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SULPHUR IN ARCHEAN VOLCANIC ROCKS OF THE CANADIAN SHIELD

E. M. CAMERON

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ABSTRACT

A total of 1098 samples of Archean volcanic rocks from four localities in the Canadian Shield have been analyzed for sulphur. Two of these areas - Noranda, Quebec and High Lake, Northwest Territories - contain massive sulphide deposits of copper and zinc. The other two localities - Yellowknife and Indin Lake, Northwest Territories - are not known to contain massive sulphides, but do contain gold deposits. The samples have been classified into basic, intermediate or acidic groups on the basis of their chemically-determined silica contents.

The frequency distributions of sulphur in these rocks are approximately lognormal. The distribution curves of the samples of basic rocks from Noranda, from Yellowknife, and from Indin Lake are similar and the average sulphur contents of these three groups are close to the limit of solubility of sulphur in basaltic melts of $\sim 0.1\%$. The sulphur contained in the original melt has probably been largely retained in the lavas as a result of rapid submarine quenching.

The cumulative frequency distribution curves for acidic rocks from Yellowknife and Indin Lake are parallel to those for basic rocks from the same areas, but at much lower sulphur levels. Curves for intermediate rocks lie midway between the basic and acidic groups. The lower sulphur content of the more siliceous volcanic rocks may, in part, be caused by a lower sulphur solubility in silica-rich melts. The major cause is, however, likely to be loss of sulphur by degassing, for many of the more silicic volcanic sequences contain a high proportion of pyroclastic rocks.

For the Noranda and High Lake samples the right-hand side of the frequency distributions are skewed to higher sulphur levels. This effect is greater for acidic than for basic samples. The net result is that for these two areas, it is the acidic groups that contain more sulphur than the basic samples. This finding is in accord with the long-known association of massive sulphide deposits with silicic volcanic rocks. It is caused by the introduction of sulphides into nearby volcanic rocks during the formation of massive sulphide deposits.

The sulphur content of Archean volcanic rocks may provide some basis for distinguishing mineralized from barren sequences. It is by no means certain, however, that such methods are more practical than surficial geochemical methods or lithogeochemical techniques based on major element changes around massive sulphide deposits.

RESUME

Un total de 1098 spécimens de roches volcaniques archéennes provenant de quatre régions du Bouclier canadien ont été analysées au point de vue de leur teneur en soufre. Deux de ces régions - Noranda, dans la province de Québec, et High Lake, dans les Territories du Nord-ouest - renferment des gîtes sulfurés massifs de cuivre et de zinc. On ne croit pas que les deux autres régions - Yellowknife et Indin Lake, dans les Territoires du Nord-Ouest - renferment de grandes quantités de sulfures, mais en revanche elles renferment des gîtes d'or. Les spécimens ont été classés en trois groupes, basique, intermédiaire et acide, sur la base de leur teneur en silice déterminée chimiquement.

La distribution en fréquence du soufre dans les roches en question est à peu près logarithmiquement normale. Les courbes de distribution des spécimens de roches basiques provenant de Noranda, de Yellowknife et d'Indin Lake sont similaires et le contenu sulfureux moyen de ces trois groupes est proche de la limite de solubilité du soufre dans des fontes basaltiques de - 0.1%. Le soufre contenu dans la fonte initiale a probablement été largement retenu dans la lave à la suite de l'extinction sous-marine rapide.

Les courbes de distribution de fréquence cumulatives se rapportant aux roches acidiques provenant de Yellowknife et d'Indin Lake sont parallèles à celles des roches basiques des mêmes zones, mais les niveaux de soufre sont plus bas. Les courbes correspondant aux roches intermédiaires se situent à michemin entre les groupes basique et acidique. Il se peut que la teneur plus faible en soufre des roches volcaniques plus siliceuses soit causée en partie par une solubilité moindre du soufre dans les fontes riches en silice. Cependant, il est probable que la cause principale de ce phénomème est une perte de soufre par dégazage, car un grand nombre de séquences volcaniques plus siliceuses renferment une proportion élevée de roches pyroclastiques.

En ce qui concerne les spécimens en provenance de Noranda et de High Lake, la partie droite de la courbe de distribution de fréquence dévie pour indiquer des niveaux de soufre plus élevés. Cet effet est plus marqué dans le cas des spécimens acidiques que dans le cas des spécimens basiques. En dernière analyse, dans ces deux zones, ce sont les groupes acidiques qui renferment plus de soufre que les spécimens basiques. Cette découverte s'accorde avec l'association, connue de longue date, de gîtes massifs de sulphure à des roches volcaniques siliceuses. On l'attribue à l'introduction de sulphures dans les roches volcaniques voisines au cours de la formation des gîtes massifs sulfurés.

La teneur en soufre des roches volcaniques archéennes peut fournir une certaine base pour établir une distinction entre les séquences

minéralisées et les séquences stériles. Rien ne prouve cependant que cette méthode soit plus pratique que les méthodes géochimiques subaériennes ou les techniques lithogéochimiques fondées sur la modification des éléments principaux autour des gîtes massifs de sulphure.

