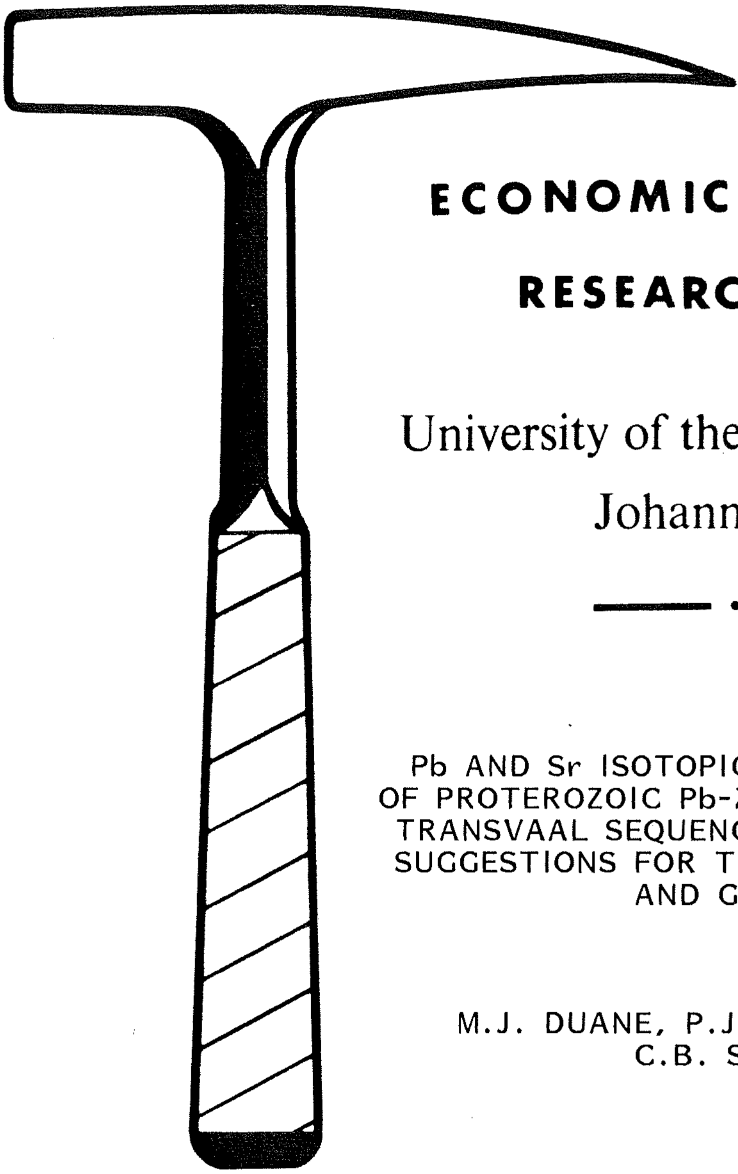

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Pb AND Sr ISOTOPIC CHARACTERISTICS OF PROTEROZOIC Pb-Zn AND Au DEPOSITS, TRANSVAAL SEQUENCE, SOUTH AFRICA : SUGGESTIONS FOR THEIR SOURCE AREAS AND GENESIS

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by

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ABSTRACT

Pb-Zn and Au deposits in the Transvaal and Griqualand West Sub-Basins are potential important contributors to the South African mineral industry. The formation of epigenetic, low-temperature (less than 250°C), carbonate-hosted Mississippi Valley-type (MVT) Pb-Zn deposits similar to those in the Transvaal Basin is generally attributed to dewatering of a sedimentary basin during the latter stages of its evolution. Au deposits in similar environments may be considered to have formed by similar processes, but at more elevated temperatures.

The source of the metals for carbonate-hosted deposits in general is still contentious. In the case of Phanerozoic MVT deposits Pb could have been stripped from sedimentary rocks and/or from basement, but the source of the radiogenic Sr in these deposits has not been unequivocally identified.

The Pb-isotope results for Transvaal galenas indicate district-wide variation for the different Pb-Zn deposits, but little within-deposit variation. Pb in the Pb-Zn deposits becomes more radiogenic to the north and east in the Transvaal Sub-Basin, and towards the south in the Griqualand West Sub-Basin. Variations in initial $^{87}Sr/^{86}Sr$ in gangue minerals associated with the ores from both types of mineralization (Pb-Zn and Au) also exhibit district-wide variation and basement-derived Sr-isotopic signatures. The basement sources most likely to have contributed to the Sr-isotope budget have Rb/Sr ratios that range from 0.20 - 0.53. The Sr-isotopic compositions of the gangue minerals varies antipathetically with Pb-isotopic compositions; mixing of at least two isotopically unique fluids is suggested to account for formation of the deposits. The effects of regional fluid flow and metamorphic overprinting in post-Ventersdorp - pre-Bushveld times is recognised throughout Southern African mafic and ultramafic rocks and the interval is considered to be an important time for formation of Pb-Zn and Au deposits within sedimentary rocks of the Transvaal Basin.

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1. INTRODUCTION

In contrast to the substantial amount of lead isotope data available for constraining the origin of classical MVT ore deposits, the isotopic composition of Sr in the gangue minerals accompanying the sulphides has received less attention. These minerals cover a wide span of the paragenetic sequence of the ore minerals and are well suited for the study of the temporal variation in the chemical composition of the ore fluids (Grant and Bliss, 1983). Gangue minerals, such as calcite, dolomite, fluorite, barite, and celestite, have very low Rb/Sr ratios. Consequently, their $^{87}Sr/^{86}Sr$ ratios are a measure of the isotopic composition of the Sr in the mineralizing fluids at the time of ore deposition.

