

Oxygen Isotopic Ratios in Quartz as an Indicator of Provenance of Dust

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3 Abstract--Quartz was isolated in the long range aerosol size range  
4 (fine silt, 1-10  $\mu\text{m}$  in diameter) from atmospheric aerosols, wind-erosive  
5 soils, soil silts, shales, and Pacific pelagic sediments of the Northern and  
6 Southern Hemispheres, to trace their provenance or origin, as part of a  
7 study of dust mineral sequestering of  $^{137}\text{Cs}$  and other products of nuclear  
8 fission. The oxygen isotopic ratio ( $^{18}\text{O}/^{16}\text{O}$ ) was determined by mass spectro-  
9 metry. The provenance has been established for this fine silt fraction  
10 which reflects the relative proportion of two classes of quartz source:  
11 (a) weathering of igneous and metamorphic rocks (high temperature origin and  
12 low  $^{18}\text{O}/^{16}\text{O}$  ratio) and (b) of quartz crystallized in cherts and overgrowths  
13 (low temperature origin and high  $^{18}\text{O}/^{16}\text{O}$  ratio). This quartz mixing ratio  
14 is a basic model or paradigm.

15 The  $\delta^{18}\text{O}$  value ( $^{18}\text{O}/^{16}\text{O}$  relative to standard sea water, expressed as  
16 parts per thousand, per mil,  $^{\circ}/\text{oo}$ ) of quartz, once established in a rock, is  
17 very resistant to change at temperatures below  $350^{\circ}\text{C}$ . A selectivity of  
18 quartz origin in detrital sediments evidently occurs, with high temperature  
19 quartz preferentially remaining in beach sands and attenuating with fine-  
20 ness, while fine quartz from chert and chipped off overgrowths increases in  
21 the fine silt (1-10  $\mu\text{m}$  diameter) range (submodel I of the paradigm). Close  
22 proximity to eroding igneous-metamorphic mountain masses, however, results  
23 in  $\delta^{18}\text{O}$  values of fine silt quartz in the range of 12 to  $16^{\circ}/\text{oo}$ . (Quartz  
24 of beach sand has  $\delta^{18}\text{O} = 11 \pm 1^{\circ}/\text{oo}$ ). In contrast, central marine basins,  
25 lakes, and sediments that have accumulated a higher proportion of fine silt  
quartz crystallized from opal, including that from diatomaceous skeletons

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1 under the mean temperatures of sub-tropical to equatorial regions, have pre-  
2 dominantly  $\delta^{18}\text{O}$  values of 17 to 24 ‰. This distance-from-source relation-  
3 ship (submodel II of the paradigm) is recapitulated across various systems  
4 (e.g., the ancient Cretaceous Sea sediments extending from Montana to South  
5 Dakota). The largest scale example (submodel III of the paradigm) is the  
6 contrasting  $\delta^{18}\text{O}$  (mainly  $19 \pm 2$  ‰) of fine silt quartz in the large-scale  
7 silt reservoirs of the Northern Hemisphere continents, differing from that  
8 (mainly  $12 \pm 2$  ‰) for glacially ground quartz of the Southern Hemisphere.  
9 This hemispheric difference is attributable to paleogeographic differences  
10 associated with paleoclimatic differences now altered by continental drift  
11 (plate tectonics). Extensive land areas of the Southern Hemisphere were once  
12 part of a frigid South Polar supercontinent, Gondwanaland, and associated  
13 seas in late Paleozoic-early Mesozoic times. Glacial and fluvial transport  
14 of predominantly igneous and metamorphic rocks from mountains to the conti-  
15 nental shelves resulted. These shelves are now uplifted into lands, which  
16 drifted northward into the south temperate zone in Cretaceous times. As  
17 present Northern Hemisphere continents were carried across the tropical and  
18 equatorial zones, weathering on lands under equatorial and tropical climates  
19 caused a high rate of desilication of soils, leaching of soluble  $\text{Si}(\text{OH})_4$ ,  
20 which was then carried out to sea by rivers. Extensive biogenic silica  
21 formed at sea; later crystallization into mainly fine silt quartz crystals  
22 in chert and overgrowths in these Paleozoic and Mesozoic sediments resulted  
23 in a considerable proportion of low-temperature, high- $\delta^{18}\text{O}$  value quartz in  
24 vast areas of the now Northern Hemisphere continents. The early paleoclimatic  
25 and paleogeochemical differences remain the control of the North-South Hemi-  
sphere difference in  $\delta^{18}\text{O}$  values in long-range aerosol sized quartz.

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## INTRODUCTION

The objective of this paper is to outline the way in which the oxygen isotopic composition of the tracer mineral quartz provides a key to the origin or provenance of long-range aerosolic mineral dusts.\* The oxygen isotopic ratio of quartz from a given crystalline rock reflects the temperature of the quartz crystallization; however, fine quartz of sediments usually reflects a variable proportion of quartz, some crystallized at high and some at low temperature, which is the basic model or paradigm three submodels of which will be discussed. A scale from about 10 to about 30 ‰ (per mil, explained under "methods") provides a degree of uniqueness for various provenances of dust, which varies with the particle size range used for oxygen isotopic analysis, as will be discussed.

Natural mineral dusts of the atmosphere have long been of interest as attested by more than a century of writings such as those of Darwin, Richthofen, and Sturt, as summarized by Free (1911). A bibliography (98 pp.) of early literature was published by Stunz and Free (1911). An early light microscope study, which was prophetic of the world-wide

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\*"Long range aerosolic dust" is defined here as eolian mineral or glassy particles of dust small enough to be carried high aloft and great distances by winds of moderate intensity, in practice  $<10 \mu\text{m}$  equivalent diameter (Jackson et al., 1971; Walker and Costin, 1971). Loess, in contrast, has a wide range of particle sizes, up to  $70 \mu\text{m}$  (Simonson and Hutton, 1954; Smith, 1942).