SULPHUR IN ARCHEAN VOLCANIC ROCKS OF THE CANADIAN SHIELD

Introduction

In spite of the economic importance of sulphide ores, there have been relatively few studies of the distribution of sulphur in mineralized or barren rocks. It is the purpose of this paper to provide some basic data on the distribution of sulphur in Archean volcanic rocks from four localities in the Canadian Shield (Fig. 1). The rocks from two of these areas are hosts to massive sulphide deposits, and gold deposits occur in three of the areas. The samples analyzed have been collected over the past few years for a variety of purposes.

The writer and his colleagues have previously studied the distribution of sulphur in ultramafic rocks of the Shield (Cameron et al., 1971). Some of the ultramafic bodies are associated with nickel-copper sulphide ores that are believed to have segregated from the original magma. These ultramafic bodies contain small amounts or "microdeposits" of copper and nickel sulphides that are genetically related to the larger ore segregations. On analyzing rock samples from these bodies the presence of the microdeposit population is revealed by a characteristic skewing of the frequency distribution for sulphur. The frequency distributions may, therefore, be used to assess the ore potential of ultramafic rocks.

In the case of massive sulphide and gold deposits of the Canadian Shield, their association with volcanic host rocks is not as direct as with the ultramafic rocks and nickel-copper sulphides. These deposits do not appear to be the immediate product of segregation from adjacent volcanic rocks and thus there may not be as direct a relationship between the sulphur content of these volcanic rocks and the presence of nearby sulphide ores.

Since it was thought likely that the sulphur content would vary with the basicity of the volcanic rock samples, they were analyzed for silica as well as sulphur. This allowed the samples to be placed in one of three groups for comparing their sulphur content: basic (40.0 - 54.9% SiO_2), intermediate (55.0 - 67.9% SiO_2) or acidic ($\geq$ 68.0% SiO_2). Field classification into similar groups tends to be inaccurate.

Acknowledgements

The writer would like to thank W.R.A. Baragar for critically reading this manuscript.

Sampling

Noranda, Quebec. 701 fist-sized samples of volcanic rock were collected along a north-south and an east-west traverse through the Noranda area (Fig. 2). Where outcrop allowed, the samples were collected at regular 100-foot intervals. This area contains a number of massive sulphide base metal deposits and also gold deposits associated with the volcanic rocks.

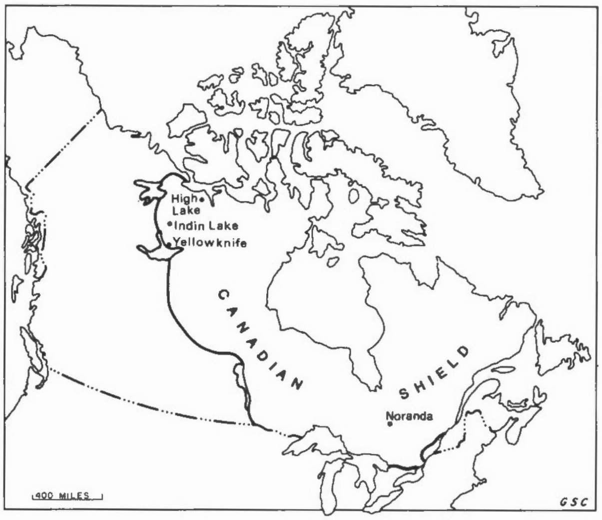

Figure 1. Sampling locations in the Canadian Shield.

High Lake, N.W.T. This area contains an undeveloped deposit of 5.2 million tons of 3.5% Cu and 2.5% Zn. 94 samples were collected at regular 100- or 200-foot intervals along three traverses normal to the strike of the volcanic rocks (Fig. 3). Two of these traverses are relatively close to the deposit; the third is two miles to the south.