MVT Pb-Zn deposits are characterized by their heterogeneous and radiogenic Pb-isotopic compositions even within a single crystal (Hart *et al.*, 1983). Although some studies have concluded that the sulphide Pb in MVT ore deposits was derived from the underlying crystalline basement (Kanasewich, 1962; Heyl *et al.*, 1966), these interpretations are not unique. Other isotopic and geologic studies have concluded that Pb was derived from the sedimentary column (Doe and Delevaux, 1972.; Doe, 1979.; Clay, 1986). Application of other isotope systems (e.g. S) and fluid inclusion studies to the problem do not allow an unequivocal choice to be made at this stage between the models (Sverjensky, 1986). High geothermal gradients within the Transvaal Basin during the early emplacement stages of the Bushveld Complex (and Molopo Farms Complex) could have driven ore-bearing, saline fluids out of the centre of the basin and focussed them in the faulted margins of the basin. The process of large-scale dewatering of Transvaal sedimentary rocks (and underlying basins) may be only one manifestation of a regional overprint caused by thermal-tectonic events recorded in mafic and ultramafic rocks of southern Africa. The dewatering model presented here is a modification of that of Jackson and Beales (1967) and Garven and Freeze (1984a, b).

The authors' attempt to use Sr-isotopic compositions (from both shales and gangue minerals), in combination with Pb-isotope ratios from galenas, to identify the reservoirs from which the ore fluids may have derived their Pb and Sr. Sr-isotope data on carbonate gangue minerals in ore deposits of the MVT exist for the Phanerozoic, but limited data exist for similar Proterozoic sulphide deposits. For example, the average $^{87}Sr/^{86}Sr$ for calcite, fluorite and fluorapatite from the Broken Hill deposit (Australia) is 0.716 ($\pm$ 0.003; Shaw, 1968), but the data are not used in a constructive way to propose a genetic model for the deposit.

The following Pb-Zn deposits were sampled: Hendrina, Buffelshoek, Doornhoek, Rhenosterhoek, Bokkraal, Koster, Carletonville, Leeuwbosch, Silverhills, Pering, Bushy Park and Kalkfontein. Four Au deposits were sampled: Olifantsgeraampte, Grootfontein, Rietvallei and Little Joker (Fig. 1). The relative stratigraphic positions of these deposits are shown in Figure 2.

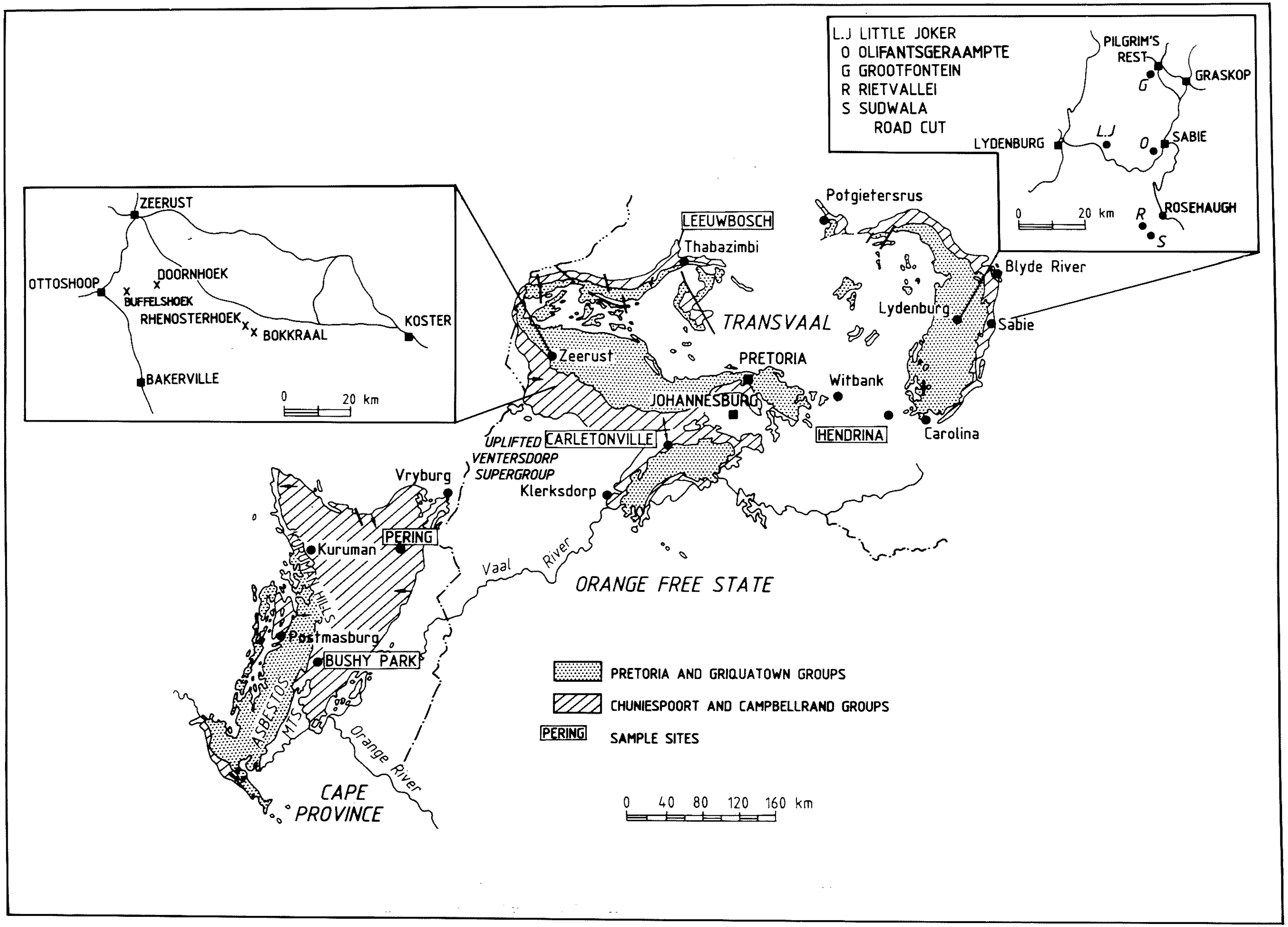

Figure 1: Locality map showing the distribution of Transvaal Sequence and sample sites. (Adapted from Truswell, 1977).