1 systems of eolian dusts that will be traced herein, used the clause  
2 "dust which follows the winds of the world" (Udden, 1898).

3 Heating of atmospheric air masses carries eolian dust to  
4 great heights, believed to range from 1.5 to 5 km altitude (Prospero and  
5 Carlson, 1972). Interaction of these heated air masses with cold fronts  
6 carry the dusty air in the cyclonic and jet-stream circulation systems  
7 (Danielsen, 1968; Jackson et al., 1973). For example, dusts are blown  
8 from dry wadis in the Sahara desert and the dust clouds cross northward  
9 into Italy, Switzerland, and Austria and eastward into the Middle East  
10 (Yaalon and Ganor, 1973). Saharan dusts also pass westward across the  
11 tropical north Atlantic Ocean (Delany et al., 1967; Prospero, 1968; Syers  
12 et al., 1969). Dust accretion occurs on land and as pelagic sediments  
13 (Windom, 1969).

14 Periodic recharge of fine material in beds of braided streams allows  
15 intermittent wind action during dry periods to add increments of eolian  
16 silt (loess) to deposits in Alaska (Péwé, 1968) and probably to loess deposits  
Insert 17 generally (Ruhe, 1969; Smith, 1942), with aerosol dust added (Fig. 1).

Fig. 1 18 Similarly, atmospheric generation of eolian dust in dry areas of Asia gives  
19 rise to dust which is carried on the Westerlies and deposited as pelagic  
20 sediments ("red muds") of the North Pacific Ocean (Rex and Goldberg, 1958;  
21 Clayton et al., 1968, 1972; Rex et al., 1969) and quartz bearing silts in  
22 highland soils of Hawaii (Jackson et al., 1968, 1971). A major divergence  
23 in oxygen isotopic composition of long range aerosol size quartz occurs  
24 between the Northern and Southern Hemispheres (Clayton et al., 1972;  
25 Jackson et al., 1972; Mokma et al., 1972), which has invoked considerations

1 of the interrelationships of dust to paleoclimate, geomorphology, and  
2 continental drift (Jackson, 1974a, 1975). Amounts of dusts from soils vary  
3 with climate, and vary with forest, grass, and tundra vegetation under  
4 glacial climates; such dusts may quickly kill grass vegetation (Kukla et al.,  
5 1972). Tracing the provenance of terrestrial dust systems by oxygen  
6 isotopic composition therefore has important relevance to one cause-and-  
7 effect relationship of terrestrial climate to the atmosphere-ocean-cryosphere  
8 system. The micaceous mineral component of atmospheric dusts sequesters  
9  $^{137}\text{Cs}$  of fallout (Syers et al., 1972), a health safety factor adding importance  
10 to the study.

11 The inorganic mineral portion of the aerosol arises mainly from wind  
12 erosion of soil (Hidy and Brock, 1970; Jackson et al., 1973), which  
13 Goldberg estimated (quoted by Peterson and Junge, 1971) to be  $500 \times 10^6$   
14 tons  $\text{yr}^{-1}$ . The amount of dust is greatly affected by agricultural  
15 cultivation and other soil disturbances. Volcanic emanations, extra-  
16 terrestrial influx, sea salt, biological particles, and S and N compounds  
17 generally make up lesser components of aerosol dust. Aerosolic dust  
18 originating from soil and alluvial sediments is small in amount,  $2 \times 10^{11}$   
19 kg  $\text{yr}^{-1}$  compared to  $93 \times 10^{11}$  kg  $\text{yr}^{-1}$  carried in the fluvial load from  
20 estimated  
21 land to sea, but is double the present/load carried by glacial ice,  $1 \times 10^{11}$   
22 kg  $\text{yr}^{-1}$  (Judson, 1968; Jackson, 1974a).

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## METHODS

### Isolation of quartz

3 A 2- to 5-g sample of soil, dust, or other sediment is treated with 6N  
4 HCl at 100°C to destroy carbonates and some clay and then with 30% H<sub>2</sub>O<sub>2</sub> to  
5 destroy readily oxidizable organic matter. The desired size fraction  
6 (often 1-10  $\mu\text{m}$  in diameter) is then obtained by sieving, gravity sedimen-  
7 tation, and centrifugation (Jackson, 1974b). Fractions still showing black  
8 color (some shales) are boiled in a glass beaker for 2 minutes in 5 ml of  
9 concentrated HNO<sub>3</sub> (with drops of concentrated H<sub>2</sub>SO<sub>4</sub> present) and then 2 ml  
10 of 60% HClO<sub>4</sub> is added and the suspension is heated to fumes for a few  
11 minutes (hood) thereby removing the carbon and leaving a white residue;  
12 evolution of CO<sub>2</sub> in the subsequent Na<sub>2</sub>S<sub>2</sub>O<sub>7</sub> fusion is thus avoided (Jackson,  
13 1974b). Monomineralic quartz is then obtained by fusion of a 0.2 or 0.3 g  
14 sample in 12 g of Na<sub>2</sub>S<sub>2</sub>O<sub>7</sub> in a silica crucible, slowly at first and finally  
15 until only a slightly moist salt cake remains. After the crucible and  
16 contents are cooled, the cake is dissolved in 50 ml of, and the residue  
17 washed with, 3N HCl. These treatments do not affect the oxygen isotopic  
18 composition of quartz (Sridhar et al., 1975). Finally the residue is  
19 treated with 10 ml of 30% H<sub>2</sub>SiF<sub>6</sub> reagent (Syers et al., 1968) modified by  
20 the use of quartz flour sized at 1-100  $\mu\text{m}$  to quench the commercial H<sub>2</sub>SiF<sub>6</sub>  
21 and thus prevent its attack on the fine quartz being isolated (Jackson,  
22 1974b; Jackson et al., 1976). The suspension is allowed to stand 3 days  
23 at 18±1°C, being mixed twice daily during this period. The sample is  
24 rinsed momentarily with 0.1N HF and then washed with H<sub>2</sub>O. A small sub-  
25 sample is X-rayed at this point to check for feldspars. If they are