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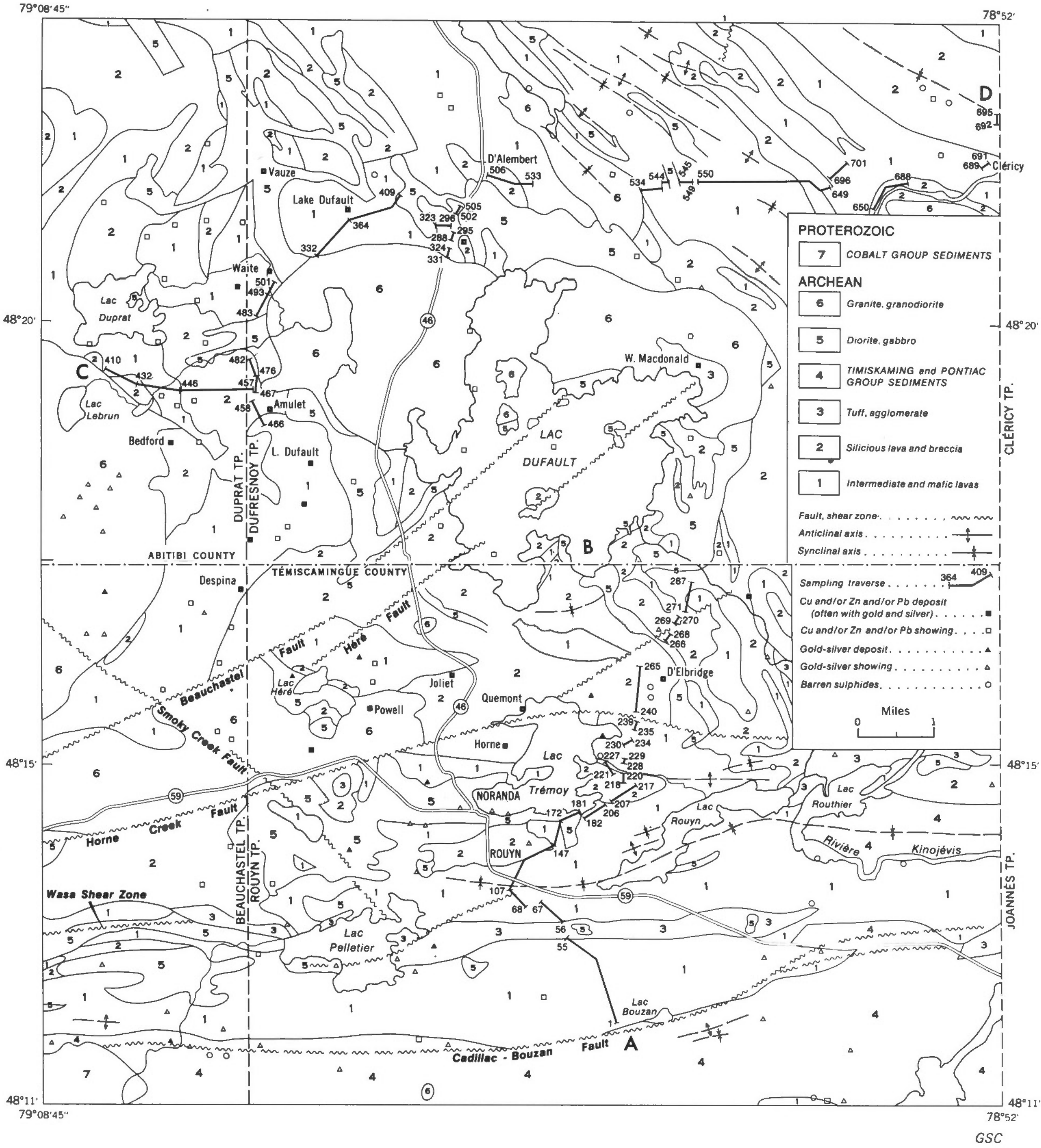

Figure 2. Sampling traverses, Noranda, Quebec. Geology after J.I. Sharpe (Quebec Department of Mineral Resources Map 1653, 1967).