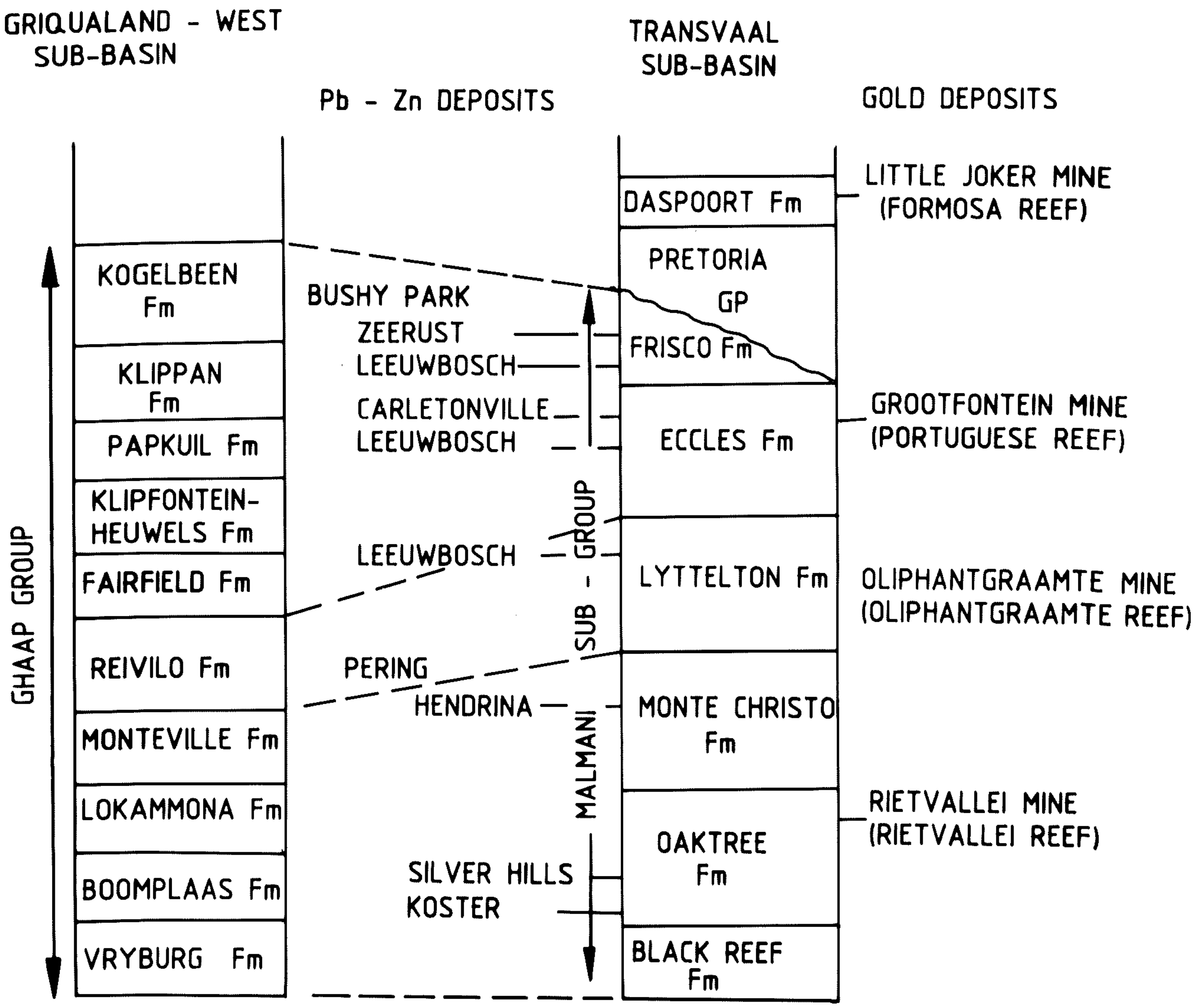

Figure 2: Schematic stratigraphic column showing the location of lead-zinc and gold deposits within the Transvaal sequence. Stratigraphy of the Griqualand West sub-basin after Beukes (1978).

2. GEOLOGIC SETTING

The Transvaal Basin is currently preserved as two structurally defined sub-basins i.e. the Transvaal Sub-Basin and the Griqualand West Sub-Basin (Fig.1). The two sub-basins are presently separated by an uplifted arch-like structure which exposes the Ventersdorp Supergroup (Fig.1). Carbonate lithologies are correlatable across this arch (Clendenin *et al.*, 1988) and the original extent of the whole basin was in excess of 500,000 km^2 (Tankard *et al.*, 1982). Stratigraphically, the rocks of the Transvaal Sequence (circa 2.2 Ga) can be broadly divided into the Pretoria and Chuniespoort Groups and the Black Reef Formation. Correlatable lithologies in the Griqualand West Sub-Basin are the Griquatown and Campellrand Groups and the Vryburg Formation, respectively (SACS, 1980). The Pb-Zn mineralization under discussion in this paper is hosted in the dolomites of the Chuniespoort/Campellrand Group (Fig.1). Major styles of Pb-Zn mineralization in the dolomites of the Transvaal Sequence are:

(1) breccia bodies/pipes formed as a result of karsting and/or carbonate dissolution associated with sulphide precipitation,
(2) unconformities or faults which acted as conduits for the mineralizing fluids, and
(3) replacement mineralization which may be important at some localities (e.g. Carletonville).

