

OXYGEN AND HYDROGEN ISOTOPE EVIDENCE
FOR LARGE-SCALE CIRCULATION AND
INTERACTION BETWEEN GROUND WATERS
AND IGNEOUS INTRUSIONS, WITH PARTICULAR
REFERENCE TO THE SAN JUAN VOLCANIC
FIELD, COLORADO

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ABSTRACT

The 33 to 25 m.y.-old intrusive and volcanic rocks from western San Juan Mountains (Silverton-Ouray area of Colorado) are all abnormally low in O¹⁸ relative to most igneous rocks, particularly along the eastern edge of the Silverton Caldera where the average $\delta O^{18} = -5$. The only exceptions to this are certain plutons emplaced into Paleozoic and Mesozoic sedimentary rocks. Therefore, although in the volcanic rocks very large-scale convective circulation systems involving heated meteoric ground waters were established by the epizonal igneous intrusions, this did not occur in the less permeable sedimentary sections. The very low δD values of the hydrous minerals in the igneous rocks confirm these conclusions ($\delta D = -137$ to -150). The same processes also occurred in the vicinity of the 29 m.y.-old Alamosa stock in the eastern San Juan Mountains, but the δO^{18} depletion effects are not so extreme and the δD values are much higher (-101 to -118). This seems to imply that in the mid-Tertiary, the meteoric waters were about 35 per mil different in these two areas, even though they are only 70 miles apart. The meteoric-hydrothermal alteration processes that have affected these types of rocks can in certain circumstances produce almost complete oxygen exchange in plagioclase phenocrysts while preserving igneous textural features and delicate oscillatory zoning in the plagioclase.

With two exceptions all of the low-O¹⁸ igneous rocks throughout the world are of late Mesozoic to Tertiary age; these are now known to be very extensive and quite common in all volcanic fields where there are shallow igneous intrusions. In Precambrian rocks, however, the reverse is true. Alkali feldspars from the red-rock granophyres and granites in the Muskox, Bushveld, and Duluth Complexes, and in the St. Francois and Keweenaw volcanic terranes are commonly higher in O¹⁸ than coexisting quartz. Although this could have resulted from much higher-O¹⁸ meteoric waters in the Precambrian (which would imply that the oceans also were much higher in O¹⁸), it seems more likely that this is due to very low-temperature ($\sim 150^{\circ}\text{C}$) exchange with ground-water brines that circulated through these rocks for long periods of time. All these alkali feldspars are turbid and contain disseminated hematite dust; this high oxidation state is readily explained if the alteration occurs at low temperatures. It would also explain why Rb-Sr ages on these rocks are generally younger and much more variable than are the Pb-U zircon ages. It is because zircon is very resistant to exchange, and the turbid feldspars and altered mafic minerals are not.

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INTRODUCTION

In recent years it has become well established by means of oxygen isotope analyses that certain epizonal igneous intrusions have interacted on a very large scale with meteoric ground waters. In favorable terranes, that is, in highly jointed, permeable, flat-lying volcanic rocks, these intrusions act as gigantic "heat engines" that provide the energy necessary to promote a long-lived convective circulation of any mobile H_2O in the country rocks surrounding the igneous body. These systems may represent the "fossil" equivalents of the deep portions of modern geothermal water systems such as those in Steamboat Springs, Nevada, and Yellowstone Park, Wyoming (White, 1968).

The interaction and transport of large amounts of meteoric ground waters through hot igneous rocks produces a depletion of O^{18} in the igneous rocks and a corresponding O^{18} enrichment or " O^{18} shift" in the water. Fortunately, primary, unaltered igneous rocks throughout the world display a relatively narrow range of δO^{18} , only +5.5 to +10.0 (Taylor, 1968). This provides a datum by which we can discern the effects of any processes that drastically affect the δO^{18} value of an igneous rock. It is also fortunate that the only commonly occurring natural processes that are known to produce O^{18} depletions in igneous rocks are (1) interaction of the rocks (or magmas) with meteoric ground waters or ocean waters at high temperatures, or (2) strong decarbonation and loss of CO_2 from carbonate-bearing rocks.

It should be pointed out, however, that certain rare occurrences of low- O^{18} igneous rocks are not readily explained by either of the above processes. An example is the Roberts Victor eclogite pipe discussed by Garlick *et al.* (1971). In any event, as the deuterium contents of meteoric ground waters are commonly lower than ocean water and in many cases are much lower than the primary

magmatic waters in igneous rocks, detailed geologic observations combined with both O^{18}/O^{16} and D/H analyses will usually allow a decision to be made between the possibilities mentioned above.

Low- O^{18} igneous rocks produced by interaction with meteoric ground waters have now been observed in the Skaergaard intrusion, the Stony Mountain ring-dike complex in Colorado, the Scottish Hebrides, in Iceland, in the Western Cascades, the Ag-Au deposits at Tonopah, Goldfield, and the Comstock Lode, Nevada, and in portions of the Boulder batholith and the Southern California batholith (Taylor and Epstein, 1963; Taylor, 1968, 1971, 1973; Taylor and Forester, 1971, 1973; Forester and Taylor, 1972; Sheppard and Taylor, 1974; Muehlenbachs *et al.*, 1972). The purpose of the present paper is to present O^{18}/O^{16} and D/H data on another extensive area that shows these features, namely, the San Juan volcanic field, Colorado, and then to integrate all these data to try to understand the mechanisms that produce low- O^{18} igneous rocks. Finally, comparisons will be made between these low- O^{18} rocks and a somewhat analogous set of rocks of Precambrian age that also seem to have exchanged on a large scale with hydrothermal solutions. However, instead of being depleted in O^{18} by the hydrothermal fluids, these Precambrian rocks have instead become enriched in O^{18} .

SAN JUAN MOUNTAINS, COLORADO

*Western San Juan Mountains
(Silverton Caldera)*

The δO^{18} and δD data obtained on volcanic rocks and epizonal igneous intrusions of the western San Juan Mountains are presented in Fig. 1 and Table 1. Analyses are principally from the area of the Silverton Caldera, but some samples were obtained from the Ouray area and from diorite porphyry intrusions em-

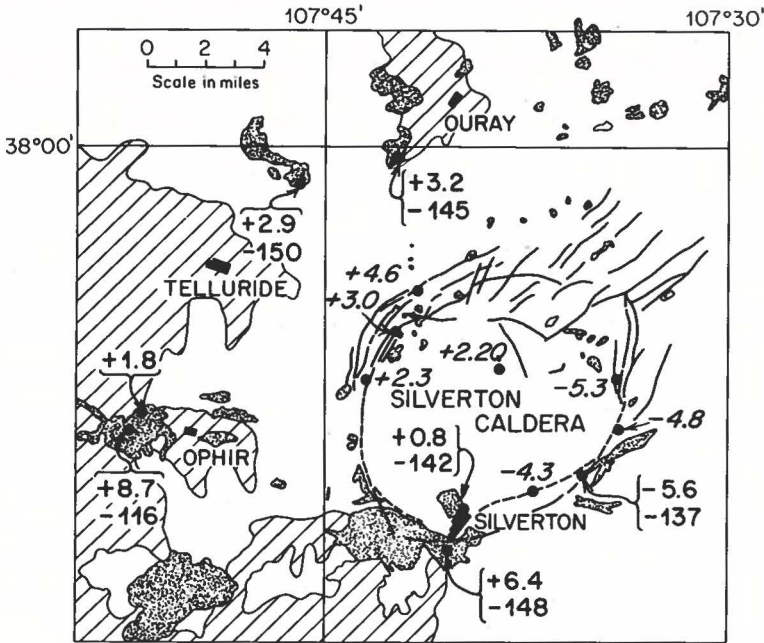


Fig. 1. Generalized geologic map of the Western San Juan Mtns., Colorado (after Luedke and Burbank, 1968), showing some of the oxygen and hydrogen isotope data obtained on igneous rocks in this area (see Table 1 for the complete set of isotopic data). The plotted δO^{18} analyses are on whole rock samples, except that Q = quartz. The analyses on volcanic rocks are given in italics, those on intrusive rocks in regular lettering. The large negative numbers (-142, -150, etc.) represent δD values on biotite or chlorite. The intrusions are shown in stippled pattern, the volcanic country rocks in blank pattern (this includes local outcrops of Precambrian basement), and the Paleozoic and Mesozoic sedimentary rocks in a diagonal-lined pattern.

placed into the Paleozoic and Mesozoic sedimentary rocks that outcrop to the west and to the south of the volcanic terrane. These various igneous rocks were largely emplaced in the late Oligocene to early Miocene about 33 to 25 m.y. ago (Luedke and Burbank, 1968; Lipman *et al.*, 1970).

The isotopic compositions of samples from the Tertiary volcanic terrane of the western San Juans are very depleted in both O^{18} and deuterium relative to "normal" igneous rocks. This is particularly true of samples collected in the deeply eroded Animas River Canyon along the eastern ring fracture of the Silverton Caldera, where four different samples have an average δO^{18} value of -5 per mil. These samples must have become depleted in O^{18} by at least 10 to

12 per mil during exchange with heated meteoric ground waters. These meteoric-hydrothermal solutions were set into convective circulation at a variety of igneous centers by the many epizonal intrusions along the ring fractures of the Silverton Caldera.