1 present, the  $H_2SiF_6$  and washing treatments are repeated (once, rarely twice  
2 may be needed). The scanning electron microscope is used for checking for  
3 volcanic ash glass or phytoliths and if present these are removed by heavy  
4 liquid separation. When the sample is free of contaminating minerals, the  
5 washed sample is treated overnight in saturated  $H_3BO_3$  solution to remove  
6 any fluorates in intersticies and washed four times with  $H_2O$  (Jackson,  
7 1974b).

8 Determination of oxygen isotopic ratio

9 Oxygen is isolated from the quartz fractions by the  $BrF_5$  procedure  
10 (Clayton and Mayeda, 1963). After measurement of oxygen yield the oxygen  
11 is converted to  $CO_2$  and analyzed isotopically by a  $60^\circ$ , 15 cm, double  
12 collecting mass spectrometer. Measured ratios are corrected for back-  
13 ground, mixing at inlet valve, and mass 44 tail (Becker and Clayton,  
14 1976). The data are reported as  $\delta^{18}O$  values, in parts per thousand (per mil,  
15  $^{\circ}/\text{oo}$ ), according to the equation,

$$16 \delta^{18}O = \frac{(^{18}O/^{16}O \text{ sample}) - (^{18}O/^{16}O \text{ standard})}{^{18}O/^{16}O \text{ standard}} \times 1000$$

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18 with standard referring to standard mean ocean water, SMOW (Craig, 1961).

19 The standard deviation for this analytical procedure is less than  $0.1^{\circ}/\text{oo}$ .

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## OXYGEN ISOTOPIC COMPOSITION OF QUARTZ

1        The oxygen isotopic composition expressed as delta value ( $\delta^{18}\text{O}$ ) in  
 2        sediments depends on the  $\delta^{18}\text{O}$  of the quartz in the crystalline rocks which  
 3        have been intermixed into the various grain sizes of the sediment. The  
 4        delta values for quartz from typical crystalline rocks are as follows:

$\delta^{18}\text{O}$  of quartz  
 (°/oo)

6        (a) From rocks of high temperature origin	
7              igneous rocks	8-10
8              metamorphic rocks	10-16
9              hydrothermal spring altered rocks	4-10
10        (b) From rocks of relatively low temperature origin	
11              dolomites and chert (40-10°C)	24-33
12              chert (80-40°C)	14-24

13        The igneous and metamorphic quartz values are from Taylor and Epstein (1962)  
 14        and Savin and Epstein (1970). The range for quartz from rocks of relatively  
 15        low temperature is from Knauth and Epstein (1976) and Kolodny and Epstein  
 16        (1976). The hydrothermal quartz values are from Clayton and Steiner (1975;  
 17        3.9 °/oo), Jackson et al. (1971; 5.5-8.3 °/oo), and Henderson et al. (1972;  
 18        9.6 to 10.0 °/oo).

### Variation with particle size in detrital sediments

20        The  $\delta^{18}\text{O}$  value of quartz, once established in a rock, is very resistant  
 21        to change at temperatures below 350°C (Clayton et al., 1968). The chert  
 22        field (upper part, Fig. 2) is mainly in the range of  $\delta^{18}\text{O}$  of 22 to 30 °/oo,  
 23        corresponding to 10 to 40°C formation temperatures as crystallization  
 24        (Knauth and Epstein, 1976) occurred at various stages of shallow burial.  
 25        The South African chert SC3 with  $\delta^{18}\text{O}$  of 14.6 °/oo shows field, morpho-  
             logical and isotopic evidence of having been metamorphosed by massive  
             basic rock intrusion (le Roux et al., 1977). Quartz from granite and

similar rocks, including an authigenic void filling in Precambrian quartzite (lower part, Fig. 2), accumulates in beach sands ( $\delta^{18}\text{O} = 11 \pm 1 \text{ ‰}$ ) and sand grains of sandstones without secondary overgrowths; however, overgrowths of chemically precipitated quartz extend the range to  $\delta^{18}\text{O} = 16 \text{ ‰}$  (Savin and Epstein, 1970). Low temperature overgrowths in pores of sandstones and siltstones may chip off during secondary transport.

A selectivity of quartz from different provenances evidently occurs according to particle size in detrital sediments (submodel I of the paradigm).

This is shown for eolian dusts and fluvial sediments by the central sloping lines of Fig. 2. The process of comminution of large high-temperature quartz grains to beach sand and finer sizes down to long range aerosolic dusts (1-10  $\mu\text{m}$  in diameter) evidently attenuates with fineness (Clayton et al., 1968,

1972; Syers et al., 1969). The  $\delta^{18}\text{O}$  values for Cambrian sandstone (Fig. 2) increase with fineness. An appreciable fraction of low temperature overgrowths may occur in the 2-5  $\mu\text{m}$  fraction. This size fraction, however, is an insignificant portion of the quartz in sandstone and, in Australian loess from beaches and New Zealand till and dust bands, has nearly the same  $\delta^{18}\text{O}$  ratio as coarser quartz silt (Table 1).