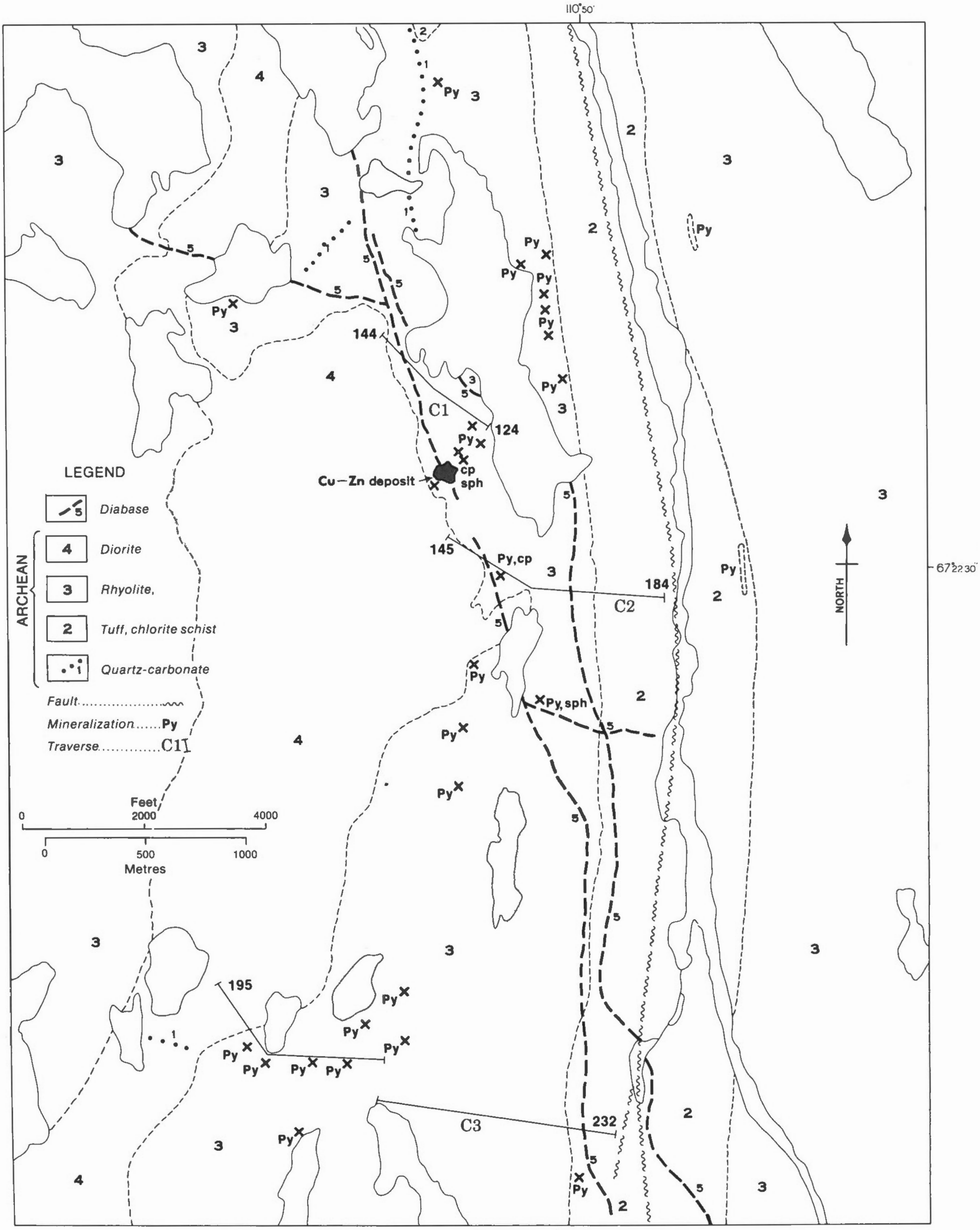

Figure 3. Sampling traverses, High Lake, N. W. T. Geology after E. O. Dearden, and B. A. Bradshaw, Kennarctic Explorations, Ltd.

Indin Lake, N.W.T. 109 volcanic rock samples were collected at regular 100-foot intervals along a number of traverses in this area (Fig. 4). As far as is known there are no massive sulphide deposits in the area, but there are abandoned gold workings that commonly occur in shear zones in volcanic rocks and sediments.

Yellowknife Area, N.W.T. 194 samples were collected by W.R.A. Baragar from the immediate vicinity of Yellowknife and from the nearby Cameron River belt. The sampling locations are shown in Baragar (1966) and in Cameron and Baragar (1971). This region is not known to contain massive sulphide bodies, but there are important gold deposits at Yellowknife.

Analysis

Sulphur. Sulphur was analyzed by heating a mixture of the sample and iron chips to 1450°C in an oxygen stream. The SO_2 evolved is absorbed in a solution containing hydrochloric acid, potassium iodide, potassium iodate and starch indicator. As the SO_2 is evolved, potassium iodate is added automatically to maintain a constant blue intensity. The method is described by Bouvier et al. (1972). The limit of detection is 0.01% S. For computation purposes, samples with less than this limit are assigned an arbitrary value of 0.005% S.

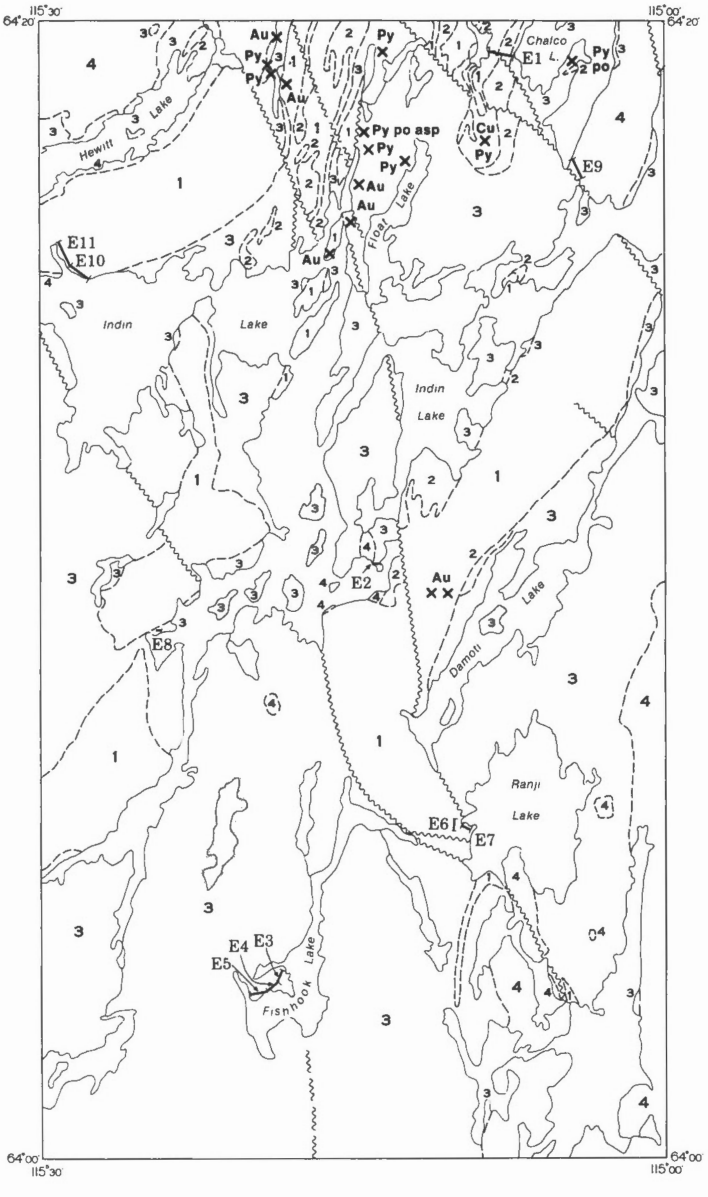

Figure 4. Sampling traverses, Indin Lake, N.W.T. Volcanic rocks from traverses E1, E2, E6, E7, E9, E10 and E11. Geology after M.S. Stanton, L.P. Tremblay, D.H. Yardley, G.M. Wright and M.L. Miller (GSC Maps 1022A and 1023A, 1954).