A detailed study of one of these intrusive centers, at Stony Mountain, six miles southwest of Ouray, was made by Forster and Taylor (1972). In this study it was shown that, even in a small geographical area, large δO^{18} variations are produced during the interaction of heated meteoric ground waters with a composite stock that has been emplaced by successive multiple intrusions of magma. In particular, although the bulk of the δO^{18} variations in the Stony Mountain complex are clearly due to exchange between

TABLE 1. Oxygen and hydrogen isotope analyses of whole-rock samples and minerals from the San Juan Mountains, Colorado

	Sample Location and Description	Mineral	$\delta O^{18}(\text{‰})$
<i>Western San Juan Mtns.</i>			
Col-1-8	Propylitized diorite porphyry, on Telluride-Rico highway, 2.6 mi N of Trout Lake.	WR	+1.8
Col-2-8	Slightly chloritized quartz diorite porphyry, north side of Telluride-Rico highway, 2 mi W of Ophir, 2.2 mi N of Trout Lake. $\delta D = -116$ (chlorite + biotite).	WR F	+8.7 +8.6
Col-8-8	Diorite porphyry, just N of La Plata, 2.2 mi N of Kroeger campground and 8.4 mi N of junction with U.S. 160.	H(+C) WR	+6.9 +9.6
Col-11-8	Granodiorite porphyry at contact with Hermosa Formation, Silverton-Durango highway, 7 mi S of Col-13-8.	WR	+5.9
Col-13-8	Medium grained augite-biotite quartz monzonite, 1 mi SW of Silverton on Silverton-Durango highway. $\delta D = -148$ (biotite).	WR Q KF B	+6.4 +6.6 +8.8 +3.9
Col-13-8a	Fine grained leucocratic granitic dike cutting Col-13-8.	WR	+5.1
Col-14-8	Porphyritic quartz monzonite, somewhat propylitized, just NE of Silverton at mouth of Cement Creek. $\delta D = -142$ (chlorite).	WR Q KF F C	+0.8 +6.3 -0.2 -1.9 -4.1
Col-16-8	Chloritized granodiorite porphyry, Animas River canyon, mouth of Cunningham Creek. $\delta D = -137$ (chlorite).	WR F C	-5.6 -5.2 -7.6
SJ-25	Altered rhyolite, Burns Formation, Animas River canyon, mouth of Eureka Gulch, 7 mi NE of Silverton.	WR	-5.3
SJ-26	Altered rhyolite, Burns Formation, Animas River canyon, mouth of Maggie Gulch, 6 mi ENE of Silverton.	WR	-4.8
SJ-33	Altered rhyolite, Burns Formation, resistant ridge overlooking Animas River canyon, 2.5 mi NE of Silverton.	WR	-4.3
Col-3R (88316)	Vein quartz from an ore body in the Gold King mine, 6 mi N of Silverton.	Q	+2.2
SJ-14	Altered Treasure Mtn. rhyolite, roadcut on U.S. highway 550, 1.1 mi S of Red Mtn. Pass.	WR	+2.3
SJ-12	Silverton Volcanic Group, roadcut on U.S. highway 550, 0.9 mi N of Red Mtn. Pass.	WR	+3.0
SJ-10	Silverton Volcanic Group, roadcut on U.S. highway 550, 2.5 mi N of Red Mtn. Pass. (San Juan Co. line).	WR	+4.6
Col-17-8	Fresh, unaltered med. gr. pyroxene gabbro from Stony Mountain, 6 mi SW of Ouray (see Forester and Taylor, 1972, and Dings, 1941). $\delta D = -150$ (biotite).	WR F B M	+2.9 +3.3 +4.0 +2.1
Col-18-8	Highly propylitized diorite porphyry in Canyon Creek, 3 mi SW of Ouray. $\delta D = -145$ (chlorite).	WR	+3.2
Col-19-8	Slightly chloritized and unalitized gabbro dike, N-trending, 1.0 mi N of Cedar Hill Cemetery, N of Ouray.	WR	+3.8
<i>Eastern San Juan Mtns.</i>			
Col-27-8	Granodiorite porphyry, just W of and above Annella Lake. Plagioclase shows well-developed, fine-scale oscillatory zoning (see Fig. 5).	Q F KF B Cpx	+6.8 +6.5 +5.8 +1.7 +4.5
Col-28-8	Andesite porphyry, Conejos formation, chloritized biotite phenocrysts, Alamosa River, 0.9 mi W of Lake Annella.	F B(+C)	+6.1 -0.9
Col-29-8	Biotite-augite granodiorite, Alamosa River stock, on road 2.5 mi E of Lake Annella, 1.4 mi W of junction to Stunner Campground.	F B	+4.8 +2.1
Col-32-8	Strongly kaolinized quartz monzonite porphyry, N side of Alamosa River stock, on road 0.7 mi W of junction to Stunner campground. $\delta D = -91$ (kaolinite).	Kaol	+4.2
Col-34-8	Strongly kaolinized quartz monzonite cut by quartz veins (3-4 mm thick), N side of Alamosa River stock, along road just W of bridge over the Alamosa River.	Q Kaol Q(vein)	+5.6 +3.9 +4.7

TABLE 1. *Continued*

Sample Location and Description		Mineral	$\delta O^{18}(\text{‰})$
Col-36-8	Uralitized quartz monzonite with turbid K feldspar, center of Alamosa River stock, on road to Platoro, 0.9 mi S of bridge over the Alamosa River. $\delta D = -101$ (chlorite + biotite).	WR	+2.3
		KF	+2.6
		M	-2.8
Col-36-8a	Same locality as Col-36-8, quartz monzonite and associated fine gr. xenolith of altered andesite.	WR	+1.5
		(qtz. monz.)	
		WR (xenolith)	+1.1
Col-37-8	Hornblende granodiorite with turbid K feldspar, Alamosa River stock, on road to Platoro, 0.3 mi S of bridge over the Alamosa River.	WR	+3.6
Col-39-8B	Augite-biotite granodiorite, turbid K feldspar, oscillatory zoned plagioclase, on road at mouth of Bitter Creek, 1.3 mi E of bridge over the Alamosa River.	F	+3.8
		M	0.0
Col-39-8A	Pink, fine gr. granitic dikelet (1 cm across) cutting Col-39-8B.	WR	+5.0
Col-41-8	Chloritized quartz monzonite with turbid alkali feldspar, along road at E edge of Alamosa River stock. $\delta D = -118$ (chlorite).	WR	+1.3

Abbreviations: WR = whole rock; F = plagioclase feldspar; KF = potassium feldspar; Q = quartz; B = biotite; C = chlorite; H = hornblende; M = magnetite; Cpx = clinopyroxene; Kaol = kaolinite. δO^{18} values are relative to SMOW (standard mean ocean water); in many cases two or more separate determinations were made on each sample, with an analytical error of ± 0.1 per mil. δD values are also relative to SMOW; the analytical error is ± 3 per mil.

heated meteoric ground waters and *solidified* igneous rocks, Forester and Taylor (1972) concluded that a low- O^{18} magma must have been produced by some process of direct or indirect exchange with the meteoric ground waters that circulated to depths of several kilometers in this highly fractured, permeable, volcanic terrane.

We therefore cannot rule out the possibility that some of the low δO^{18} values shown in Fig. 1 are the result of crystallization of low- O^{18} magmas. However, this is clearly not the case for samples such as Col-14-8 and Col-17-8 which display marked isotopic disequilibrium within an assemblage of coexisting minerals. In these two rocks, quartz-feldspar fractionations of +5.5 to +8.2 per mil ("normal" values are +1.0 to +1.5 in igneous rocks) and a plagioclase-biotite fractionation of -0.7 per mil ("normal" values are +3 to +5) are clearly the result of preferential O^{18} depletion of the feldspar relative to coexisting quartz and biotite. Several other such examples of isotopic disequilibrium were cited by Forester and Taylor (1972) and this

phenomenon has by now been well documented in several other localities (Taylor, 1968; Taylor and Forester, 1971, 1973). This implies that both plagioclase and alkali feldspar are much more susceptible to hydrothermal oxygen isotope exchange and/or recrystallization than any other common rock-forming mineral (with the probable exception of calcite).

The low- O^{18} rocks from the western San Juans all exhibit uniformly low δD values of -137 to -150, indicating that the meteoric ground waters in question were also relatively low in deuterium. If we assume that equilibration temperatures were on the order of 300°-600°C, these waters would have had δD values about 20 to 40 per mil higher than the associated chlorite or biotite with which they exchanged (Suzuoki and Epstein, 1973). This implies a δD of about -100 to -130 for these meteoric waters, and in subsequent discussion we shall assume that such waters had a uniform $\delta D \approx -115$. Applying the meteoric water equation ($\delta D = 8\delta O^{18} + 10$, Craig, 1961), this implies an initial $\delta O^{18}_{H_2O} \approx$

—16. Such waters would, of course, have undergone an O^{18} shift upward to δO^{18} values of about -5 to zero after having been involved in high-temperature exchange with most of the rock samples shown in Fig. 1. However, the extreme low- O^{18} rocks that occur along the eastern edge of the Silverton Caldera must have been in exchange equilibrium with H_2O having a δO^{18} value lower than -5 . Depending on the temperature, the δO^{18} of this H_2O may have been as low as -10 to -15 . These parameters require that the integrated water/rock ratios along the eastern ring fractures of the caldera were at least unity and were probably on the order of 5 to 10 (Fig. 10, Taylor, 1971). Note that these are minimum values because of the likelihood that an appreciable amount of water circulated upward along the fractures without completely equilibrating with the surrounding volcanic rocks.

The meteoric-hydrothermal solutions described above were not totally pervasive throughout the area shown in Fig. 1, as shown by the δO^{18} data from Col-13-8, a sample from the center of the large quartz monzonite stock just southwest of Silverton. The whole-rock δO^{18} value of $+6.4$ is only about 2 per mil lower than "normal" for a quartz monzonite (Taylor, 1968), and the quartz-alkali feldspar O^{18} fractionation of 2.2 per mil is just 1 per mil larger than "normal". Thus, only minor quantities of meteoric-hydrothermal solutions penetrated into the center of this stock (the integrated water/rock ratio may have been as low as 0.05 to 0.10). Nonetheless, such amounts of H_2O are more than enough to have overwhelmed the small amounts of primary magmatic water that may have been originally present in the biotite or hornblende of this quartz monzonite. This conclusion is of course also well documented by the very low δD value of the biotite in this rock (-145).

In addition to this example, which is a large stock emplaced along the bound-

ary between the permeable volcanic terrane and a Paleozoic sedimentary rock section, certain other igneous rocks listed in Table 1 and Fig. 1 also show little or no O^{18} depletion. It is notable that all of these represent intrusions into the sedimentary-rock section, which apparently was much less permeable to ground water than was the volcanic section. Of such samples, only a single highly altered specimen collected at an intrusive contact near the boundary between the volcanic rocks and the sedimentary rocks shows any clear-cut O^{18} depletion whatsoever (Col-1-8, $\delta O^{18} = +1.8$, left side of Fig. 1). Other samples (Col-2-8, Col-11-8, and Col-8-8) lie well within the range of δO^{18} values of normal igneous rocks. The abnormally high δD value obtained on the stock west of Ophir (Col-2-8, -116) also indicates that only minor amounts of meteoric ground water probably exchanged with this intrusion during its crystallization and cooling. Although lower than normal, this δD value is the highest yet obtained from igneous rocks in the western San Juan Mountains.

In summary, the following picture emerges during the complex late-Oligocene to early-Miocene intrusive and volcanic history of the western San Juan Mountains. Repeated cauldron subsidence, volcanic eruption, and emplacement of epizonal intrusions provided the heat energy and fracture permeability necessary to establish widespread meteoric-hydrothermal convection systems throughout the volcanic terrane shown in Fig. 1. The hydrothermal fluids circulated to depths of at least several kilometers. Water/rock ratios were at a minimum on the order of 0.5 to 1.0 and were locally higher than 5.0. Circulation in the Paleozoic and Mesozoic sedimentary rocks (and probably the Precambrian basement rocks exposed to the south in the Needle Mountains) was, however, much more restricted, as a re-

sult of lower permeabilities in these rock types.