Isotopic ratio increase in fine fractions under burial diagenesis has been reported (Yeh and Savin, 1973). In contrast, authigenic quartz of chert initially is very fine (Sayin and Jackson, 1975). Occurrence as aggregates of fine grain size permit chert particles to extend up into the 50- $\mu\text{m}$  diameter and larger sizes (AR Baxter, Fig. 2; Sayin and Jackson, 1975). Mixing of chert and other quartz chips crystallized at low temperature with varying amounts of coarser detrital particles from high temperature rocks gives the sloping curves for the eolian dust silts, both in long range aerosol size (Jackson et al., 1968, 1971), and in loess (Sayers et al., 1969), as well as in fluvial sediments (Sridhar et al., 1975) and dusts (Gillette et al., 1977). Proximity to one or the other of the two types of quartz source determines the vertical position of the sloping lines (Fig. 2), as will now be discussed.

Insert

Table 1

Variation with distance from igneous-metamorphic rock sources

The systematic progression in the  $\delta^{18}\text{O}$  values indicates that the proportions of quartz from high temperature rocks relative to authigenic quartz decreases with distance from the mountainous western shore of the Cretaceous Sea (Fig. 3). The progressively higher  $\delta^{18}\text{O}$  values for quartz toward the central basin indicate a higher proportion of chert quartz originating (a) authigenically from biogenic (skeletal) amorphous silica and (or) (b) secondarily from fine sediments bearing quartz with heavier oxygen isotopic ratios. Analogously, the Tertiary non-marine sedimentary aprons spread east as the Rocky Mountains in Colorado were uplifted have quartz with lighter oxygen isotopes in the

1-10  $\mu\text{m}$  fraction (Fig. 2), reflecting a higher proportion of quartz from igneous sources (Taylor and Epstein, 1962; Taylor, 1968; Churchman et al., 1976). A  $\delta^{18}\text{O}$  value of 16.2 ‰ (Rex et al., 1969) for quartz from a dust of Japan (Miyake et al., 1956), a value of 12.6 ‰ for quartz off the mountainous Alaska Gulf Coast (Clayton et al., 1972), and the mean whole quartz value of  $14.7 \pm 0.9$  ‰ for southwestern U.S.A. (Table 1) also illustrate the importance of proximity to igneous rock sources.

The stair-step rise in quartz  $\delta^{18}\text{O}$  values in Fig. 2 from the Tertiary through Pacific pelagic sediments and Hawaiian soils (Jackson et al., 1971) and Lake Waiau sediment on Mauna Kea summit (Woodcock et al., 1966), Texas 1-10  $\mu\text{m}$  dusts, Miami R. of Ohio and Afghanistan silts, near-shore to central basin Cretaceous shale and shales generally of central U.S.A., to Maury and Baxter soils represent increasing proximity to chert sources of quartz (Churchman et al., 1976). This relationship of the high-low  $\delta^{18}\text{O}$  values for oxygen isotopic composition with proximity to low- or high-temperature rocks, respectively, is designated submodel II of the paradigm.

Insert  
Fig. 3

1        The Baxter soil overlying chert, the quartz of which has  $\delta^{18}\text{O}$  of 29.6 ‰  
2        (Fig. 2), has a mixture of quartz from chert and loess in various silt  
3        fractions (Sayin and Jackson, 1975) with a strong size function of oxygen  
4        isotopic ratio, indicated by the slope for various size fractions for two  
5        soil horizons. Thus size is very important when chert quartz is abundant.

6        Maury silt loam soil of central KY overlies cherty Ordovician dolomite  
7        the fine silt quartz of which has  $\delta^{18}\text{O} = 28.1$  ‰ (Syers et al., 1969).  
8        The silt in which this Paleudult (ancient humid region soil) more than three  
9        meters deep formed has quartz  $\delta^{18}\text{O}$  values of 19.6 to 20.9 ‰ (Fig. 2).  
10      The silt appears, therefore, to have originated mainly from silty shale  
11      interbeds of the dolomite (rocks of Maysville and Eden age) and loess  
12      derived therefrom, since these rocks make up the cuesta of the Lexington, KY  
13      basin in which the Maury soil occurs.

14      The mean  $\delta^{18}\text{O}$  value for eight WI loess 1-10  $\mu\text{m}$  quartz fractions  
15      (mixed dolomite-igneous rock area) is 21.3 ‰; the mean for this size  
16      quartz fraction from a column of 11 MS loesses is 17.9 ‰ (Fig. 2),  
17      presumably reflecting a larger influx of the igneous rock component into  
18      the eolian mixture (unpublished data). Although loess is generally gen-  
19      erated under periglacial conditions from glacial alluvium along streams,  
20      warm-climate loess is also known (Obruchev, 1945), originating from beaches  
21      of Victoria, Australia, for example (Jackson et al., 1972). Particle size  
22      fractionation of dusts occurs during transport (Johnson, 1976) and this may  
23      affect the proportions of quartz from igneous rock versus chert sources.

1      Variation with latitude of earth

2      Northern Hemisphere. An impressively narrow range of  $\delta^{18}\text{O}$  values  
3      (17 to 21  $^{\circ}/\text{o}$ ) for the long range aerosol size quartz particles of dusts  
4      (Table 1)  
5      and sediments/has been noted for North Pacific (Clayton et al., 1968, 1972;  
6      Jackson et al., 1968, 1971; Rex et al., 1969), Central North America  
7      (Churchman et al., 1976), and South East Asian areas (Syers et al., 1969).