The only presently operating Zn-Pb mine in the Transvaal dolomites is Pering Mine, southwest of Vryburg in the northern Cape Province (Fig.1). Here mineralization is hosted around the perimeter of a large collapsed karst structure and also occurs in sub-horizontal, stratabound layers (Wheatley *et al.*, 1986). Breccias, formed as a result of carbonate dissolution associated with sulphide precipitation, are also important host structures. Other Pb-Zn occurrences in the dolomites of the Transvaal Sequence are small and have been mostly worked out; however, they are similar in many respects to the Pering deposit.

The gold deposits of the eastern Transvaal have received renewed interest over the last few years. Gold occurs throughout the stratigraphic section from the Black Reef Formation to the Pretoria Group shales (Fig. 2). The most common form of mineralization is in the form of lensoid reefs which are broadly conformable to bedding, but their economic value is localized to carbonaceous sections in the reefs. Vertical cross-cutting reefs also host gold mineralization, but this study is restricted to the flat reefs. Gold mineralization is commonly associated with pyrite in quartz and shale. A wide variety of other sulphides and sulphosalts also occur, and may host minor gold mineralization (Zeitsman, 1967; Meyer *et al.*, 1986). Up to three periods of gold mineralization and numerous gangue phases have been recognized in other areas (Maske *et al.*, 1986.) and the bulk of the gold is associated with the last major fluid-mobilization event. Similarly numerous periods of mineralization can be identified at the Frankfort Mine (Meyer *et al.*, 1986). The regional geology, structural characteristics and reef horizons have been described in detail by Visser and Verwoerd (1960) and Zietsman (1967). From the available data it is envisaged that the horizontal fault zones acted as conduits for the mineralizing fluids and that the shales within the sequence supplied a suitable reducing environment for sulphide precipitation.

3. SAMPLES AND ANALYTICAL TECHNIQUE

Eighteen samples of galena were selected from veins, shear zones and other sites of mineralization in the dolomites. The galenas were dissolved in 6N HCl acid for 1 hour and the Pb was purified by electrodeposition. Thirteen samples of vug-filling dolomite and siderite, and a single sample of gypsum were selected from Au and Pb-Zn occurrences for Sr analyses. The relative position of each of the vug-filling gangue minerals in the paragenetic sequence was carefully noted. Each sample was thoroughly washed in distilled H_2O to remove any fine powdered carbonate rock fragments that could have adhered to the surfaces. Pure grains were powdered in an agate pestle and mortar and dissolved in open beaker with 6N HCl. $HClO_4$ was needed to dissolve the siderites. Rb and Sr concentrations were determined by isotope dilution mass spectrometry. Isotopic compositions and concentrations were obtained on a MM30 mass spectrometer. Standard SRM 987 runs averaged 0.71025 (0.44% error) during the course of this study; blank measurements for Rb and Sr concentrations were 1 ng and 5 ng, respectively.

4. RESULTS

The Sr and Rb concentrations of the vug-filling, white, fine-grained, rhombic dolomite and siderites from various Au and Pb-Zn deposits are characteristically low and yield high $^{87}Sr/^{86}Sr$ ratios (Table 1) by comparison with the country rocks; published data for the Transvaal dolomites range from 0.7042 - 0.7094 (Faure and Powell, 1972). The initial ratios of the gangue minerals range from 0.7098 - 0.7360 (Table 1). The Leeuwbosch occurrence has several generations of siderite and one generation of late dolomite. One sample of siderite and the vug-filling dolomite were investigated for their Rb and Sr compositions from this deposit. In general, with the exception of the Sudwala road-cut calcite and Leeuwbosch dolomite, Rb concentrations are less than 0.25ppm with relatively high Sr concentrations (Table 1). Thus the $^{87}Sr/^{86}Sr$ ratios are considered to be close to the initial ratio of the gangue minerals, but were back calculated to 2.05 Ga, the age of the Bushveld Complex (a time considered to be important for regional thermal metamorphism in the Transvaal Basin and hence fluid migration; Armstrong, 1987).

Pb isotopic compositions for galenas (Table 2) from the Transvaal Basin show regional and district-wide variations on a uranogenic and thorogenic Pb plot (Fig. 3a and 3b). Amongst the least radiogenic end-members are the galenas from Pering, while the most radiogenic and thorogenic are the galenas from the northern and eastern Transvaal. The model ages young to the north and east in the Transvaal and to the south in the northern Cape Province.

5. DISCUSSION AND CONCLUSIONS

The salinities, stable isotopes (Pinckney and Rye, 1972), and temperatures of MVT ore fluids are commonly considered to be similar to oilfield brines (Hannah and Stein, 1984.; Heyl *et al.*, 1974; Carpenter *et al.*, 1974). Furthermore, high $^{87}Sr/^{86}Sr$ ratios for oilfield brines have recently been reported (Chaudhuri *et al.*, 1987) and are very similar to gangue-carbonate minerals in Phanerozoic MVT ore deposits (Kessen *et*

TABLE 1

Sr–Isotope Data for Gangue Minerals from Pb–Zn and Au Occurrences

Sample	Locality	Sr(ppm)	Rb(ppm)	$^{87}Sr/^{86}Sr$	Ro
Calcite	Hendrina	53	0.03	0.71536 (+4)	0.71531
Siderite	Buffelshoek	49	0.16	0.72804 (+4)	0.72776
Siderite	Rhenosterhoek	58	0.08	0.72643 (+7)	0.72631
Dolomite	Pering	45	0.07	0.73061 (+7)	0.73047
Siderite	Leeuwbosch	2	0.05	0.73815 (+40)	0.73600
Dolomite	Leeuwbosch	20	10.70	0.72399 (+40)	0.67819
Eastern Transvaal Gold Deposits					
Gypsum	Grootfontein	No Data		0.71843 (+3)	No Data
Siderite	Sabie	208	0.17	0.70987 (+4)	0.70980
Magnesite	Sabie	66	0.08	0.71304 (+4)	0.71293
Calcite	Sabie	289	0.16	0.71353 (+2)	0.71348
Dolomite	Rietvallei	823	0.21	0.72178 (+3)	0.72184
Siderite	Little Joker	6	0.03	0.72090 (+7)	0.72048
Calcite	Sudwala	234	19.40	0.70811 (+13)	0.70129