Silica. Two quite different methods were used for the determination of silica. The High Lake and Indin Lake samples were analyzed by direct-reading emission spectrometer. After fusion of the samples with lithium tetraborate containing strontium tetraborate and cobalt oxide as stabilizers and internal standards, the powder was sparked on a "Tape machine" of an A.R.L. Quantometer. Accuracy is approximately ±1% SiO_2.

The Noranda and Yellowknife samples were analyzed by X-ray fluorescence. The untreated powder was excited by a chrome-target tube and the SiK_α line measured. Accuracy is approximately ±2% SiO_2.

Results

Indin Lake and Yellowknife. The frequency distributions for sulphur in these areas are given in Figures 5 and 6 respectively. For Indin Lake and Yellowknife the distributions for basic, intermediate and acidic samples form three nearly parallel curves lying at decreasing sulphur values with increasing silica content. The distributions appear to be lognormal since the logarithmically-plotted percentile values lie along a straight line. Lognormal distribution is most apparent for the basic group because the scatter around the line is small. It is less clear for the acidic and intermediate distributions that show a greater scatter, and a greater proportion of samples close to or below the detection limit for sulphur.

For the Yellowknife samples, the frequency distributions are similar to Indin Lake. One notable difference is that the curve for intermediate rocks is skewed upwards to higher sulphur values, possibly indicative of sulphur mineralization in these samples. Relative to Indin Lake, the basic samples from Yellowknife are somewhat poorer in sulphur (Table 1).

Noranda and High Lake. The frequency distributions for sulphur in these areas are given in Figures 7 and 8. These mineralized volcanic sequences have distributions that are substantially different from the Indin Lake and Yellowknife areas which do not contain massive sulphide bodies. The distribution curves are steeper, indicative of a higher dispersion or variance of the values, and continue up to much higher sulphur levels. These differences are greater for the more siliceous samples, so that the sequence of decreasing sulphur levels, basic, intermediate and acid, noted above for the Indin Lake and Yellowknife volcanics, is reversed in the upper parts of the Noranda and High Lake distribution curves.

The frequency distribution for basic samples from Noranda is no different from those of the above unmineralized areas; in fact sulphur is more abundant in the basic samples from Indin Lake (Table 1). Sulphur is very abundant in the High Lake samples and here even many of the basic samples are mineralized.

In Figure 9 the frequency distributions for the combined volcanic samples from the four areas are compared. The mineralized sample sets from High Lake and Noranda are noticeably skewed relative to those from Yellowknife and Indin Lake. It is of interest to note that the cumulative frequency distributions for the latter two areas lie along a straight line. They show what appears to be a near-perfect lognormal distribution, although, in fact, they are composed of a mixture of separate distributions (Figs. 5 and 6).

Discussion

The geochemistry of sulphur in volcanic rocks is likely to be much more complex than that of the rock-forming elements. Sulphur contained in molten rocks may be lost either in the vapour state or in solution during the eruption and subsequent cooling of the lavas. Further, sulphur may be introduced into permeable lavas by sulphur-bearing waters or by those mineralization processes accompanying the formation of sulphide ores.

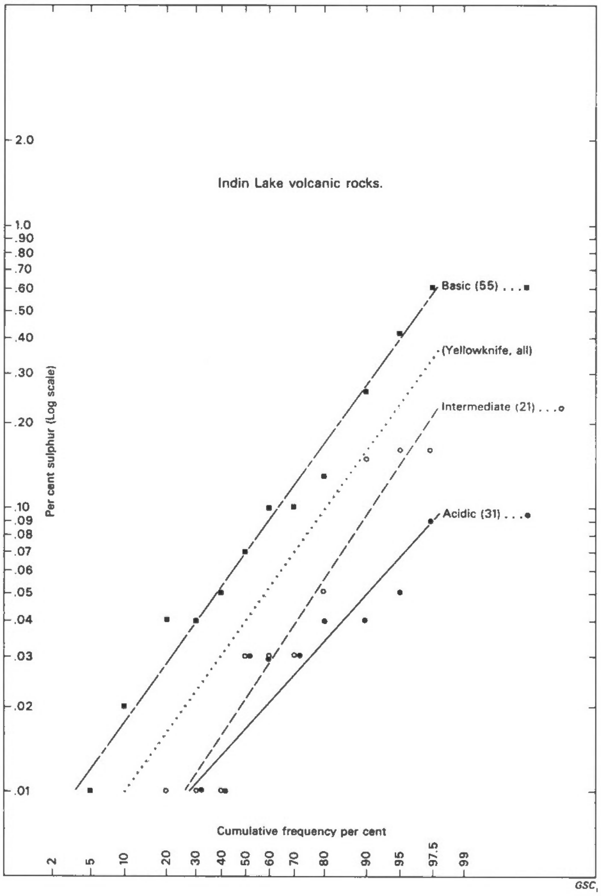

Figure 5. Frequency distributions for sulphur, Indin Lake volcanic rocks.