Very little isotopic data is available on the ore deposits of the western San Juans, but by analogy with data obtained on other epithermal mineral deposits in volcanic terranes (Taylor, 1973), it is practically certain that most of the ore deposits in the San Juan volcanic field were formed from hydrothermal fluids that contained a significant component of meteoric ground water. This statement is supported by the low- O^{18} quartz obtained from an ore body in the Gold King mine near the center of the Silverton Caldera (Fig. 1). The δO^{18} value of this quartz (+2.2) indicates formation from a hydrothermal fluid with $\delta O^{18} \approx -5$ to -10 , if the temperature of deposition was in the range 200° – $400^{\circ}C$ (Clayton *et al.*, 1972). Such a low- O^{18} fluid must be dominantly

(or wholly) of ground water origin. Even more definite statements of this sort can be made concerning a vein dickite from the Ouray area ($\delta O^{18} = -6.2$, $\delta D = -141$, see Sheppard *et al.*, 1969), and various low- O^{18} vein quartz samples near the Stony Mountain stock (Forester and Taylor, 1972).

*Eastern San Juan Mountains
(Alamosa River Stock)*

Isotopic analyses were made on several samples from the composite granodiorite-quartz monzonite Alamosa River stock from the southeast part of the San Juan volcanic field (see Table 1 and Fig. 2). The K-Ar age of this stock is 29.1 m.y. (Lipman and others, 1970). The geology of this area, which lies within the Platoro Caldera, is described by Lipman and Steven (1970).

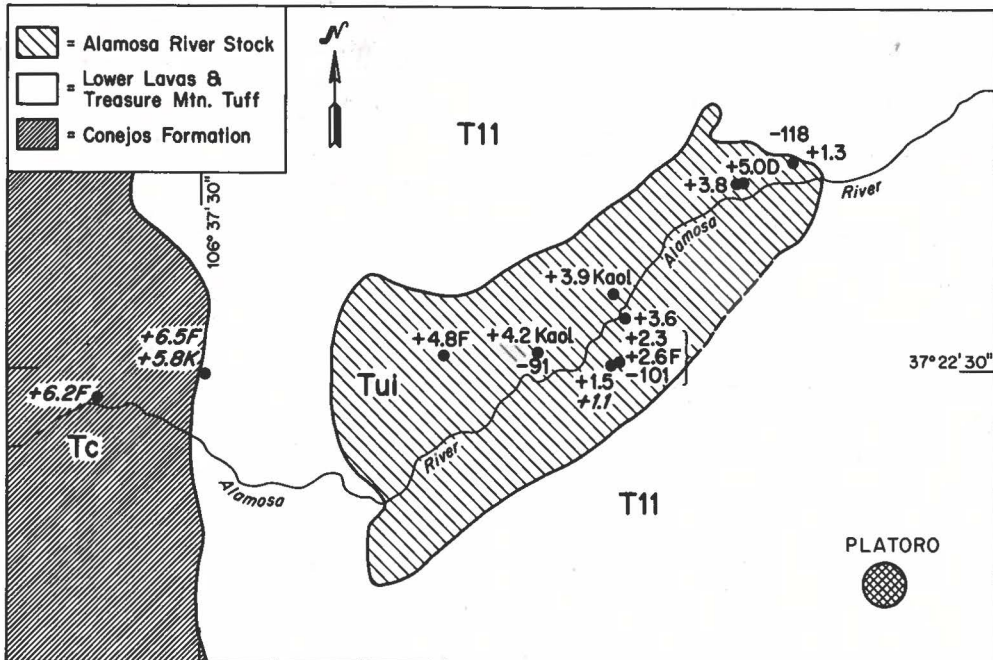


Fig. 2. Generalized geologic map of the Alamosa River stock and vicinity, San Juan Mtns., Colorado (after Lipman and Steven, 1970). The plotted numbers indicate some of the oxygen and hydrogen isotope data obtained on igneous rocks in this area (see Table 1 for a complete list of the isotopic data). The notation is identical to that given in Fig. 1, except that D = granitic dike, F = plagioclase, K = K feldspar, and Kaol = kaolinite.

The δO^{18} values of samples from the Alamosa River stock are abnormally low relative to "normal" igneous rocks (Taylor, 1968), although the O^{18} depletions are not quite so extreme as those described above from the western San Juans. The observed O^{18} depletions must have locally amounted to as much as 5 per mil (e.g., Col-36-8a, $\delta O^{18} = +1.5$). Therefore, the same general type of meteoric-hydrothermal convective system must have been set up after emplacement of this stock, but judging by the observed isotopic effects in the stock and in its country rocks, this system either involved meteoric water with a higher δO^{18} value or the amounts of water involved were less. Also, the convective circulation could have persisted to lower temperatures where the mineral- H_2O O^{18} fractionations are larger. One indication that the latter might have occurred is the very extensive kaolinitic alteration zone present on the north side of the Alamosa River stock. The δO^{18} values of two kaolinite samples from this part of the stock are 3.9 and 4.2. Assuming these formed in the temperature range 100° – $200^\circ C$, they imply a δO^{18} of about -5 to zero in the low-temperature sulfataric-type hydrothermal solutions that formed the kaolinite (Sheppard *et al.*, 1969).

An interesting aspect of the hydrogen isotope data shown in Fig. 2 is that the samples from the eastern San Juans are markedly richer in deuterium than those from the Silverton area only 70 miles to the northwest (~ -110 vs ~ -145 , respectively). It is clear from the δO^{18} data that meteoric-hydrothermal systems were important in both localities, and the time of formation of the calderas and associated epizonal intrusions was late Oligocene to early Miocene in both areas (Lipman and others, 1970). Therefore, the meteoric ground waters in the two areas apparently had markedly different δD values about 30 m.y. ago. This cir-

cumstance conceivably could be explained as follows: (1) The elevation of the volcanic plateau in the Silverton area may have been appreciably higher than in the Platoro area. This could have allowed greater snowfall in the Silverton area and thus a lower δD in the integrated annual local precipitation. This is actually the case today in the two

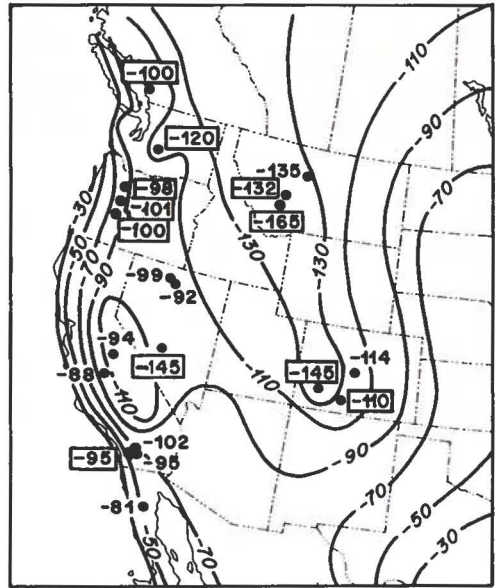


Fig. 3. Map of the western United States showing generalized contours of δD in present-day meteoric surface waters (rivers, lakes, etc.) after Friedman *et al.* (1964). The lettered numbers represent δD analyses (or averages for a given locality) of biotite, chlorite, or hornblende in a variety of Tertiary or late Mesozoic epizonal igneous intrusions (data from the present work; Taylor, 1973; Taylor and Epstein, 1968; Shieh and Taylor, 1969; Sheppard and Taylor, 1974). The numbers enclosed in rectangles are for samples that are depleted in O^{18} and are thus known to have interacted with abundant meteoric ground waters. For example, the $\delta D = -110$ and -145 in southwest Colorado represent the data from Table 1 on the Eastern and Western San Juan Mtns., respectively. Apparently, a number of the other intrusions with "normal" δO^{18} values also have exchanged with small quantities of meteoric ground waters. Note for comparison that most deep-seated igneous rocks from regional metamorphic or batholithic terranes have δD values of about -60 to -80 (Taylor and Epstein, 1966; Turi and Taylor, 1971; Godfrey, 1963).

areas. (2) The topographic barriers in the mid-Tertiary may have been such that the western San Juans obtained most of their rainfall and snowfall from Pacific storms, whereas the southeastern San Juans obtained most of their precipitation from air masses formed over the Gulf of Mexico. This pattern is also generally true today in the two areas, and it leads to higher deuterium values in the surface waters of northern New Mexico (see Friedman and others, 1964, and the map in Fig. 3). (3) The meteoric-hydrothermal systems may in fact have been terminated at different times in the two areas. The exact ages of such hydrothermal alteration zones are not easily determined, and the isotopic precipitation patterns could be expected to be different at different times in the Tertiary.

Again, as is the case in the Silverton district, several areas of ore mineralization are found in the vicinity of the Alamosa River stock, notably the Summitville district to the north and the Platoro district to the southeast. By analogy, these areas of hydrothermal alteration and ore deposition also very likely involve fluids that contain a dominant meteoric-water component. Note that, based on the D/H data in Fig. 2, the δD value of this meteoric water would have been about -80 , implying an initial δO^{18} of about -11 rather than the -16 suggested in the Silverton area. Therefore, this might also in part explain why the δO^{18} values of the Alamosa River stock are higher than in the western San Juans.

HYDROGEN ISOTOPE VARIATIONS IN LOW- O^{18} IGNEOUS ROCKS IN NORTH AMERICA

The scale and the widespread nature of meteoric-hydrothermal convective circulation systems in volcanic-intrusive terranes are shown in Figs. 3 and 4. They are summary diagrams based upon data

from a variety of sources, as well as upon the data given in Tables 1 and 2.

The map in Fig. 3 shows δD values of chlorite, biotite, and hornblende from a variety of igneous rocks throughout western North America. The δD values enclosed in rectangles represent samples (or averages for a locality) where the igneous rocks are abnormally low in O^{18} and are known to have exchanged at high temperatures with meteoric ground waters. The idea here is that if meteoric water effects are at all discernible in the O^{18}/O^{16} data, the D/H values of the hydroxyl-bearing minerals will totally reflect the D/H ratio of the local meteoric water, making some allowance for the variations introduced by differences in temperature and chemical composition of the minerals. In general, the lower the temperature or the higher the Fe/Mg ratio in the mineral, the larger is the water-mineral D/H fractionation (Suzuoki and Epstein, 1973), and the heavier would be the calculated δD of the H_2O .