8      We have found similar values for shales of Austria, loess in the central  
9      part of the Peoples Republic of China, and an eolian cap on a volcanic cone  
10     off the coast of Korea (unpublished data). Saharan dusts crossing the  
11     tropical North Atlantic have been collected on Barbados, Cape Verde  
12     Islands, and the Canary Islands and the  $\delta^{18}\text{O}$  values (unpublished data) for  
13     the 1-10  $\mu\text{m}$  quartz fraction also fall in the broad black bar for the  
14     Northern Hemisphere in Fig. 4. A scatter of values through the broad  $\delta^{18}\text{O}$   
15     spectrum is found for hydrothermal and chert systems, each representing  
16     more homogeneous temperature and geomorphological mixing processes.

17     Southern Hemisphere. In contrast to the values of 17 to 21  $^{\circ}/\text{o}$  for  
18     1-10  $\mu\text{m}$  quartz of eolian derived central North Pacific pelagic sediments  
19     and Hawaiian highlands and Northern Hemisphere continents, the comparable  
20     values in the South Pacific pelagic sediments (Clayton et al., 1968, 1972)  
21     range downward to 13  $^{\circ}/\text{o}$  (mean,  $15.1 \pm 1.5 ^{\circ}/\text{o}$ , Table 1). The regression  
22     with south latitude below  $6^{\circ}\text{S}$  is  $r = -0.73$  (Jackson et al., 1973). Eolian  
23     quartz from dusts falling on 11 soils developed in quartz-free basalt  
24     (6,000 to 4.5 million years old) has mean  $\delta^{18}\text{O}$  values of  $14.9 \pm 0.5 ^{\circ}/\text{o}$  and  
25      $14.5 \pm 0.2 ^{\circ}/\text{o}$  for the 2-5 and 5-20  $\mu\text{m}$  fractions, respectively, (Table 1)

Insert

Fig. 4

1 in southwestern Victoria and southeastern South Australia (Mokma et al.,  
2 1972). The  $\delta^{18}\text{O}$  values for 22 quartz samples from dust fall on glaciers  
3 and the fine and medium silt fractions of soils in New Zealand (Mokma  
4 et al., 1972) have means of  $13.2 \pm 0.2$  to  $13.5 \pm 0.4$  ‰ (Table 1). The  
5 lower  $\delta^{18}\text{O}$  values of eolian quartz in the soils and glacial ice of the  
6 Southern Hemisphere was attributed to the extensive source of aerosolic  
7 dust in the arid lands of Australia (Mokma et al., 1972). Those in the  
8 pelagic sediments were attributed to "a dominance in the Southern Hemisphere  
9 of older crystalline rocks of Australia and southern Africa" (Clayton  
10 et al., 1972). The long range aerosol size quartz (1-10  $\mu\text{m}$  diameter)  
11 isolated from 28 shales, mixed sediments, and soils in a large area of  
12 southern Africa (le Roux et al., 1977) has a mean value of  $12.2 \pm 2.1$  ‰  
13 (Table 1), represented by the black bar in Fig. 4. Again, there are  
14 scattered points for quartz of hydrothermal and unmixed chert systems  
15 formed at various temperatures scattered throughout the  $\delta^{18}\text{O}$  scale. The  
16 temperate latitudes of these continents make them partly arid lands subject  
17 to wind erosion of dust with low  $\delta^{18}\text{O}$  quartz (Jackson, 1975; le Roux  
18 et al., 1977). Extensive deposits of eolian clay (Parna) carried as silt  
19 size aggregates occur in east central Australia (Butler, 1956). Quartz  
20 from unmetamorphosed cherts (R125, 2000 m.y., Fig. 2) and old dolomites  
21 (RS13, 700 m.y., Fig. 2) in the Southern Hemisphere (le Roux et al., 1977)  
22 fall in the usual range for Northern Hemisphere Paleozoic cherts. This  
23 persistence of high  $\delta^{18}\text{O}$  values and the persistence of hemispheric differences  
24 in the oxygen isotopic compositions for detrital quartz (Fig. 3) confirm, on  
25 a long time scale, the evidence (Clayton et al., 1977) for resistance of silt

1 sized quartz to appreciable oxygen isotopic composition change in unmeta-  
2 morphosed sediments. Shales in situ appear generally not to receive an  
3 appreciable influx of quartz crystallized at low temperature.

4 During the several hundred million years of the Phanerozoic Eon, the  
5 Northern Hemisphere continents were located in tropical and equatorial  
6 latitudes (Drewry et al., 1974). The soils were subject to the usual  
7 intense desilication at these latitudes (Jackson, 1975), with an accompanying  
8 discharge of soluble  $\text{Si(OH)}_4$  into rivers and oceans where biogenic silica  
9 (Houck et al., 1975) was deposited and from which chert quartz gradually  
10 formed. As the present Northern Hemisphere continents were carried north-  
11 ward across the equator by plate tectonics (Drewry et al., 1974), during and  
12 subsequent to the Cretaceous Period, the ocean sediments were uplifted to form  
13 great continental plains with their abundance of low-temperature, high  $\delta^{18}\text{O}$   
14 chert quartz. Some land areas in the Southern Hemisphere were part  
15 of a frigid South Polar supercontinent, Gondwanaland, and associated seas  
16 in late Paleozoic time. Glacial and fluvial erosion of predominantly  
17 igneous and metamorphic rocks, with low  $\delta^{18}\text{O}$  values of quartz from the  
18 mountains of continental sutures, carried silts onto continental shelves  
19 during the late Carboniferous, Permian, and Triassic periods (le Roux  
20 et al., 1977). These continental shelves were uplifted into continents as  
21 plate motions carried the Southern Hemisphere continents northward into  
22 their present positions, along with the low  $\delta^{18}\text{O}$  quartz in their silty  
23 sediments. Of course, large portions of the sediments of these continents  
24 were formed before and after the Gonwanaland glacial periods and some areas  
25 extended into warm climates even in times of Gonwanaland's existence. The  
sediments laid down at those times and places contain quartz with oxygen  
isotopic ratios resembling Northern Hemisphere quartz (Fig. 4).