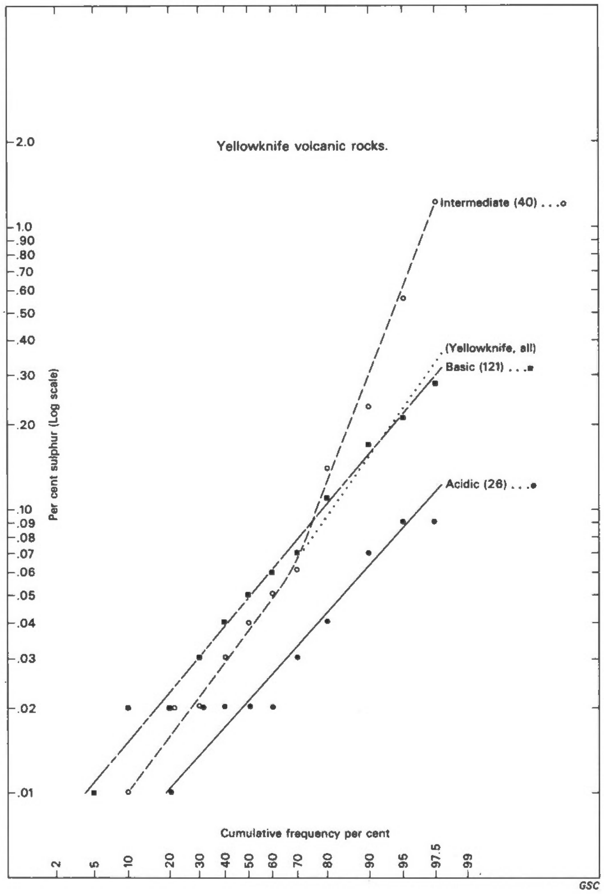

Figure 6. Frequency distributions for sulphur, Yellowknife volcanic rocks.

TABLE 1. Statistical data on sulphur (as per cent) in volcanic rocks from the Canadian Shield.

Location		Number of Samples	Arithmetic Mean	Standard Deviation	Geometric Mean	Median
Indin Lake:	All samples	109	.067	.093	.036	.04
	Basic	55	.103	.117	.065	.07
	Intermediate	21	.034	.043	.020	.03
	Acidic	31	.025	.019	.017	.03
Yellowknife:	All samples	194	.072	.117	.042	.04
	Basic	121	.072	.078	.048	.05
	Intermediate	40	.102	.214	.043	.04
	Acidic	26	.027	.022	.021	.02
Noranda:	All samples	701	.119	.318	.043	.04
	Basic	214	.091	.182	.043	.05
	Intermediate	372	.127	.357	.045	.04
	Acidic	114	.124	.295	.037	.02
High Lake:	All samples	94	.205	.542	.044	.04
	Basic	22	.144	.421	.031	.04
	Intermediate	54	.177	.555	.046	.04
	Acidic	16	.249	.490	.045	.04

Note: Basic plus intermediate plus acidic do not equal total samples because some samples measure less than 40% SiO_2.

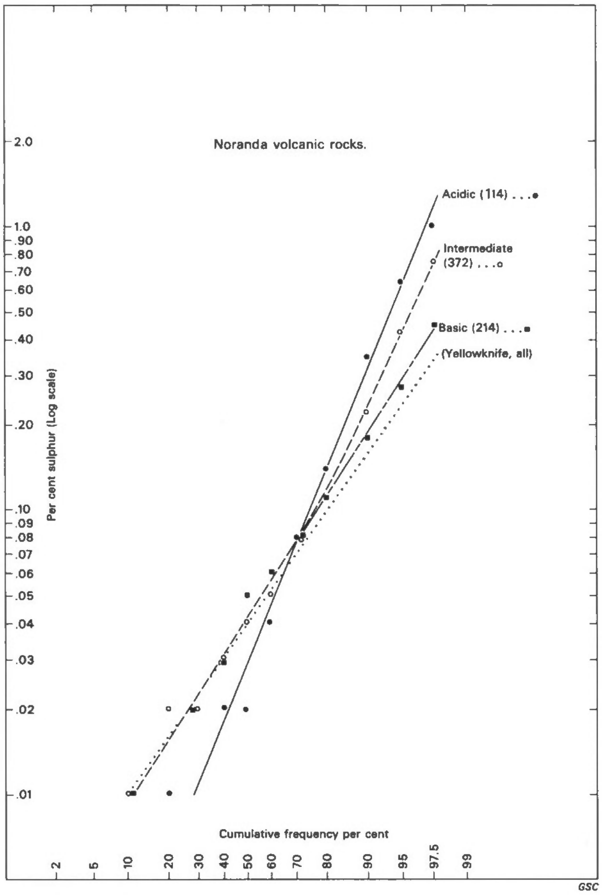

Figure 7. Frequency distributions for sulphur, Noranda volcanic rocks.