Note in Fig. 3 that in spite of the above complications, there is a general correspondence between the δD of low- O^{18} igneous rocks and geographical position. Most of the samples shown on Fig. 3 are mid- to early-Tertiary. The samples from the Rocky Mountains-Great Basin area are consistently lower in δD than those from the areas near the Pacific Coast. As shown by the δD contours of present-day surface waters, this is also the general isotopic pattern produced in present-day rainfall and snowfall; this correlation is to be expected in view of the fact that the general shape of the North American continent in the Tertiary is known to have been roughly similar to the present-day configuration. The δD values calculated for waters that would have coexisted in equilibrium with the low- O^{18} igneous rocks from Fig. 3 at high temperatures are in general somewhat heavier than the values of present-day surface waters. This is, however, to

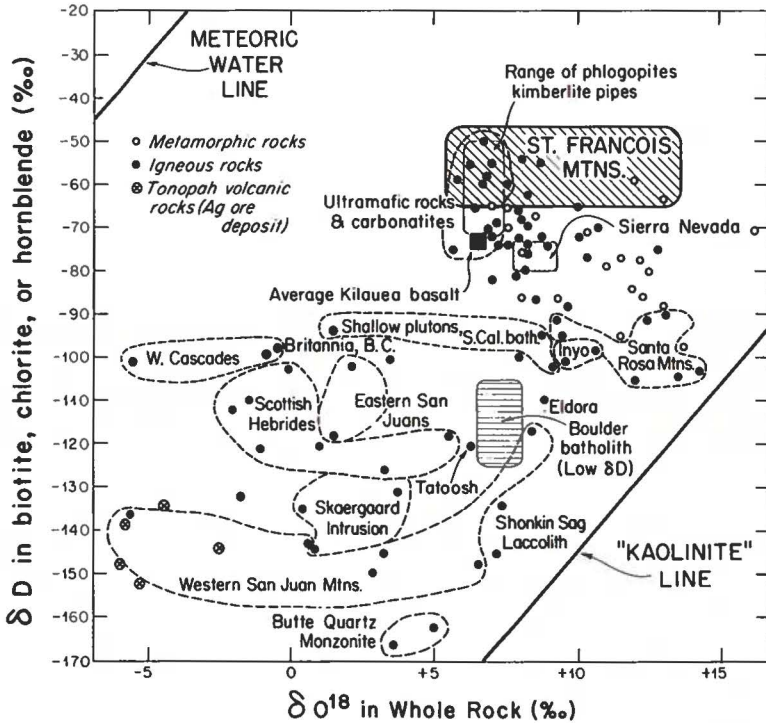


Fig. 4. Plot of δD in biotite, chlorite, or hornblende vs δO^{18} in whole rock for a variety of igneous and metamorphic rocks. The data are from the present work, and from Sheppard and Epstein (1970), Sheppard and Taylor (1974), Taylor (1973), Taylor and Epstein (1966, 1968), Shieh and Taylor (1969), Turi and Taylor (1971), Friedman (1967), and Godfrey (1963, Group II only). The samples labeled Butte quartz monzonite actually represent the early dark micaceous (biotite) alteration in the Butte ore deposit. Also shown is the general range of δO^{18} and δD from Precambrian igneous rocks of the St. Francois Mtns., Missouri (Wenner and Taylor, 1972). The meteoric water line (Craig, 1961) and sedimentary kaolinite line (Savin and Epstein, 1970) are shown for reference. Note that there is a rough correspondence between low δD values and low δO^{18} values but that at a given locality the δD is roughly constant while the δO^{18} may vary considerably.

be expected because the climate is known to have been generally warmer in the mid- to early-Tertiary than it is at present (Axelrod, 1964).

The δD values shown in Fig. 3 that are not enclosed in rectangles are from epizonal to mesozonal plutons which do not show any significant O^{18} depletion. In some instances even these plutons show a D/H correlation similar to that established for the low- O^{18} igneous rocks. This probably indicates that these epizonal igneous intrusions (all of which are emplaced into crystalline rocks or sedimentary rocks, not into permeable volcanic rocks) only exchanged with very

tiny amounts of meteoric H_2O , enough to affect the D/H ratios of the hydroxyl-bearing minerals, because these contain only a small amount of hydrogens, but not enough to appreciably affect the much larger oxygen reservoir present in the whole-rock systems.

THE PROBLEM OF LOW- O^{18} MAGMAS

Reference was made above to the probable emplacement of the inner diorite in the Stony Mountain complex as a low- O^{18} magma with $\delta O^{18} \approx +2.5$ (Forester and Taylor, 1972). Such low- O^{18} silicate liquids also were apparently produced at least in minor amounts during the very

TABLE 2. Oxygen isotope analyses of other igneous rock samples analyzed in the present work

	Location and Sample Description	Mineral	$\delta O^{18}(\text{‰})$
IC.2	Obsidian, fresh, nonhydrated, postglacial, Hrafninnuhryggur, near Myvatn, Iceland, donated by I. Carmichael (see Wright, 1915; Carmichael, 1962).	WR	+3.0
Brit-4	Quartz monzonite porphyry, 4000-ft level of Britannia Mine, British Columbia. $\delta D = -100$ (biotite).	WR	+3.8
		Q	+7.1
		F	+2.8
		KF	+2.8
O-23-9	Granodiorite, fine gr., center of Champion Creek stock, Bohemia Mining District, Oregon (see Fig. 7). All plagioclase grains show well-developed oscillatory zoning. Typical examples are shown in Fig. 5b and c.	B	-1.9
		Q	+6.2
		F (phenocryst)	+2.6
O-28-9	Hydrothermally altered andesite, very fine gr., porphyritic, 1.5 mi W of contact of Nimrod stock, McKenzie River, Western Cascade Range, Oregon (see Fig. 5 of Taylor, 1971). Many of the abundant andesine phenocrysts show moderately well-preserved oscillatory zoning, but recrystallization is much more apparent than in O-23-9. The quartz in this rock is present in aggregates and has clearly been recrystallized at some stage in the hydrothermal alteration process.	WR	+1.6
		Q	+0.3
		F (phenocryst)	-4.1
		WR	-5.0

Abbreviations: Same as in Table 1.

latest stages of differentiation of the Skaergaard intrusion in east Greenland (Taylor, 1968; Taylor and Forester, 1973). However, the first definitive evidence for the existence of large volumes of low- O^{18} magmas was found in a number of recent volcanic rocks from Iceland (see Table 2 and Muehlenbachs *et al.*, 1972; Muehlenbachs, 1973). Many such fresh, unaltered low- O^{18} lava flows in Iceland have now been identified by Muehlenbachs and his co-workers. They range in chemical composition from rhyolite to basalt. However, none of these magmas exhibits an O^{18} depletion anywhere near as large as that of many epizonal intrusions or altered volcanic rocks such as those from the Animas River Canyon shown in Fig. 1. The δO^{18} values of the low- O^{18} magmas are typically about +4 to +5 and none have been found to be lower than +2 per mil.

Thus, although low- O^{18} magmas certainly are generated in volcanic-intrusive terranes, it is clear that most of the O^{18} depletion observed in areas such as the San Juan volcanic field and the other

localities noted in Fig. 4 is due to circulation of heated meteoric ground waters along joints and fractures in solidified igneous rocks. This effect is usually easily discerned by demonstrating a correlation between δO^{18} and grain size (e.g., Forester and Taylor, 1972), or by δO^{18} analyses of quartz-feldspar or feldspar-pyroxene mineral pairs. If the more easily exchanged feldspar is found to be abnormally depleted in O^{18} relative to the quartz or the pyroxene, it can be concluded that the abnormal depletion must have occurred after magmatic crystallization. However, if the δO^{18} values of the relatively resistant minerals such as quartz and augite are also lower than "normal," there will in general be some ambiguity about whether or not these minerals might have crystallized from a low- O^{18} magma. For example, the quartz from Col-14-8 and Col-34-8 (Table 1) might have formed from low- O^{18} silicate melts with $\delta O^{18} \approx +4$ to +5.

It is easy to envisage how convective circulation and migration of large

amounts of meteoric ground water can occur in highly fractured rocks under essentially hydrostatic conditions. This represents the typical situation observed in modern hot spring systems (e.g., see White, 1968). However, it is much more difficult to envisage how significant amounts of water could migrate into a magma from an essentially hydrostatic fissure system lying outside an epizonal magma chamber. The magma must be under a lithostatic pressure that would be a factor of 2.5 to 3 higher than the hydrostatic pressure in the fissure system. The fractures that provide the major conduits for H₂O circulation outside the intrusion obviously cannot be present in the immediate contact zone at the edge of the magma body; otherwise they would be forcibly filled with magma. Therefore, if H₂O is to gain access directly to the magma, it must be by grain-boundary diffusion up a thermal gradient through the hot contact zone and into the magma (see Shaw, this volume).

Once the H₂O is inside the magma chamber, convective circulation of the silicate melt or successive injections of new magma can in principle aid the diffusion process in distributing this H₂O through the interior of the magma body. Nonetheless, this is likely to be a relatively slow process, even though most of these magmas are probably initially undersaturated with respect to H₂O. The well-studied Skaergaard intrusion (Wager and Brown, 1967) is a beautiful example of a magma body that definitely did not become depleted in O¹⁸ throughout 95 to 99% of its crystallization history, even though it is known that a large-scale meteoric-hydrothermal convection system was set up outside the pluton throughout its crystallization (Taylor and Forester, 1973). In principle this should have been a relatively favorable example, because of the very low H₂O content of the initial magma.

The difficulties outlined above are compounded by the fact that very large amounts of H₂O are required to produce any significant δO^{18} lowering of the magma. First of all, any H₂O finally able to diffuse into the magma would probably have already undergone a significant O¹⁸ shift to much higher δO^{18} values than those characteristic of the cool ground waters in the surrounding terrane. For example, in the San Juan volcanic area, it would be remarkable if such H₂O had a δO^{18} much lower than -5 at the time of influx into the silicate melt. To produce even a modest δO^{18} lowering of a magma from a "normal" value of about +7 to +4 with such H₂O would require a water/rock ratio of about 0.3. This is far more H₂O than can be dissolved in a magma at such shallow depths in the Earth's crust, thereby implying that most of this H₂O would have to diffuse or bubble through the magma chamber and then out again!

It is therefore doubtful that any model utilizing only direct influx of low-O¹⁸ meteoric H₂O into a magma chamber can by itself account for more than about a one per mil lowering of the δO^{18} of a large magma body. However, as pointed out by Taylor (1968), other things being equal, we would expect larger O¹⁸ effects to be observed in small magma bodies, such as the thin sheets of granophyric melt formed during the latest stages of crystallization of the Skaergaard magma. It is also possible that for a short time H₂O pressures in the vicinity of a pluton may be locally raised to values equal to or slightly greater than the lithostatic pressure when a catastrophic event like cauldron subsidence occurs, perhaps bringing water-rich rocks directly into contact with magma. However, this would be a transient effect and would in any case be more likely to cause violent steam explosions than to greatly enhance migration of H₂O directly into a magma.