## CONCLUSIONS

1       Quartz of detrital sediments varies in oxygen isotopic ratio depending  
2       on the proportions of quartz crystallized at low temperatures (high  $\delta^{18}\text{O}$   
3       value) and at high temperatures (low  $\delta^{18}\text{O}$  value); this is a basic model or  
4       paradigm. The quartz  $\delta^{18}\text{O}$  value increases with decreasing particle size,  
5       from  $11 \pm 1\text{ ‰}$  for coarse sands to intermediate values for silts to 25 or  
6       30 ‰ for fine silt and clay size of chert ( $\sim 1-5\text{ }\mu\text{m}$  diameter); this is  
7       designated submodel I of the basic paradigm.

8       Proximity to an igneous rock erosional source decreases the  $\delta^{18}\text{O}$  value  
9       of the long range aerosol size ( $1-10\text{ }\mu\text{m}$  diameter) quartz of sediments, while  
10      proximity to silica sources crystallized in contact with natural open waters  
11      produces higher quartz  $\delta^{18}\text{O}$  values; this is designated submodel II of the  
12      basic paradigm.

13      Analyses of present day atmospheric aerosols and eolian-derived soils,  
14      Pacific pelagic sediments, and now-raised Phanerozoic marine sediments show  
15      that the Northern and Southern Hemispheres have separate large-scale reser-  
16      voirs of the fine grain sizes that contribute to aerosol dusts. These can  
17      be identified by distinctive values of  $^{18}\text{O}/^{16}\text{O}$  ratios of the quartz therein.  
18      The difference in quartz  $\delta^{18}\text{O}$  value of about  $12 \pm 2\text{ ‰}$  in Southern Hemi-  
19      sphere mixed detrital sediments and about  $19 \pm 2\text{ ‰}$  in those of the  
20      Northern Hemisphere (for constant size, the  $1-10\text{ }\mu\text{m}$  size fraction) results  
21      from the presence of a considerably larger proportion of quartz having low-  
22      temperature origin and higher  $\delta^{18}\text{O}$  values (chert, silica overgrowths, etc.)  
23      in the Northern Hemisphere reservoirs. This  $\delta^{18}\text{O}$  difference in aerosol sized  
24      quartz geochemistry between the two Hemispheres, attributable to paleographic-  
25      paleogeomorphic-paleoclimatic (polar-equatorial) differences associated with  
      continental drift, is designated submodel III of the basic paradigm.

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Table 1 -- Mean oxygen isotopic ratios of aerosol-sized quartz isolated from dusts, soils and sediments

Area	Kind of soil or sediment	Quartz size range (μm)	Number of samples (mean of)	Mean $\delta^{18}\text{O}$ (%)	Reference
Mid-USA	Shales "	1-10 ws*	30 31	20.7±1.8 19.1±2.2	Churchman <i>et al.</i> , 1976
SW. USA	Soils	1-10	13	19.6±0.3	Sridhar <i>et al.</i> , 1977
		1-10	18	19.0±0.2	" " "
	Dusts	1-10	21	18.4±0.8	Gillette <i>et al.</i> , 1977
E. Cent. TX, AZ	Soils	1-10	5	17.7±0.6	Sridhar <i>et al.</i> , 1977
Hawaii (4 Islands) <sup>†</sup>	Soils	1-10 ws	5 25	18.3±0.2 17.6±0.5	Sridhar <i>et al.</i> , 1977
N. Pacific	Pelagic	ws <sup>§</sup>	8	17.5±0.5	Jackson <i>et al.</i> , 1971
S. Pacific	Pelagic	2-5	7	15.1±1.5 <sup>#</sup>	Clayton <i>et al.</i> , 1972
Australia, Victoria	Loess in basaltic soils	2-5 5-20	10 11	14.9±0.5 14.5±0.2	Mokma <i>et al.</i> , 1972
SW. USA	Soils	ws	8	14.7±0.9	" " "
New Zealand					Sridhar <i>et al.</i> , 1977
N. Island	In till	5-20	5	13.6±1.0	" " "
S. Island	" "	5-20	7	13.2±0.2	" " "
S. Island	" "	2-5	7	13.3±0.1	" " "
S. Island	Glacial dust bands	ws	3	13.5±0.4	" " "
S. Africa	Soils and sediments	1-10	28	12.2±2.1	Le Roux <i>et al.</i> , 1977

\* Whole quartz from samples.

<sup>†</sup> Including for 1-10 μm two lake sediments from on top of Mauna Kea, and excluding hydrothermal area for ws.

<sup>§</sup> Weighted average calculated from size fractions, excluding Gulf of Alaska.

<sup>#</sup> 9° 13' to 58° 26' S. latitude.