The dolomite from Leeuwbosch may have been introduced after Bushveld times since the initial ratio is meaningles for Bushveld ages.
Samples from Sabie are from Olifantsgeraampte Mine.

al., 1983; Chaudhuri *et al.*, 1983). Ravenhurst *et al.*, (1987) recently reported data for evaporites from the Gays River deposit (Maritime Canada), and showed that anhydrite in the sequence has ratios significantly higher (0.7090) than the host dolomite (0.7083), and that estimated for Phanerozoic seawater (Veiser and Compston, 1976; Veiser *et al.*, 1983). The results of the study are considered by Ravenhurst *et al.*, (1987) to suggest that the ore fluids did not equilibrate isotopically with the host rocks and were derived from evolved crustal sources, such as granite detritus or basement. In the Transvaal Sub-Basin, shales of the Pretoria Group above the dolomites have $^{87}Sr/^{86}Sr$ ratios that range from 0.708 - 0.725 at 2.05 Ga (data from Hunter and Hamilton, 1978). This supports the view that the shales may have been an important source for the radiogenic Sr.

A plot of the possible source compositions and initial ratios for the gangue minerals is shown in Figure 4. The plot illustrates the possible Rb/Sr evolution curves for basement rocks from Swaziland and the southeast of the Transvaal Basin (Davies and Allsopp, 1976). If the deposits formed between 2.2 - 2.0 Ga the initial ratios can be used to constrain the provenance of the Sr in the samples (Davies and Allsopp, 1976).

TABLE 2

Galena Pb Isotope Data for Pb-Zn and Au Occurrences

Sample	Locality	$^{206}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$
Western Transvaal				
PR1	Zeerust	14.809 (2)	15.029 (2)	34.100 (5)
PR2	Zeerust	14.831 (2)	15.027 (3)	34.040 (6)
PR8a	Zeerust	14.778 (2)	15.048 (2)	34.156 (6)
PR15	Zeerust	14.780 (1)	15.072 (1)	34.316 (4)
BK1	Zeerust	14.770 (1)	14.978 (1)	33.955 (3)
BK2	Zeerust	14.843 (2)	15.048 (2)	34.124 (5)
Kafferskraal	Zeerust	14.53 (4)	15.17 (6)	34.55 (14)'
Witkop	Zeerust	15.00 (12)	15.17 (11)	34.48 (24)'
Strydfontein	Zeerust	14.72 (12)	15.38 (12)	34.97 (28)'
Bokkraal	Zeerust	15.03 (9)	15.28 (8)	34.72 (17)'
Doornhoek	Zeerust	15.19	15.50	35.28'
Central Transvaal				
Broederstroom	Pretoria	15.11 (20)	15.33 (15)	34.84 (35)'
Knoppieslaagte	Pretoria	14.66 (3)	15.10 (3)	34.25 (7)'
Boschkop	Brits	16.20 (10)	15.80 (11)	36.23 (25)'
Leeuwenkloof	Pretoria	14.92	15.06	34.21'
PR16	Koster	14.303 (6)	14.835 (7)	33.910 (16)
PR5A	Carletonville	14.818 (1)	15.050 (2)	34.043 (4)
PR14	Carletonville	14.884 (3)	15.161 (3)	34.367 (7)
MY 546	Carletonville	14.961 (6)	15.083 (7)	34.145 (13) *
MY 547	Carletonville	14.778 (10)	15.016 (10)	33.963 (27) *
PR9	Silverhills	14.411 (3)	14.858 (3)	33.801 (7)
73a	GGMA	24.7 (2)	18.5 (2)	35.0 (5)°
73b	GGMA	24.2 (3)	18.4 (2)	34.7 (4)°
Northern Transvaal				
Vogelstruisdraai	N. Marico	15.14 (5)	15.42 (5)	35.21 (11)'
Genadendal	N. Marico	16.14 (13)	15.96 (11)	36.76 (26)'
Spitzkop	Thabazimbi	16.93 (12)	15.73 (16)	36.40 (15)'
PR3	Leeuwbosch	15.096 (2)	15.237 (2)	34.783 (5)
PR7	Leeuwbosch	15.144 (1)	15.295 (2)	34.943 (4)
Eastern Transvaal				
PR4	Hendrina	17.236 (9)	15.629 (8)	36.791 (2)
Northern Cape (Griqualand West Sub-Basin)				
PR6A	Pering	13.530 (1)	14.582 (1)	33.301 (3)
PR12	Pering	13.430 (3)	14.537 (3)	33.236 (8)
PR13	Pering	13.534 (1)	14.675 (1)	33.627 (3)
PR10	Bushy Park	13.815 (2)	14.667 (3)	33.464 (6)
Banghoek	Hay Division	14.18 (9)	14.78 (7)	33.43 (17)'
Geelbek Dam	E. Marydale	13.59 (18)	14.81 (19)	34.01 (44)'
Kalkfontein	Griquatown	14.03 (7)	14.88 (17)	33.90 (7)'
Langrug	Griquatown	14.20 (6)	14.88 (6)	33.90 (14)'
Balloch	Niekerkshoop	14.15 (14)	14.96 (12)	34.13 (27)'
Bushy Park	Griquatown	14.30 (9)	14.98 (9)	34.13 (20)'

PR and BK denotes data collected by authors of this paper
* denotes data from Clay (1986).
° denotes data from Burger *et al.*, (1962); samples 73a and 73b from Black Reef
' denotes data from Nicolaysen *et al.*, (1958).