Obviously, more data must be gathered on this problem, but it seems likely that

some model or models involving indirect exchange between magmas and meteoric water will be necessary to account for most low- O^{18} magmas. The most logical ways in which this might occur are: (1) by direct melting, above a magma chamber, of water-rich country rocks that had already been hydrothermally altered and strongly depleted in O^{18} by the meteoric-hydrothermal circulation system set up above the intrusive body; (2) by large-scale assimilation and dissolution of such low- O^{18} rocks directly into the magma; (3) by sinking of low- O^{18} xenoliths through the magma chamber, because the H_2O in the abundant hydrous minerals would certainly be driven off into the magma, and because in hydrous magmas such xenoliths rapidly exchange O^{18} with their host magma (Shieh and Taylor, 1969) although they may show no evidence of dissolution by the silicate melt; and finally (4) by direct exchange between the liquid magma and O^{18} depleted country rock at the edge of the magma chamber or along a fissure through which the magma penetrates. The fourth mechanism is actually a variant of (3), and it is known to occur invariably in deep-seated plutonic or mesozonal environments whenever small bodies of magma are intruded into country rocks having a distinctly different O^{18}/O^{16} ratio (Taylor and Epstein, unpublished data; Shieh and Taylor, 1969; Turi and Taylor, 1971).

Direct melting of hydrothermally altered roof-rocks above a magma chamber would be favored by the large amounts of H_2O present in that environment. If these melted rocks remain separate from the underlying magma (because of lower density or low mixing rates), such melts conceivably could have δO^{18} values as low as any of the hydrothermally altered rocks from which they formed. However, if either of the assimilation mechanisms is involved, extreme O^{18} depletions in the magmas would not be expected.

Note that the more complicated the intrusive igneous history, the more likely it is that one or all of these four mechanisms will be actuated. In particular, multiple intrusion, ring-dike formation, repeated cauldron subsidence, and periodic explosive activity would all be expected to be accompanied by the developments outlined above. The lack of O^{18} depletion of the main mass of Skaergaard magma may be due to the fact that the Skaergaard intrusion represents almost an end-member example of a single, simple textbook intrusion of basaltic magma that subsequently underwent a relatively straightforward sequence of fractional crystallization.

Continued assimilation or melting of water-rich hydrothermally altered rocks at the top of a magma body emplaced into a permeable volcanic terrane would after a period of time undoubtedly lead to H_2O saturation in the upper portions of the magma chamber. Water pressure could then exceed lithostatic pressure and an explosion might ultimately result, causing eruption of low- O^{18} magma from the top of the chamber. Release of pressure on the underlying H_2O -undersaturated magma column would cause it to vesiculate as well, and the entire magma column might then erupt. Thus, meteoric water may be the underlying cause of many ash-flow tuff eruptions in volcanic fields such as the San Juan Mountains.

EVIDENCE FOR PRESERVATION OF MAGMATIC ZONING IN CERTAIN PLAGIOCLASE CRYSTALS THAT HAVE UNDERGONE O^{18} EXCHANGE

One of the interesting textural features of many of the low- O^{18} igneous rocks from the San Juan Mountains (and other areas) is the presence of delicate, oscillatory zoning in much of the plagioclase. This is clearly a magmatic phenomenon, and most explanations of this feature have emphasized the role of H_2O pressure in some way. Repeated build-up and sub-

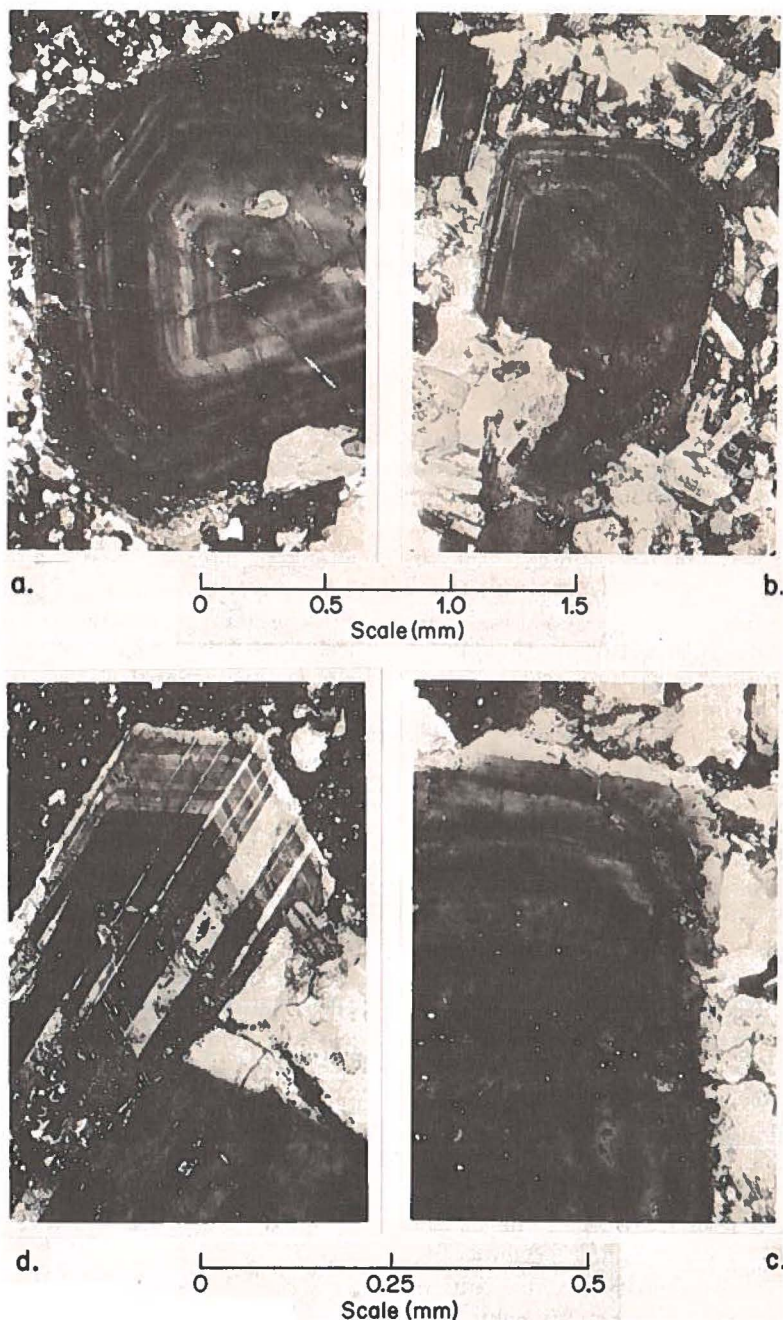


Fig. 5. (a) Photomicrograph (X-Nicols) of a typical plagioclase phenocryst ($\delta^{18}\text{O} = +6.1$) in Col-28-8 from the Eastern San Juan Mtns. (see Table 1). Although this rock has suffered some low- O^{18} alteration ($\delta^{18}\text{O}$ of chloritized biotite = -0.9), the plagioclase has not undergone any appreciable O^{18} exchange, compatible with the well-preserved oscillatory zoning. (b) Photomicrograph (X-Nicols) of a typical plagioclase phenocryst ($\delta^{18}\text{O} = +2.6$) in sample O-23-9 from the Western Cascades (see Fig. 7 and Table 2). All plagioclase phenocrysts in this granodiorite sample from the Champion Creek stock show well-developed oscillatory zoning, even though the plagioclase has clearly undergone marked O^{18} depletion after its original crystallization from the granodiorite

sequent partial release of H_2O pressure in the magma, recurring on a fairly regular basis, seems to be a very adequate way to produce such oscillatory zoning. This is because of the sensitive response of the temperature of the liquidus in any silicate melting diagram to fluctuations in P_{H_2O} . In the light of the previous discussion involving incorporation of meteoric H_2O into magmas, it is not unreasonable to expect that this type of plagioclase would be very common in volcanic-intrusive terranes.

Many of these oscillatory-zoned plagioclases are strongly depleted in O^{18} . Photomicrographs of some examples are shown in Figs. 5 and 6. The question immediately arises as to whether this textural feature implies that the oxygen in such plagioclase grains was "frozen in" at the time of magmatic crystallization, and has not been subsequently disturbed by exchange with the meteoric-hydrothermal solutions that have permeated many of these rocks. This is an important concept because if it is true, it means that the presence of well-preserved oscillatory zoning in a low- O^{18} plagioclase would demand that the plagioclases have crystallized from a low- O^{18} magma.

Unfortunately, the answer to the above question is both yes and no. The ambiguities arise mainly because of difficulty in quantifying some measure of what is meant by "well-preserved oscillatory zoning." It is certainly true that if one takes a random collection of igneous rocks from an area like the San Juan volcanic field and arranges them into a sequence from "best preserved" to "worst preserved," subsequent O^{18} analyses will show that this sequence correlates roughly with the δO^{18} values (highest to

lowest, respectively). An example of such a "well-preserved," high- O^{18} sample is given in Fig. 5a. However, the state of preservation of the magmatic zoning in the plagioclase also correlates with the degree of hydrothermal alteration of the rock in general, and it is certainly to be expected that the amounts of chlorite, epidote, calcite, actinolite, etc., in the rock should correlate roughly with O^{18}/O^{16} ratio, as in fact they do.

Nevertheless, sufficient O^{18} data now exist to state fairly positively that the oscillatory zoning in a plagioclase grain can survive almost complete oxygen isotope exchange with meteoric-hydrothermal fluids. Several examples of this effect are illustrated in Figs. 5 and 6. It must be emphasized that none of the O^{18} analyses of these plagioclases is on a single grain; they all represent analyses of 10–15 mg separates of a large number of grains, and it cannot be demonstrated that all grains show oscillatory zoning nor that the zoning is equally "well preserved" in all cases. However, one excellent example in which almost every plagioclase grain in the thin section is zoned in this fashion is sample O-23-9 (Table 2) from the Western Cascade Range, Oregon (Fig. 5b). The location of this sample is shown on the map in Fig. 7, modified after Fig. 4 of Taylor (1971).

Sample O-23-9 was chosen for O^{18} analysis in the present study because the plagioclase in thin-section shows such well-developed oscillatory zoning; it had not been analyzed previously by Taylor (1971). Note that this sample well illustrates the point made above about a correlation between δO^{18} value and "degree of preservation," because all other analyzed samples of the Champion Creek

magma. The whole-rock $\delta O^{18} = +1.6$, indicating that the fine-grained groundmass has undergone even more marked O^{18} -depletion than have the plagioclase phenocrysts. (c) Photomicrograph (X-Nicols) at higher magnification of another typical plagioclase phenocryst from O-23-9. (d) Photomicrograph (X-Nicols) of a reasonably typical plagioclase phenocryst in SM 147, an inclusion of San Juan Tuff in the Stony Mtn. ring-dike complex (Forester and Taylor, 1962). These plagioclase grains are strongly depleted in O^{18} ($\delta = +2.3$) relative to values in "normal" igneous rocks, but it is possible that this effect may in part be due to crystallization from a low- O^{18} magma (see text).