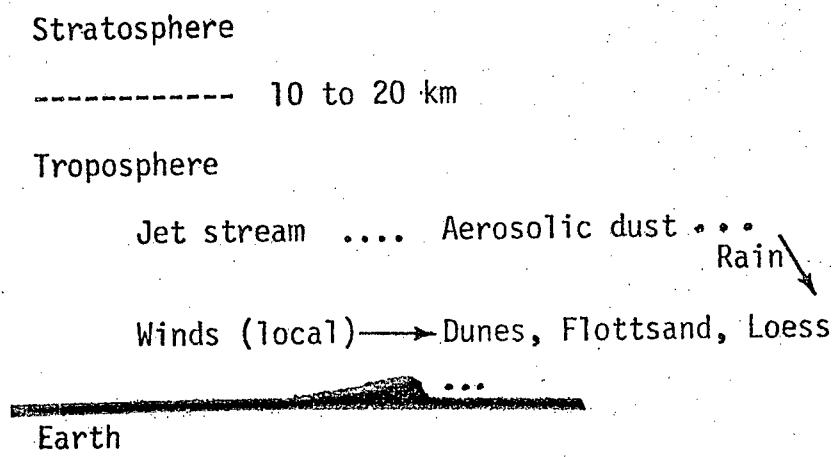


Fig. 1. Relationship of wind-blown soil particles, local dunes (~ 1 mm particles), flottsand, loess (5 to 70  $\mu\text{m}$  particles), and long-range aerosolic dust (1-10  $\mu\text{m}$  particles) carried in high winds and brought down in rain.

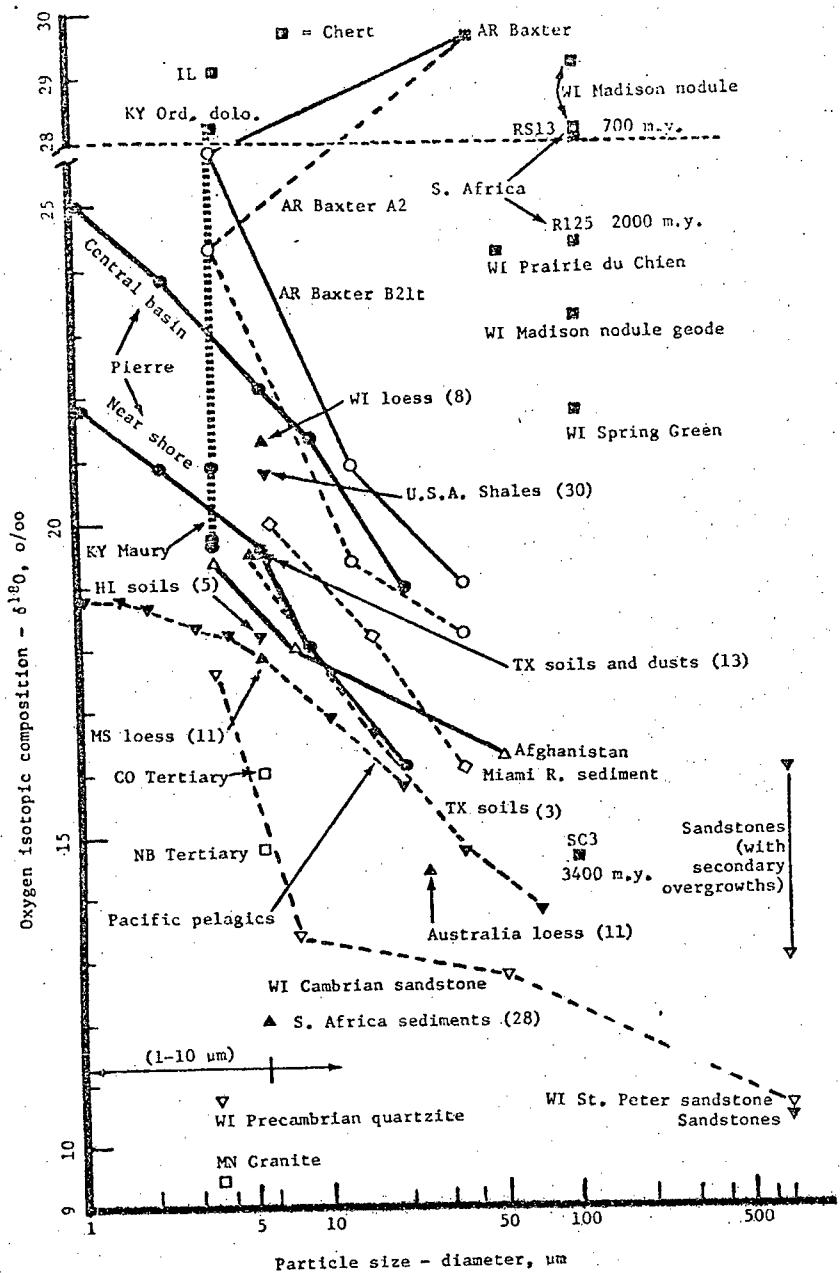


Fig. 2. Variation in quartz oxygen isotopic composition with particle size and provenance of detrital sediments. Chert and high temperature rock and Maury and Baxter soil data from Syers et al. (1969), Savin and Epstein (1970), Sayin and Jackson (1975), and Sridhar et al. (1975); North Pacific pelagic data from Clayton et al. (1972); Pierre shale data from Sridhar et al. (1975); shale mean and Tertiary data from Churchman et al. (1976); and Texas dust data from Gillette et al. (1977) and Sridhar et al. (1977). Numbers in parentheses indicate number of samples averaged.