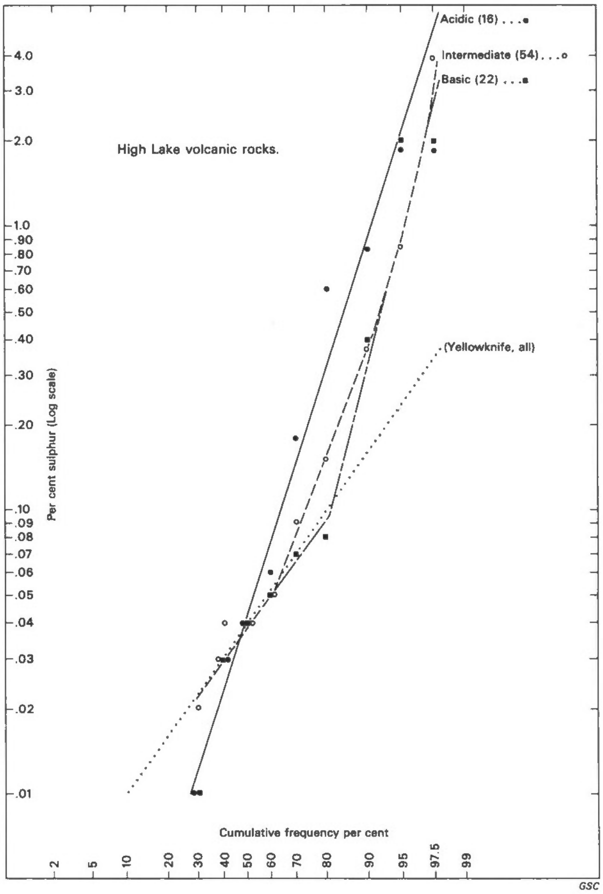

Figure 8. Frequency distributions for sulphur, High Lake volcanic rocks.

Rapid quenching of a lava under pressure inhibits the vesiculation that permits degassing or that provides the permeability required for soaking the rock with sulphur-bearing solutions or gases. Moore and Fabbi (1971) showed that basalts that were erupted on the ocean floor and were thus rapidly quenched contain more sulphur (0.068% S for 38 analyses) than those erupted subaerially (0.011% S for 9 analyses). Sulphur is more abundant in the glassy rims of basaltic pillows than in the more slowly cooled interior (0.078% S versus 0.054% S for 9 pairs of samples).

The mean sulphur content of the basic volcanic samples from Indin Lake, Yellowknife, and Noranda are, at 0.103% S, 0.072% S and 0.091%, similar or greater than the values quoted above for modern rapidly-cooled basalts. Most basalts from the Archean of the Canadian Shield are believed to have been extruded subaqueously (Baragar and Goodwin, 1969). Pillow structures are a common feature of the sequences sampled for this study, and of Archean basalts in general. These observations, plus the apparently homogeneous nature of the frequency distributions for basic rocks shown in Figures 5-7, suggest that the basic volcanic samples from Noranda, Indin Lake and Yellowknife contain close to the original sulphur content of the molten lava flow. Haughton et al. (unpublished) carried out experimental studies that allowed them to estimate the limit of sulphur solubility in basaltic melts at approximately 0.1% S. It would seem significant that the mean sulphur values quoted earlier in this paragraph are close to this value, indicating perhaps that they were derived from melts saturated in sulphur.

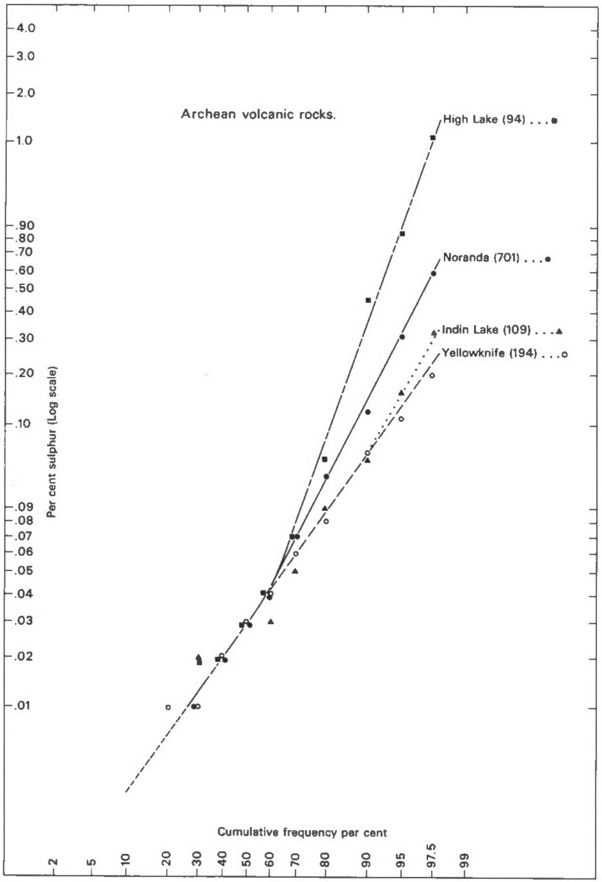

Figure 9. Frequency distributions for sulphur, Archean volcanic rocks.

The study of Haughton et al. showed that sulphur solubility of silicate melts is positively related to temperature and to the Fe, Ca, Mg, and Ti contents. It is inversely related to the oxygen fugacity and the Si, Al, Na and K contents. Silica-rich melts near their crystallization temperature should have a lower sulphur solubility than basaltic melts. This may, in part, be an explanation for the lower sulphur contents of the intermediate and acidic volcanic rocks from Indin Lake and Yellowknife. However, most acidic volcanic sequences from the Shield, including those sampled here, show a high proportion of pyroclastic rocks. This indicates that degassing, with probable sulphur loss, was taking place during eruption. The pyroclastic rocks are often permeable, allowing further sulphur loss after deposition, or, alternatively, gain of sulphur.