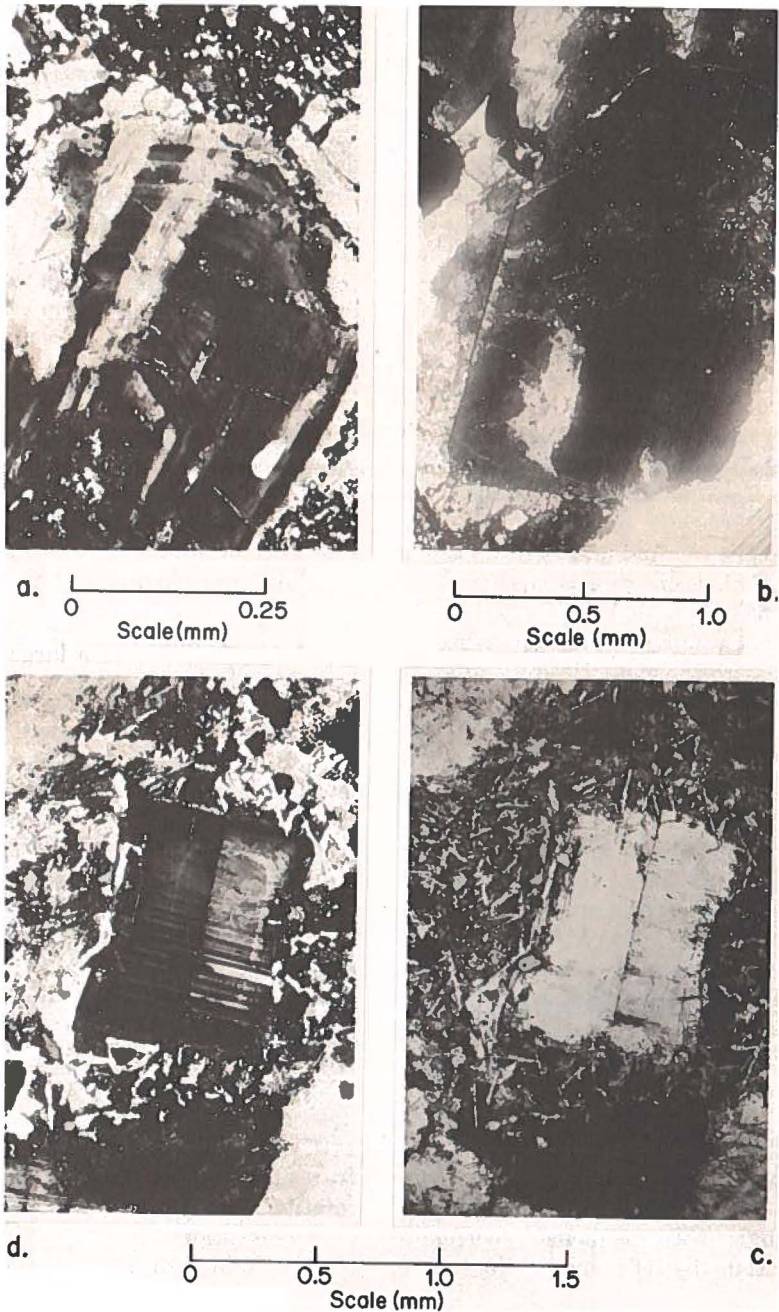


Fig. 6. (a) Photomicrograph (X-Nicols) of some of the better preserved oscillatory zoning shown by plagioclase phenocrysts in O-28-9, a propylitically altered andesite from the Western Cascade Range, Oregon (see Table 2). Many of the plagioclase phenocrysts in this rock show evidence of recrystallization in thin section, but remnant oscillatory zoning is common. The rock has undergone intense O^{18} depletion (δO^{18} whole rock = -5.0), and so have the plagioclase phenocrysts (δO^{18} = -4.1). (b) Photomicrograph (X-Nicols) of G-260, a gabbro from the Upper Border Group (α) of the Skaergaard intrusion. The labradorite crystals have preserved a normal magmatic zoning (calcic cores) even though their oxygen has been totally exchanged (δO^{18} plagioclase = $+2.0$;

stock shown in Fig. 7 are considerably more depleted in O^{18} . The O-23-9 plagioclase is drastically out of equilibrium with its coexisting quartz and after crystallization must have been depleted in O^{18} by at least 2 per mil. Some of the cores of the O-23-9 plagioclase grains are recrystallized, but not enough to explain the entire O^{18} effect. It thus seems clear that although the oscillatory zoning in the plagioclase of O-23-9 is somewhat "fuzzy" and not as sharp as, for example, that shown by the high- O^{18} sample in Fig. 5a, the basic zoning pattern has been essentially preserved during the O^{18} recrystallization process. Another grain shows this at higher magnification in Fig. 5c.

Analyses were also made on an altered andesite (O-28-9) from the most O^{18} -depleted locality in the Western Cascades (Table 2, see Fig. 5 of Taylor, 1971). Most of the abundant fine-grained, plagioclase phenocrysts in this rock are oscillatory-zoned, but the zoning is less well preserved than in O-23-9 (Fig. 6a). This plagioclase ($\delta O^{18} = -4.1$) has been depleted in O^{18} by about 10 per mil during hydrothermal alteration, based on comparisons with analogous volcanic rocks outside the area of propylitic alteration (Taylor, 1971).

Other samples which suggest the same conclusion are SM 147 (Fig. 5d), from an inclusion of San Juan tuff in the Stony Mountain ring-dike complex (see Forester and Taylor, 1972). The zoning in the plagioclase of this sample is exceptionally well preserved, even though the plagioclase has a relatively low $\delta O^{18} = +2.3$. Unfortunately, we do not know how much O^{18} depletion may have occurred in the San Juan tuff magma prior to eruption,

so we do not know exactly how much δO^{18} lowering of this sample is due to later rock-water interaction.

Some of the best examples of preservation of zoning during O^{18} exchange are rocks of the Upper Border Group of the Skaergaard intrusion (Taylor and Forester, 1973). Some of these are coarse-grained (Fig. 6). Although they do not show the type of delicate oscillatory zoning discussed above, they do show pronounced magmatic zoning and here there is no ambiguity whatsoever; all the oxygen in these Skaergaard plagioclases has been totally exchanged with meteoric-hydrothermal solutions, as clearly shown by the strong isotopic reversal in the plagioclase-clinopyroxene O^{18} fractionations. The plagioclase has been depleted in O^{18} by at least 3 to 4 per mil, while the coexisting clinopyroxenes are virtually unaffected.

In summary, virtually all the plagioclase grains in the O^{18} -depleted igneous rocks shown in Tables 1 and 2 and Fig. 4 are in fact "pseudomorphs" after the original igneous plagioclase grains. In many cases, the magmatic textures in the rocks are almost perfectly preserved (a good example is the Skaergaard intrusion); and the internal zoning in the feldspars is also often found to be preserved. However, in areas of progressively more intense meteoric-hydrothermal alteration, the internal plagioclase zoning is progressively destroyed, just as are the other primary igneous textural features in the most strongly altered rocks. In general, the calcic cores of the oscillatory-zoned plagioclase grains are destroyed first, and the oscillatory zoning itself tends to become less and less distinct.

δO^{18} clinopyroxene = +3.8, Taylor and Forester, 1973). (c) Photomicrograph (plane light) of G-238, a granophyric ferrodiorite from the Upper Border Group (β) of the Skaergaard intrusion. The tiny prismatic crystals surrounding the plagioclase are quartz that has inverted from tridymite. (d) Photomicrograph (X-Nicols) of G-238. The andesine grains in this rock characteristically show a few well-developed oscillatory zones, even though the oxygen in the feldspar crystals has been totally exchanged (δO^{18} plagioclase = +1.7, δO^{18} clinopyroxene = +4.6).

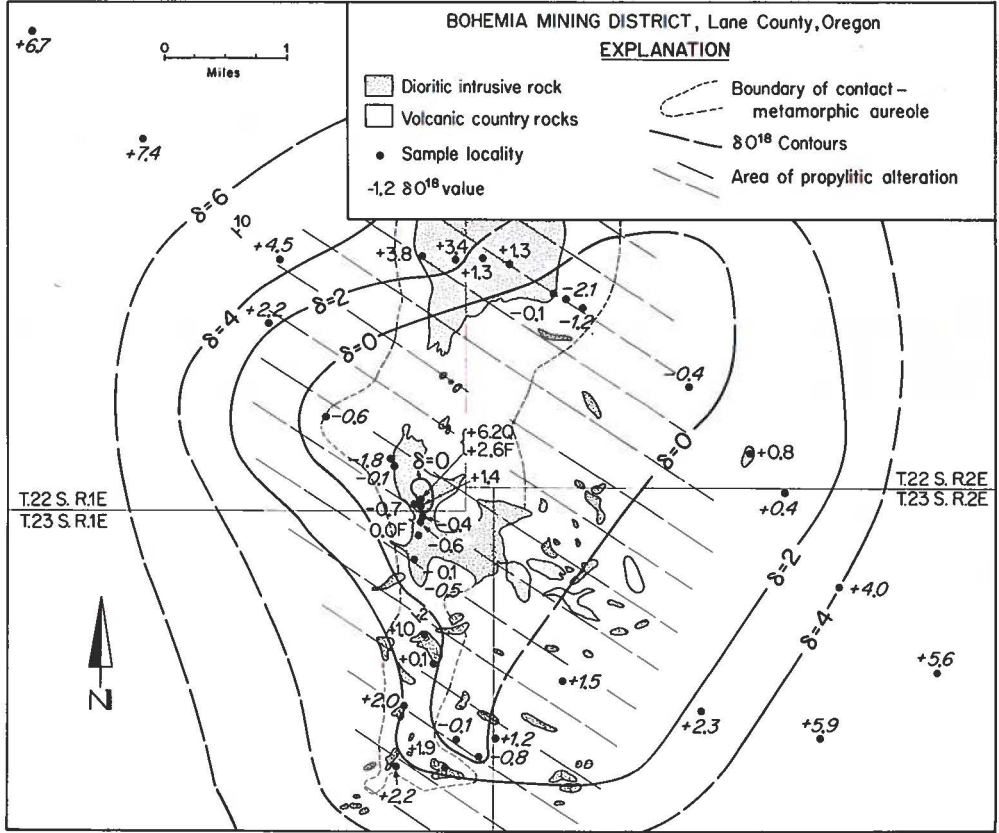


Fig. 7. Generalized geologic map of the Bohemia Mining District, Western Cascade Range, Oregon, showing $\delta^{18}\text{O}$ values on whole-rock samples of epizonal intrusions (regular lettering) and volcanic country rocks (italic lettering), modified after Fig. 4 of Taylor (1971). Notation: Q = quartz, F = plagioclase. New $\delta^{18}\text{O}$ data on granodiorite sample O-23-9, discussed in the text, are shown on the map ($\delta^{18}\text{O}$ quartz = +6.2, $\delta^{18}\text{O}$ plagioclase = +2.6). This sample is located near the center of the Champion Creek stock; it has the highest $\delta^{18}\text{O}$ value of any granodiorite sample analyzed from the stock, and its plagioclase also shows the best-preserved oscillatory zoning (see Fig. 5b).