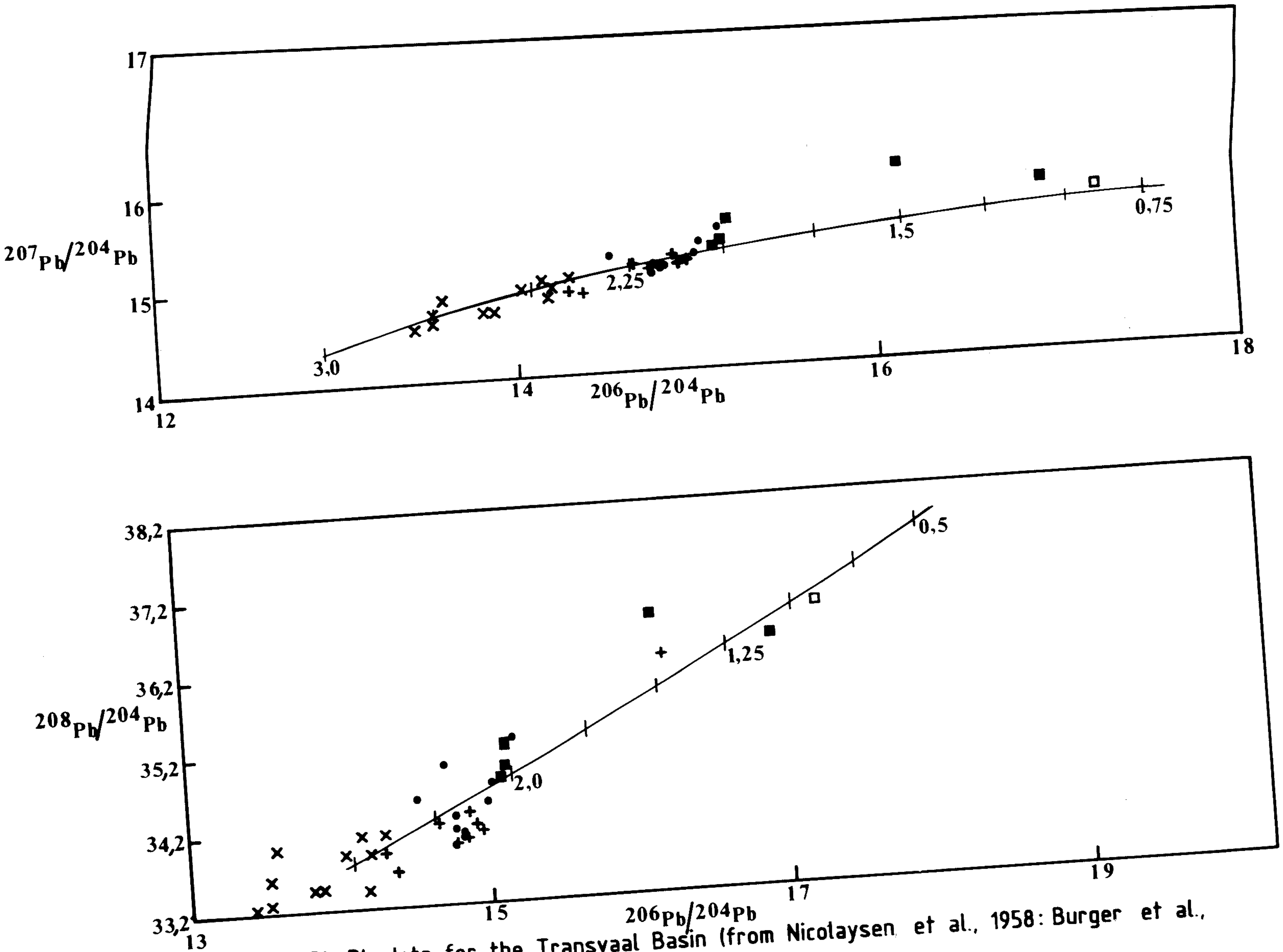

Fig. 3. Combined Pb-Pb data for the Transvaal Basin (from Nicolaysen et al., 1958: Burger et al., 1962: Clay, 1986: and this paper,)

