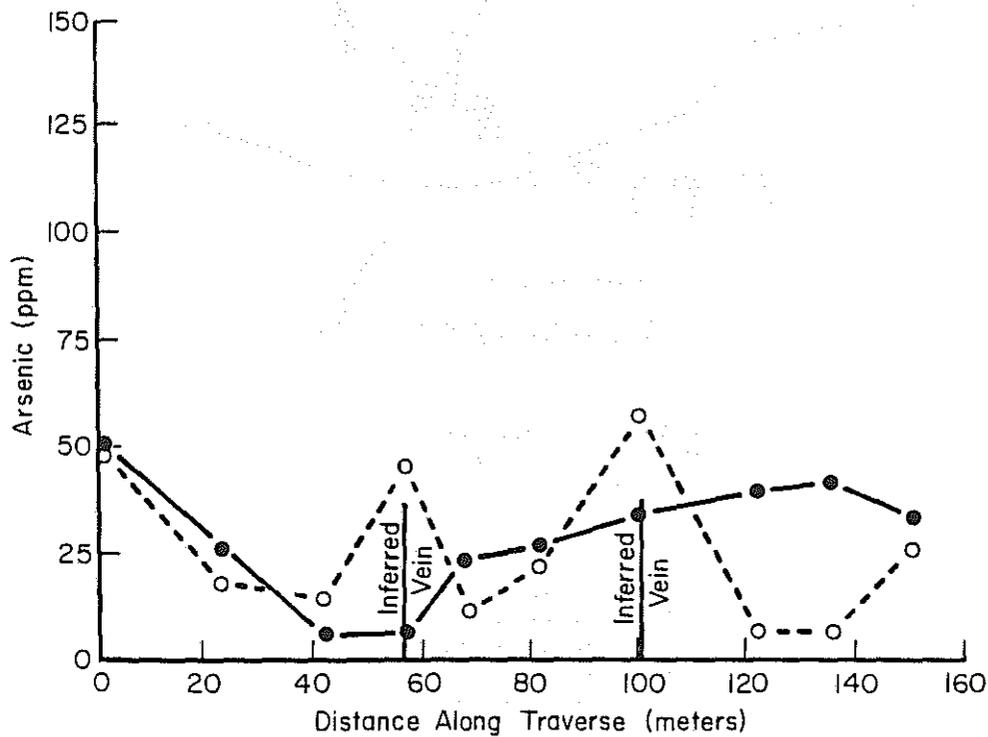


Figure C3.   Arsenic concentrations in alder and Labrador tea
along lower traverse of Ryan Lode.
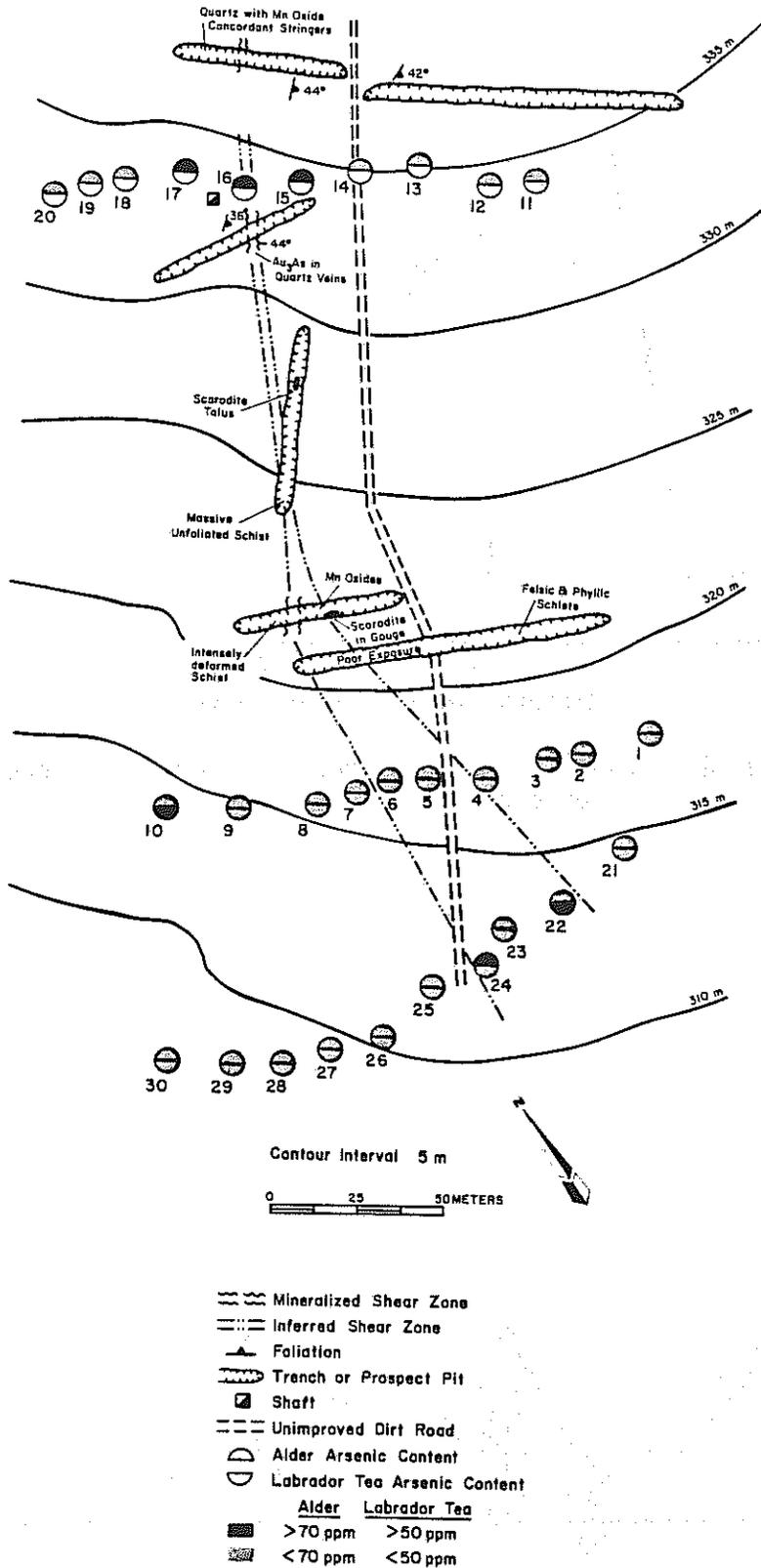
Figure C4. Biogeochemical traverses of
Ryan Lode.