Solutions clearly penetrated the plagioclase crystals along cracks and imperfections, and it is reasonable to believe that they preferentially recrystallized the more calcic zones, thus exchanging oxygen with the feldspar but retaining as a relict texture the gross features of the original feldspar grains. If this occurs by a fine-scale solution and redeposition process in nature as it seems to do in some cases in the laboratory (O'Neil and Taylor, 1967), then the primary reason for partial preservation of the oscillatory zoning may well be the relative insolubil-

ity of aluminum in these solutions; the Al/Si ratio in the plagioclase grain will control the Na/Ca ratio regardless of how easily the Na and Ca exchange. It is also possible that under some hydrothermal conditions oxygen may diffuse through the feldspar lattice more easily than do the cations; this would also allow O^{18} exchange while preserving the oscillatory cation zonation. There is, in fact, some experimental evidence that this may be the case (see Yund and Anderson, this volume).

HIGH-O¹⁸ PRECAMBRIAN IGNEOUS ROCKS

A remarkable feature of the oxygen isotope investigations carried out on igneous rocks (Taylor, 1968; 1971; 1973; this work), is that practically without exception the occurrences of low-O¹⁸ igneous rocks are of Tertiary or late Mesozoic age (see Fig. 4). Only two such occurrences are known from Precambrian terranes, a 600 m.y. old granite from the Seychelles Islands and an isolated occurrence of granite in the 1450–1500 m.y. old volcanic-intrusive terrane in the St. Francois Mountains, Missouri. Neither of these two examples is strongly depleted in O¹⁸, as the whole-rock δ values are no lower than +3.0 and +5.0, respectively (Taylor, 1968; Wenner and Taylor, 1972). In part, the lack of low-O¹⁸ rocks in older terranes is undoubtedly due to a lesser degree of sampling, as well as to the fact that later erosion and metamorphism may have removed many of the examples of such rocks. However, in spite of these qualifications, enough O¹⁸ data are on hand to indicate clearly that some peculiar process was involved in the petrologic history of some of these older igneous rocks.

Not only are low-O¹⁸ rocks largely absent from Precambrian terranes, many examples of these older igneous rocks throughout the world display exactly the opposite effects and are in fact abnormally high in O¹⁸! In most instances these high whole-rock δ O¹⁸ values are accompanied by strong disequilibrium in quartz-feldspar O¹⁸ fractionations, but instead of the feldspar being abnormally depleted in O¹⁸ as in the Tertiary examples, the feldspars consistently have higher δ O¹⁸ values than the coexisting quartz. Localities where this phenomenon has been shown to exist are shown on a map of North America in Fig. 8, and the isotopic data are shown in Figs. 9 and 10. Such effects have now been observed in the granophyres and granitic rocks of the Muskox, Bushveld, and Duluth layered igneous complexes, and in various parts

of the Keweenaw volcanic terrane, as well as throughout the sequence of ash-flow tuffs and epizonal granitic intrusions in the St. Francois Mountains, Missouri. Other examples occur in a pegmatitic vein in the Precambrian basement rocks brought up in the drill-core cuttings of a well at Sandhill, West Virginia, and in the hydrothermal alteration zones along fractures in the Precambrian Basement complex at Hopedale and Nain in Labrador.

All of the above examples have the following characteristics in common. The disturbed alkali feldspars are all turbid and brick-red in color due to disseminated hematite dust. The mafic minerals are partially or completely converted to chlorite. Several of the samples display granophyric textures. Except for the ubiquitous presence of hematite dust and their high-O¹⁸ character, these samples are in fact similar in character and to a certain extent in geologic setting to the low-O¹⁸ Tertiary igneous rocks described above. This is particularly true of the St. Francois Mountains terrane.

To what then should we attribute the reversed oxygen isotope behavior in these Precambrian rocks? There are basically only two possibilities: either (1) the hydrothermal solutions were much higher in O¹⁸ than those involved in the Tertiary occurrences; or (2) O¹⁸ exchange and alteration occurred at much lower temperatures (~150°C) in the Precambrian examples. Either of these hypotheses presents difficulties, and sufficient data are not yet available to settle this problem. In either case, very large amounts of H₂O must have circulated through and exchanged with these Precambrian rocks.

Hypothesis (1) demands either that the meteoric ground waters in the Precambrian were much higher in O¹⁸ than Tertiary or present-day ground waters, or that these Precambrian granitic rocks contained far higher concentrations of primary magmatic water than did the younger igneous rocks. There is no in-

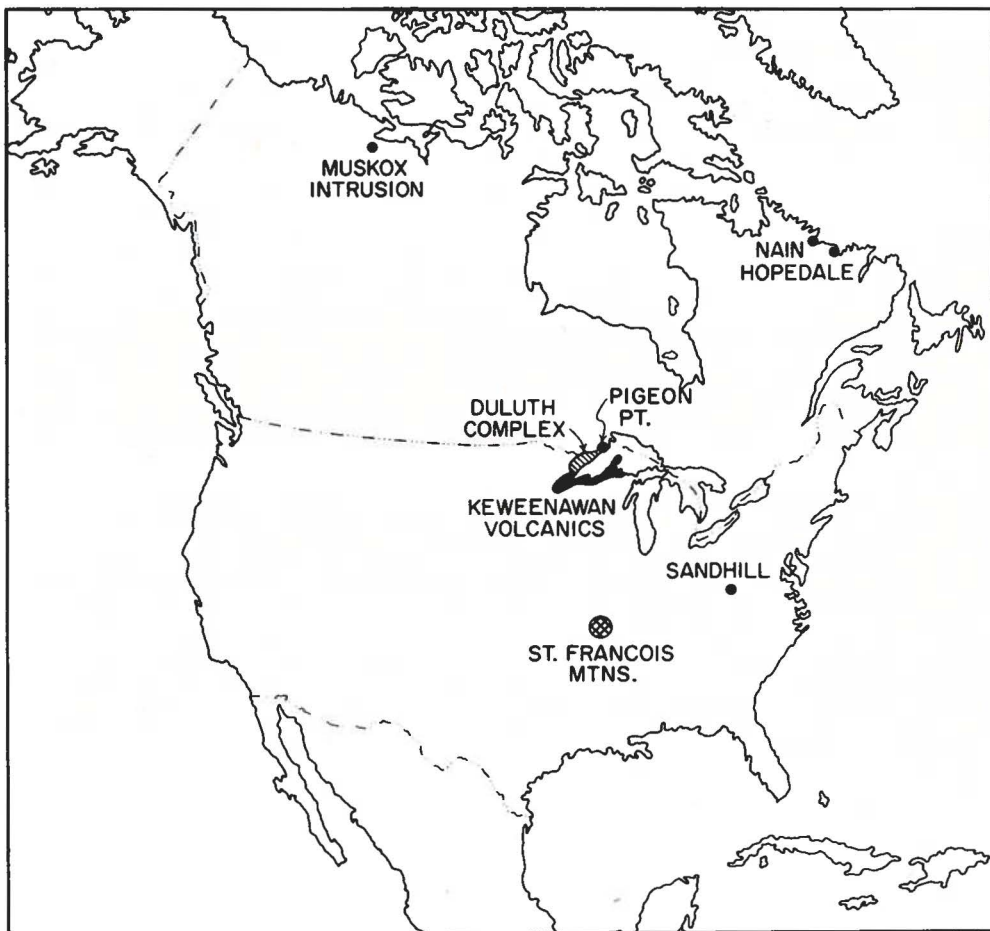


Fig. 8. Map of North America showing localities of Precambrian rocks discussed in the text, all of which show "reversed" quartz-feldspar O^{18} fractionations, abnormally high whole-rock δO^{18} values, and dissemination of hematite dust throughout the feldspar grains. To these North American localities we should also add the brick-red Bobbejaankop granite from the Bushveld Complex, South Africa.

dependent evidence that the latter was true, and if the former was true the only way it could logically come about would be if the Precambrian oceans were at least 5 to 10 per mil heavier in O^{18} than they are at present. The position of the meteoric water line on a δD - δO^{18} plot such as that shown in Fig. 4 is essentially fixed by the isotopic composition of the oceans existing at the time.

If the ocean was originally derived by degassing of the Earth's interior at high temperatures, its original δO^{18} value

should have been about +6 rather than its present value of zero. This is because such H_2O would have been in exchange equilibrium with basalts and ultramafic rocks in the mantle, and these rocks have $\delta O^{18} = +5$ to +6. At about $1000^\circ C$ the O^{18} fractionation between such rocks and H_2O is close to zero. Thus it is not unreasonable that at some time in the Earth's history the oceans may have been approximately 6 per mil richer in O^{18} than they are at present. However, as soon as sedimentation began, with con-

sequent precipitation of low-temperature, high-O¹⁸ marine minerals (such as cherts, clay minerals, carbonates) the oceans should have become steadily lower in O¹⁸ with time. Material-balance calculations imply that the "locking-up" of so much O¹⁸ in these low-temperature sedimentary rocks must be balanced by loss of O¹⁸ in some other part of the system, and the only suitable candidate is the ocean water reservoir itself (see Silverman, 1951; Savin and Epstein, 1970).

Thus, a reasonable mechanism does exist by which the early Precambrian oceans may have been a few per mil high in O¹⁸. However, many of the high-O¹⁸

igneous rock samples under discussion were formed in the interval 1100 to 1500 m.y. ago. At this relatively late stage in the Earth's history, the oceans had been in existence for at least 2000 m.y. and cycles of sedimentation had occurred many times. It therefore seems unlikely that these later Precambrian oceans would still have had a primordial δO^{18} value $\approx +6$. In fact, based upon data on the evolution of δO^{18} values of cherts with time, Perry (1967) has presented an entirely different model for the oceans, proposing that the oceans were lower in O¹⁸ during the Precambrian than they are at present.