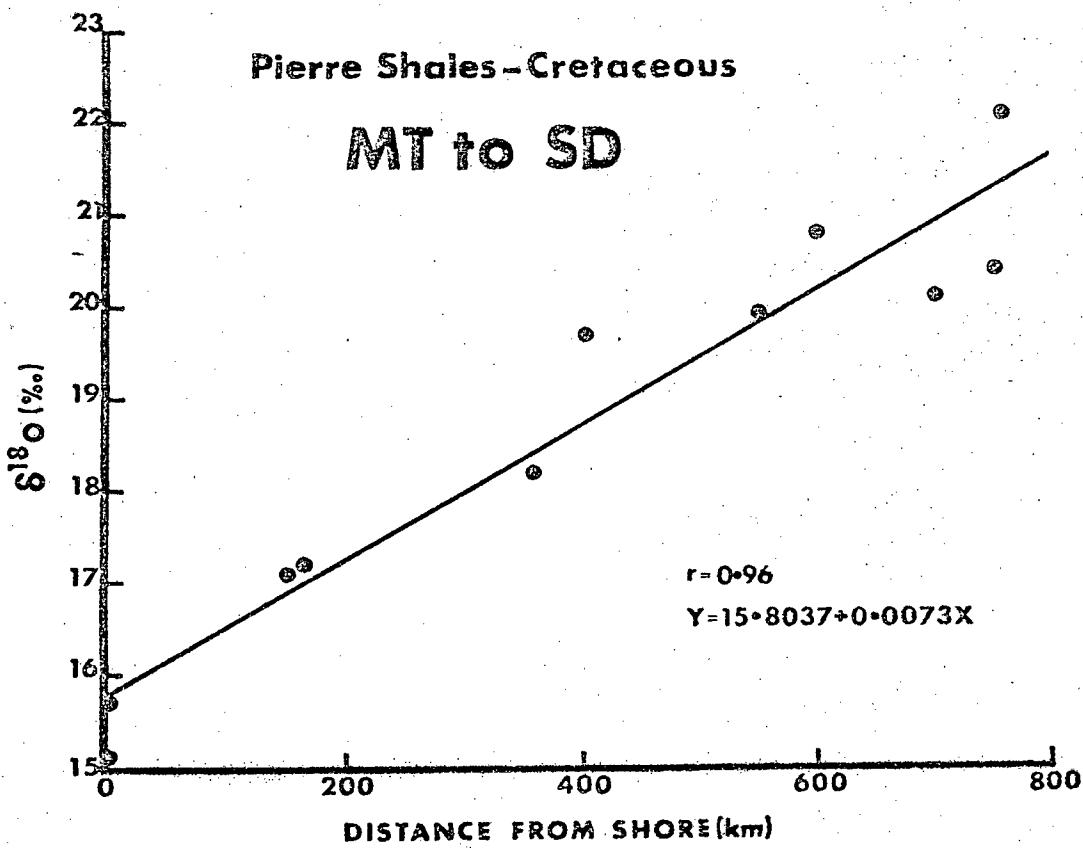


Fig. 3. Variation in quartz oxygen isotopic composition ( $\delta^{18}\text{O}$ ) of the Pierre shale and related rocks with distance from mountainous shore of the Cretaceous Sea from Montana (MT) to South Dakota (SD). Numerical data from Churchman et al. (1976), for quartz from whole shales.

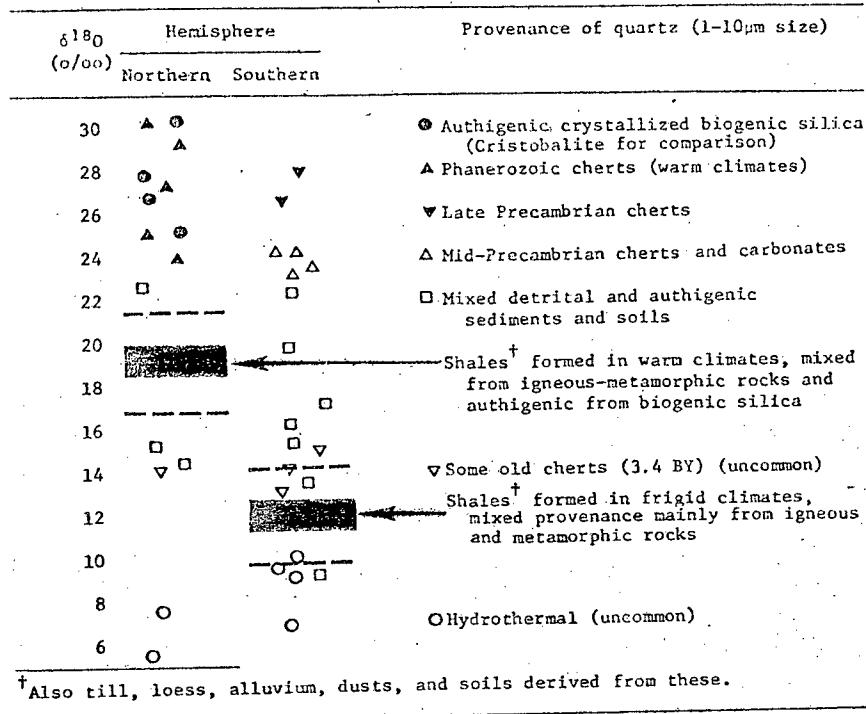


Fig. 4. Divergence in Northern and Southern Hemisphere quartz oxygen isotopic ratios ( $\delta^{18}\text{O}$ ) of long range aerosol size (1-10  $\mu\text{m}$ ) particles. — = sigma. Northern Hemisphere data are from Jackson et al. (Hawaiian highland eolian caps, 1968, 1971), Clayton et al. (Pacific pelagic sediments, 1968, 1972), Rex et al. (world wide eolian circulation, 1969), Syers et al. (southeast Asia and tropical North Atlantic dusts, 1969), Henderson et al. (cristobalite, 1972), and Churchman et al. (Central U.S.A. shales, 1976). Southern Hemisphere data are mainly from Clayton et al. (Pacific pelagic sediments, 1972), Jackson et al. (Australian soils and dusts, 1972), Mokma et al. (New Zealand soils and dusts, 1972), Henderson et al. (a few hydrothermal quartz values, 1972), and le Roux et al. (southern Africa soils, sediments and cherts, 1977).