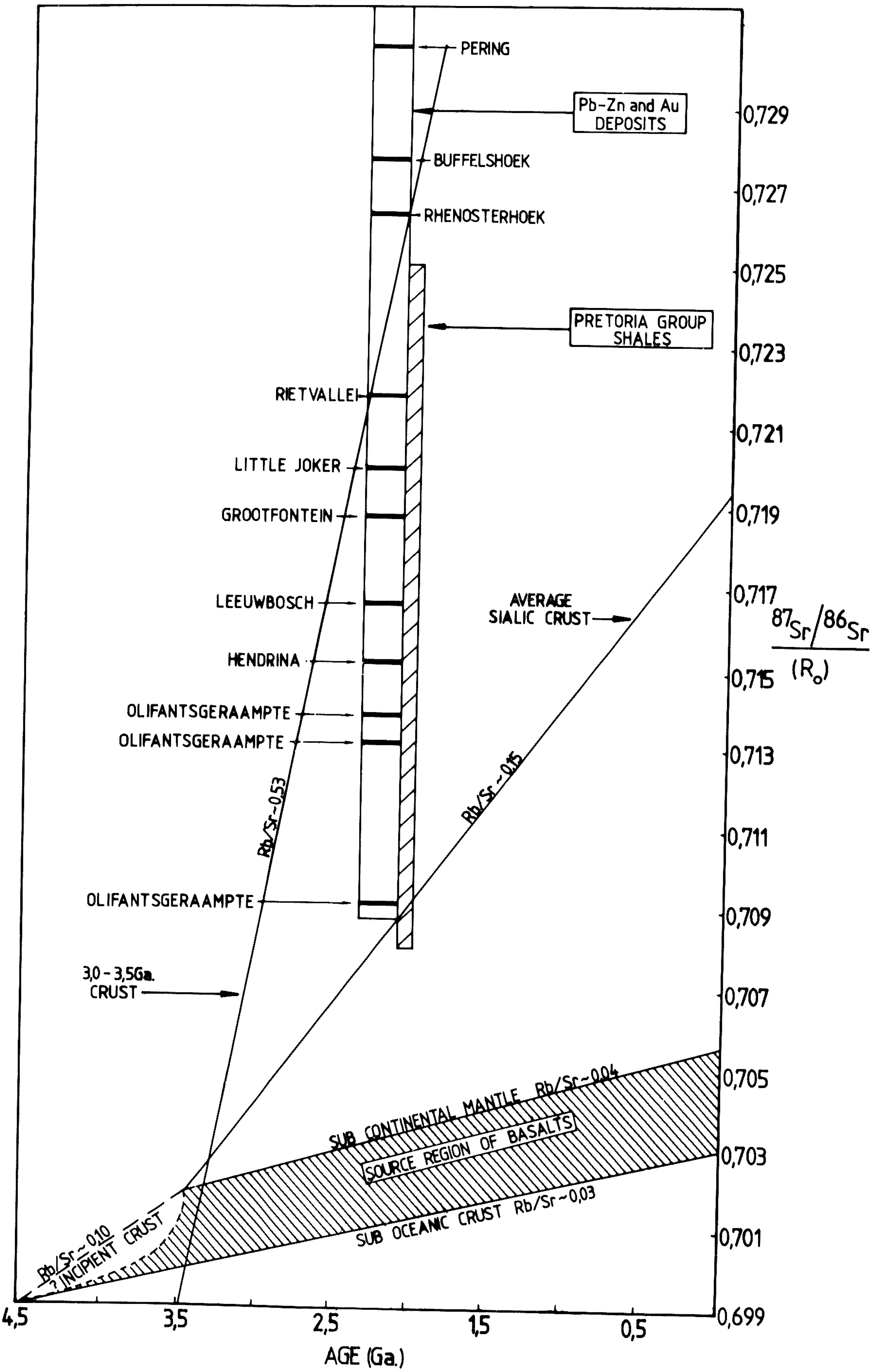

Figure 4: *Initial $^{87}Sr/^{86}Sr$ (R_o) versus age (Ga) plot for possible source compositions for Sr in gangue minerals from Pb-Zn and Au occurrences within the Transvaal Basin. Note the regional variations which are in contrast to the Pb-Pb isotope variations for galenas.*

Preliminary T_{DM} and T_{CHUR} Nd model ages on shales from the Chuniespoort Group within the central Transvaal Basin (Koster) show that 3.0 Ga material was being shed into the basin during deposition of the shales. Possible sources may have been the granite-greenstone terranes which surround the basin.

The gangue minerals record more radiogenic $^{87}Sr/^{86}Sr$ ratios than MVT gangue minerals, even with the time factor taken into consideration. The 3.0 - 3.5 Ga metamorphic crust (with Rb/Sr = 0.53) and the younger igneous basement granites (Rb/Sr = 0.15; Davies and Allsopp, 1976) that form the basement in the eastern Transvaal, may have contributed radiogenic Sr to the shales in the Transvaal Basin. Like classical MVT districts in which the ore fluids have the ability to mix and penetrate the crust during orogeny (Duane and De Wit, 1988, in press) the ore fluids within the Transvaal Basin appear to have undergone regional migration via unconformities and faults to shallower crustal levels.

Available ore genesis models for classical MVT deposits propose that the ore fluids had access to Sr and Pb reservoirs with higher time integrated Rb/Sr and U/Pb ratios as the mineralization continued. The Transvaal galenas have Pb-isotope compositions that define a quasi-linear array and could be interpreted as mixing of fluids accessing Pb reservoirs with higher time integrated U/Pb to the north and east. Possible source rocks for the least radiogenic end-members could be the lavas of the Ventersdorp Supergroup (Armstrong, 1987), since back-calculated (to 2.2. Ga) ratios for these rocks plot in the field between the Transvaal and northern Cape Province galenas. In a similar study of galenas from carbonate-hosted deposits of Western Newfoundland, Swinden *et al.*, (1988) noted that lead in the ores defines linear trends in which the lead-isotope ratios increase with stratigraphic height in the Cambro-Ordovician sequence. These authors attribute the stratigraphic dependency of the Pb-isotope compositions to sources within the shales. The present writers note similar characteristics for the Transvaal galenas and suggest that the unique Pb-isotope compositions present in each deposit may have been derived by similar processes. The Pb-Pb isotope signature systematically changes with stratigraphic height and to the northeast in the Transvaal Sub-Basin implying that progressively more radiogenic Pb is available at source (i.e. in the shales).

In the northern Cape migrating fluids may have traversed the lavas of the Ventersdorp Supergroup during orogeny, which may account for the non-radiogenic Pb-isotope signature. Pb-isotope compositions for galenas from the Transvaal Sub-Basin imply a source with a higher $^{238}U/^{204}Pb$, but the Sr-isotope compositions for the gangue minerals are less radiogenic than the northern Cape Province minerals. The paragenetic data for the ores indicate that the sulphides were emplaced prior to the gangue minerals in most cases and it may be that the fluids arrived at the site of deposition via different paths. This observation is consistent with the fluid-mixing models for classical MVT Pb-Zn mineralization (Sverjensky, 1986).