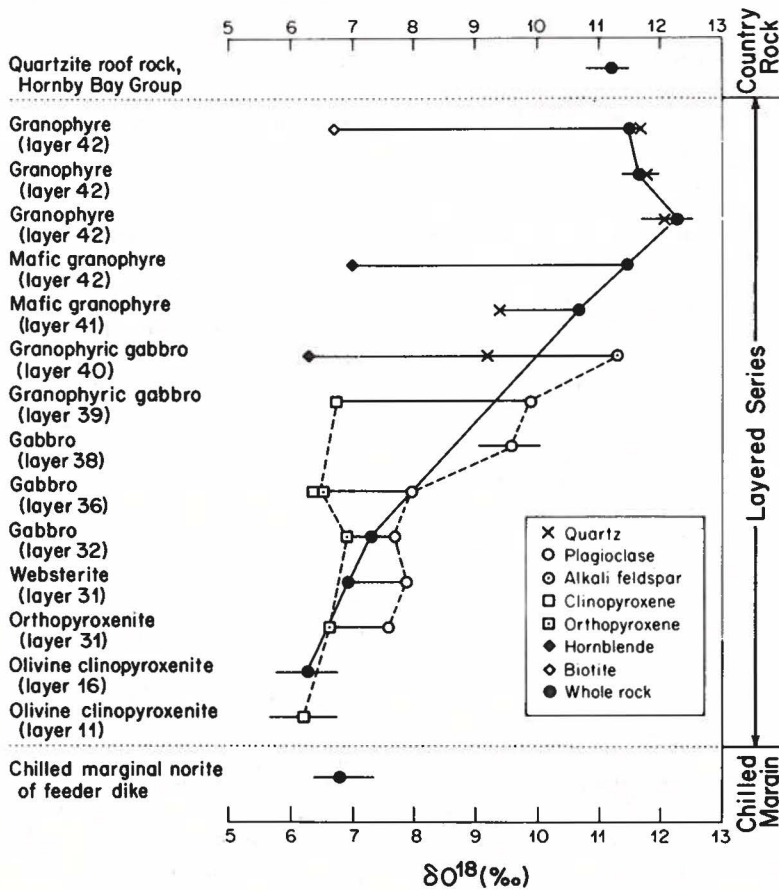


Fig. 9. Oxygen isotope data obtained on minerals and rocks from the Muskox intrusion, northern Canada, after Taylor (1968). Note the "reversed" quartz-feldspar fractionations in some of the late-stage, red-rock granophyres, and the strong O¹⁸-enrichment in the feldspars at the top of the intrusion.

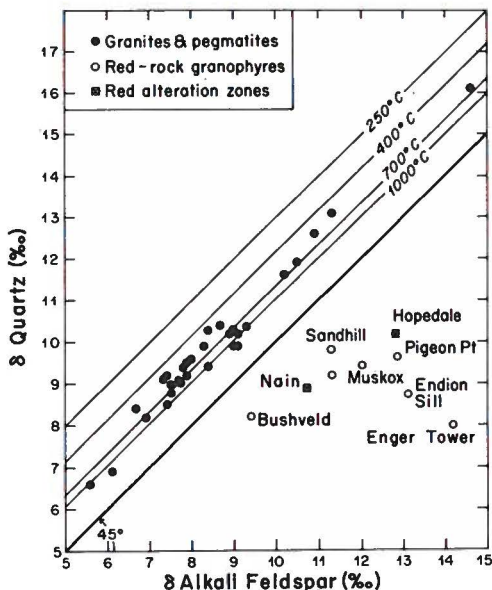


Fig. 10. Plot of δO^{18} in feldspar vs δO^{18} in quartz for various igneous rocks, including the Precambrian samples indicated in Fig. 8. The isotherms drawn parallel to the 45° line represent the equilibrium quartz-alkali feldspar O^{18} fractionations at various temperatures, based on the partial exchange quartz- H_2O experiments of Clayton *et al.* (1972) and the feldspar- H_2O experiments of O'Neil and Taylor (1967). Note that most of the "normal" igneous rock samples (black circles) exhibit fairly reasonable isotopic "temperatures," whereas all of the Precambrian red-rock samples represent drastic nonequilibrium. Data on the "normal" samples is from Taylor (1968) and Taylor and Epstein (1962 and unpublished data). Note that the nonequilibrium quartz-feldspar data-points for the low- O^{18} Tertiary examples discussed in the text would plot well to the left of this diagram. For the Nain and Hopedale data-points, which represent red hydrothermal alteration zones along fractures in gneiss, there is no change in δO^{18} quartz in going from the unaltered gneiss into the alteration zone (Taylor, 1967).

The history of oxygen isotopic evolution of the oceans with time remains a problem of prime geochemical importance, and further investigations of Precambrian igneous rocks that have exchanged with hydrothermal waters of surface origin may cast some light on this problem. However, an evaluation of the presently available evidence strongly

suggests that the Precambrian oceans 1000 to 1500 m.y. ago were probably not significantly higher in O^{18} than they are at present. Well-preserved limestones and cherts formed at this time are not significantly different from analogous Paleozoic and Mesozoic samples (Knauth, 1973). Also, material-balance calculations by Savin and Epstein (1970) indicate that sedimentation has probably been responsible for no more than about a 1 per mil depletion of the oceans in the last billion years.

The extended discussion above leads us to the tentative conclusion that hypothesis (2) above is the most likely explanation of the high- O^{18} , brick-red feldspars in Precambrian igneous rocks. It is in fact likely that such rocks were in contact with circulating water at 100° – $200^\circ C$ for extended periods of time because they have all been deeply buried under younger sedimentary or volcanic rocks which would have contained connate waters or meteoric ground waters. For example, soon after the Muskox intrusion had completed crystallization, the entire area was buried under the very thick section of Coppermine River Basalt flows (Irvine and Smith, 1967). As another example, brines with δO^{18} values of -4 to $+4$, δD values of -40 to $+20$, and salinities of 1 to 6 gram-equivalents per liter are common throughout sedimentary basins in the mid-continent and Gulf Coast regions of the United States (Clayton *et al.*, 1966). Brines with similar characteristics have been identified in fluid inclusions of minerals that make up the Mississippi Valley type Pb-Zn deposits in the Paleozoic sediments of the mid-continent region of the U.S. (Hall and Friedman, 1963). Ore deposits of this type are common throughout the world in such sedimentary sections, and various geothermometric techniques suggest they formed at about 100° – $200^\circ C$. Also, the anomalous character of the Pb isotopic compositions in the galenas of these deposits strongly suggests that the hydrothermal solutions from which they

were derived must have circulated through the underlying Precambrian basement (Heyl *et al.*, 1966).

At 100° to 200°C, the equilibrium alkali feldspar-H₂O oxygen isotope fractionation varies from +17.5 to +9.6 (O'Neil and Taylor, 1967). Exchange between feldspars and one of the aforementioned brines at these temperatures would lead to feldspar δO^{18} values similar to those shown in Fig. 10. The relatively heavy δD values of such brines are also compatible with the high δD values obtained on Keweenawan and St. Francois igneous rocks, as shown in Fig. 4, and by Forester and Taylor (unpublished data). Oxygen isotope exchange would be much more likely with such a brine than with low-salinity meteoric ground waters because the brine would not be in cation equilibrium with the feldspar and would thus tend to attack the feldspar (Orville, 1963; O'Neil and Taylor, 1967). Exchange with such low-temperature hydrothermal fluids would be strongly oxidizing, thereby explaining the ubiquitous hematite dust disseminated through the feldspars (note that because of the shapes of oxygen buffer curves on a plot of P_{O_2} vs temperature, a simple lowering of temperature will move a system into steadily more oxidizing conditions and finally into the hematite field of stability (see Eugster, 1959)).

It is conceivable that some of the iron necessary to form the ubiquitous hematite dust was originally present in solid solution in the alkali feldspars (e.g., see Ernst, 1960). However, it is even more likely that this Fe is released during the hydrous alteration of the coexisting mafic minerals in the rocks (D. R. Wones, personal communication). In any case, the constant association of high O^{18}/O^{16} ratios and hematite dust in these feldspars is clearly not a coincidence; this implies the hematite was definitely *not* formed by a simple exsolution process but must have been formed by the hydrothermal event that produced the high δO^{18} values in the feldspars (Taylor,

1967). Inasmuch as the hydrothermal O^{18} exchange process requires a complete breaking and re-forming of all Si-O and Al-O bonds throughout the feldspar crystal, it is perfectly reasonable that the Fe was derived from outside the original feldspar grain, as proposed by Wones.

Why, then, are these particular Precambrian alkali feldspars so susceptible to exchange by such low-temperature hydrothermal fluids? The answer probably lies in the fact that many of the feldspars that have been through a high-temperature hydrothermal exchange event (such as has affected all of the Tertiary examples discussed previously) are commonly turbid and full of imperfections and fluid inclusions. Such feldspars are probably much more susceptible to hydrothermal exchange during a later, lower-temperature event. One indication that this two-stage process might have occurred, at least in the St. Francois Mountains, is the apparent local preservation of some low- O^{18} rocks that may reflect an earlier episode of exchange with "normal" low- O^{18} meteoric waters (Wenner and Taylor, 1972). Certainly the general similarity between the geologic history of the Precambrian St. Francois Mountains volcanic terrane and the Tertiary San Juan volcanic field would imply that if low- O^{18} meteoric ground waters were present during the Precambrian, they would have interacted on a large scale with the St. Francois igneous rocks just as they have in the San Juans.

One other phenomenon that might have been produced by the postulated low-temperature hydrothermal event is a disturbance of Rb-Sr systematics in these rocks. Since these rocks were clearly "open" to a great deal of water that may have been very saline, any trace elements present in the highly altered feldspars and mafic minerals would be expected to have undergone redistribution. If this event occurred a significant time interval after primary crystallization of the red-rock granophyres and volcanic rocks, it could produce abnormally young Rb-Sr

isochron ages. Just such a pattern is developed in the Keweenaw, Duluth, and St. Francois Mountains terranes because the Rb-Sr ages invariably show a greater spread and are slightly younger than the U-Pb concordia ages obtained from zircons (Faure *et al.*, 1969; Chaudhuri and Faure, 1967; Silver and Green, 1963; Bickford and Mose, 1972). Bickford and Mose (1972) report that the Rb-Sr ages in the St. Francois Mountains tend to be "chaotic" and are up to 13 percent younger than the zircon ages; the zircon ages establish fairly clearly that the primary igneous events in this area occurred over a narrow time interval about 1475 m.y. ago. Note that recently formed zircons that have suffered little radiation damage or metamictization are not very susceptible to alteration during a hydrothermal alteration event (L. T. Silver, personal communication).

The envisaged hydrothermal event may have been responsible for such effects as the extensive serpentinization of the Muskox intrusion, and both there and in other areas such an event could have occurred intermittently over a very long time, perhaps extending into the Paleozoic Era or later. The postulated temperatures of 100°–200°C might either be due to another episode of magmatic activity in the various localities or simply to slight enhancement of the normal geothermal gradient (with perhaps just enough lateral temperature variation to produce some convective circulation of the H₂O). In any case, this second stage of the two-stage process might have little or nothing to do with the original magmatic heat in the intrusion. Therefore, although the "deuteric" explanation of these isotope data proposed previously (Taylor, 1967, 1968) may still have some local validity, it is very doubtful that it can serve as a general explanation (unless, of course, the oceans and hence the meteoric waters were in fact much higher in O¹⁸ during the mid- to late-Precambrian than they are at present).

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