In the Shield, massive sulphide deposits containing zinc, copper, lead and silver, and also gold deposits, are associated with silicic volcanic rocks (Lang et al., 1970). Sangster (1972, p. 3) emphasized the close spatial association between acidic volcanic agglomerates or coarse pyroclastic rocks and massive sulphide ores. It is, therefore, not surprising that it is the acidic volcanic rocks which show the greatest difference in sulphur distribution between sequences that do and do not contain these deposits.

In the case of many of the more highly mineralized samples from High Lake and Noranda, the sulphides appear to have been introduced into preexisting volcanic rocks. These sulphides may be observed along fractures and bedding planes or in the vugs of altered volcanics. In the case of certain other samples it is difficult to tell whether the visible sulphide content is primary or secondary. The sulphides that were introduced into the volcanics were, no doubt, introduced along feeder pipes (Roscoe, 1965) or along shoots at the time of formation of the main sulphide body. The sulphide content of the samples from these mineralized areas is therefore governed by their distance from the deposit, rather than by stratigraphic controls, as might be the case with a high primary sulphide content in the lavas. Thus at High Lake, traverse C1, nearest the deposit, contains a mean of 0.48% S for 21 samples; traverse C2, 0.19% S for 36 samples; and traverse C3, most distant from the deposit, only 0.06% S for 37 samples. In the Noranda area the same is true. Sulphur is not uniformly distributed across the area, but is more

TABLE 2. Distribution of sulphur (as per cent) along sampling traverses in Noranda area. For location of samples see Figure 2. (N: number of samples; $\overline{X}$: arithmetic mean.)

Sample Number	ACIDIC		INTERMEDIATE		BASIC	
	N	$\overline{X}$	N	$\overline{X}$	N	$\overline{X}$
1 - 67	4	.03	26	.10	37	.07
68 - 107	0	-	11	.12	29	.06
108 - 147	7	.29	2	.07	31	.06
148 - 172	1	.65	8	.04	16	.08
173 - 181	0	-	4	.13	5	.12
182 - 217	1	.14	27	.10	8	.08
218 - 239	0	-	15	.48	6	.17
240 - 265	21	.31	4	.10	1	1.09
266 - 287	5	.09	16	.05	1	.04
288 - 331,502-505	4	.07	30	.08	14	.22
332 - 409	6	.06	70	.04	2	.03
410 - 457	12	.03	25	.12	11	.07
458 - 482	4	.16	21	.78	0	-
483 - 501	3	.04	10	.04	6	.17
506 - 533	3	.03	23	.09	2	.03
534 - 549	2	.11	7	.04	7	.18
550 - 649,696-701	30	.05	49	.06	27	.08
650 - 688	7	.07	21	.03	11	.02
689 - 695	4	.02	3	.12	0	-

abundant in volcanic rocks near the massive sulphides (Fig. 2 and Table 2). Acidic volcanics near the centre of the camp (Rouyn-Delbridge sector) are enriched in sulphur, but those sampled elsewhere are not. Intermediate volcanic samples are only enriched in part of the Rouyn-Delbridge sector and near the Amulet Mine. Basic samples contain higher amounts of sulphide only in an area 1/2 mile east of the Lake Dufault Mine. These data, of course, only confirm observations that may readily be obtained in the field.

Use of Sulphur For Exploration

During reconnaissance exploration, it would be advantageous to estimate the ore potential of a volcanic belt or area on the basis of a relatively small number of samples of volcanic rocks. Since the bulk of volcanic sequences in the Shield are composed of basaltic rocks (basaltic rocks, 60%; andesitic, 28%; salic, 12%; Baragar and Goodwin, 1969) it is further desirable that the reconnaissance methods be based, in part at least, on sampling basic rocks. The frequency distribution for basaltic rocks from Noranda, Yellowknife and Indin Lake are all quite similar. At first sight it may thus appear that there is no direct criterion of ore potential in the sulphur content of these rocks. However, all three sets of basic rocks show average sulphur levels near the maximum that can be held in solution in basaltic melts. It is possible that all three distributions indicate magma compositions suitable for the fractionation of sulphur-rich liquids. Although the three areas do not all have known massive sulphide deposits, they all do have gold deposits.

Turning now to the more silicic rocks, their frequency distribution curves are quite sensitive to the presence of sulphide mineralization. The acidic rocks from Indin Lake and Yellowknife have only a fractional percentage of the samples with 0.1% S or more, while 25 per cent of those from Noranda and High Lake contain this amount or greater. Systematic sampling of volcanic belts may possibly be a useful method of locating mineralized areas, but it should be emphasized that the data presented here indicate that only those volcanic rocks in the immediate vicinity of massive sulphide deposits contain higher amounts of sulphur. It is by no means certain that such an approach is more practical than surficial geochemical methods, like lake sampling, or lithogeochemical methods based on metasomatic changes in major element chemistry around massive sulphides (for example, magnesium enrichment at High Lake; Allan et al., 1972). If rock samples are taken to appraise the ore potential of a belt, they should be analyzed for elements other than sulphur in order to distinguish volcanic rocks containing only barren iron sulphide mineralization from those which are associated with copper, zinc, lead, and silver and other useful metals.

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