Pb-isotope compositions for the Witwatersrand Supergroup sulphides display a wide range on conventional histogram plots (Fig. 5) by comparison with data for the Ventersdorp Supergroup and the Transvaal Sequence rocks. The non-radiogenic Pb-isotope compositions for all three systems appear to form a distinct group and imply that migrating fluids carried Pb from the same source into the Witwatersrand and Ventersdorp Supergroups and the Transvaal Sequence at approximately the same time.

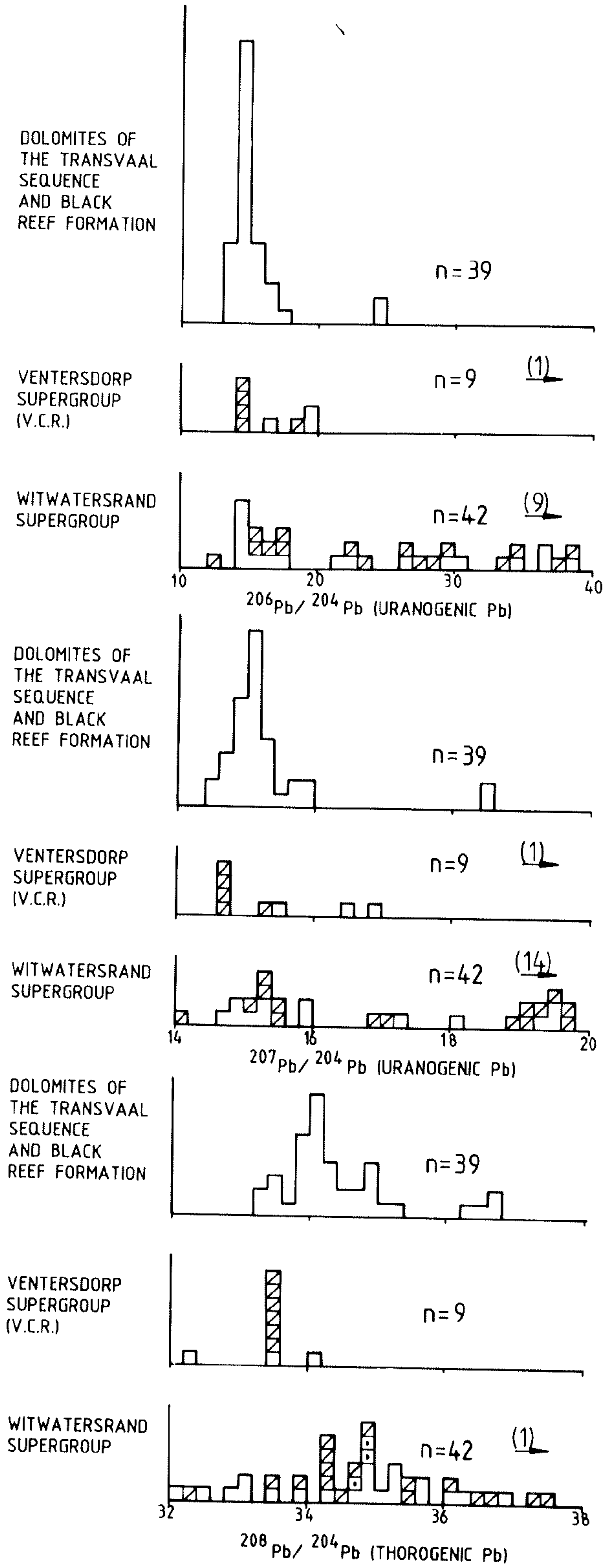

Fig. 5. COMPOSITE HISTOGRAM OF SELECTED Pb ISOTOPE DATA

□ Pb GALENA ▨ Pb PYRITE ⊡ Pb SPHALERITE

DATA FROM NICOLAYSEN ET AL. (1958), BURGER ET AL. (1962), KöPPEL & SAAGER (1974), GIUSTI ET AL. (1986), CLAY (1986) & THIS PAPER

NUMBERS IN PARENTHESIS INDICATES SAMPLES OFF SCALE

The exact timing of igneous events preceding the emplacement of the Bushveld Complex are unknown but Cawthorn *et al.*, (1981) presented an estimate of the parental magma composition and geological age constraints. The first thermal imprint of the Bushveld Complex on the shales of the Transvaal Basin could conceivably have caused fluid migration and mineralization within the dolomites before the main Bushveld igneous event at 2.05 Ga. On the other hand, orogeny-driven fluid movements could be responsible for the location of the ore occurrences at higher stratigraphic levels. Armstrong (1987) presented U/Pb and Rb/Sr isotope data for a number of basic and ultrabasic rocks from southern Africa which suggests that regional overprinting has occurred in post-Ventersdorp times. An apparent enrichment in U/Pb and R_0 for these rocks (Armstrong, 1987), combined with dramatic increases in $^{238}U/^{204}Pb$ immediately after the Ventersdorp volcanism, suggests that a fundamental crustal process has occurred. Burke *et al.*, (1985) have shown that the Ventersdorp volcanism coincided with, and was probably a consequence of, deep crustal rifting resulting from collision of the Kaapvaal and Rhodesian cratons at circa 2.6 Ga. Duane and De Wit (1988; in press) pointed out that continental collision can cause regional fluid-flow patterns over the entire duration of an orogeny. These authors noted dramatic increases in $^{238}U/^{204}Pb$ ratios in ores within foreland sediments away from the collisional axis of the Caledonian orogen which they interpreted as fluids accessing basement (or detritus thereof) with higher time integrated U/Pb ratios as the mineralization progressed. Continental collision may be an important aspect of metallogenesis in the Transvaal Basin, but further research is needed before positive causes, be it the intrusion of the Bushveld Complex or collisional tectonics, can be assigned as major causative factors in the genesis of these Pb-Zn and Au ore deposits.

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