

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

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

The $\delta^{18}\text{O}$ value ($^{18}\text{O}/^{16}\text{O}$ relative to standard sea water, expressed as parts per thousand, per mil, $^{\circ}/\text{oo}$) of quartz, once established in a rock, is very resistant to change at temperatures below 350°C . A selectivity of quartz origin in detrital sediments evidently occurs, with high temperature quartz preferentially remaining in beach sands and attenuating with fineness, while fine quartz from chert and chipped off overgrowths increases in the fine silt (1-10 μm diameter) range (submodel I of the paradigm). Close proximity to eroding igneous-metamorphic mountain masses, however, results in $\delta^{18}\text{O}$ values of fine silt quartz in the range of 12 to 16 $^{\circ}/\text{oo}$. (Quartz of beach sand has $\delta^{18}\text{O} = 11 \pm 1$ $^{\circ}/\text{oo}$). In contrast, central marine basins, 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 ± 2 ‰) of fine silt quartz in the large-scale
7 silt reservoirs of the Northern Hemisphere continents, differing from that
8 (mainly 12 ± 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 paleochemical differences remain the control of the North-South Hemi-
sphere difference in $\delta^{18}\text{O}$ values in long-range aerosol sized quartz.

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

*"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 $<10\text{ }\mu\text{m}$ equivalent diameter (Jackson et al., 1971; Walker and Costin, 1971). Loess, in contrast, has a wide range of particle sizes, up to $70\text{ }\mu\text{m}$ (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
17 generally (Ruhe, 1969; Smith, 1942), with aerosol dust added (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

Insert
Fig. 1

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}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 500×10^6
14 tons 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, 2×10^{11}
19 kg yr^{-1} compared to $93 \times 10^{11} \text{ kg yr}^{-1}$ carried in the fluvial load from
20 land to sea, but is double the present/estimated load carried by glacial ice, 1×10^{11}
21 kg yr^{-1} (Judson, 1968; Jackson, 1974a).
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METHODS

Isolation of quartz

A 2- to 5-g sample of soil, dust, or other sediment is treated with 6N HCl at 100°C to destroy carbonates and some clay and then with 30% H₂O₂ to destroy readily oxidizable organic matter. The desired size fraction (often 1-10 µm in diameter) is then obtained by sieving, gravity sedimentation, and centrifugation (Jackson, 1974b). Fractions still showing black color (some shales) are boiled in a glass beaker for 2 minutes in 5 ml of concentrated HNO₃ (with drops of concentrated H₂SO₄ present) and then 2 ml of 60% HClO₄ is added and the suspension is heated to fumes for a few minutes (hood) thereby removing the carbon and leaving a white residue; evolution of CO₂ in the subsequent Na₂S₂O₇ fusion is thus avoided (Jackson, 1974b). Monomineralic quartz is then obtained by fusion of a 0.2 or 0.3 g sample in 12 g of Na₂S₂O₇ in a silica crucible, slowly at first and finally until only a slightly moist salt cake remains. After the crucible and contents are cooled, the cake is dissolved in 50 ml of, and the residue washed with, 3N HCl. These treatments do not affect the oxygen isotopic composition of quartz (Sridhar et al., 1975). Finally the residue is treated with 10 ml of 30% H₂SiF₆ reagent (Syers et al., 1968) modified by the use of quartz flour sized at 1-100 µm to quench the commercial H₂SiF₆ and thus prevent its attack on the fine quartz being isolated (Jackson, 1974b; Jackson et al., 1976). The suspension is allowed to stand 3 days at 18±1°C, being mixed twice daily during this period. The sample is rinsed momentarily with 0.1N HF and then washed with H₂O. A small subsample 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 interstices 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° , 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 $\delta^{18}\text{O}$ values, in parts per thousand (per mil,
15 $^\circ/\text{oo}$), according to the equation,

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

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

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

OXYGEN ISOTOPIC COMPOSITION OF QUARTZ

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

	$\delta^{18}\text{O}$ of quartz ($^{\circ}/\text{oo}$)
(a) From rocks of high temperature origin	
igneous rocks	8-10
metamorphic rocks	10-16
hydrothermal spring altered rocks	4-10
(b) From rocks of relatively low temperature origin	
dolomites and chert (40-10 $^{\circ}\text{C}$)	24-33
chert (80-40 $^{\circ}\text{C}$)	14-24

The igneous and metamorphic quartz values are from Taylor and Epstein (1962) and Savin and Epstein (1970). The range for quartz from rocks of relatively low temperature is from Degens and Epstein (1962), Knauth and Epstein (1976) and Kolodny and Epstein (1976). The hydrothermal quartz values are from Clayton and Steiner (1975; 3.9 $^{\circ}/\text{oo}$), Jackson et al. (1971; 5.5-8.3 $^{\circ}/\text{oo}$), and Henderson et al. (1972; 9.6 to 10.0 $^{\circ}/\text{oo}$).

Variation with particle size in detrital sediments

The $\delta^{18}\text{O}$ value of quartz, once established in a rock, is very resistant to change at temperatures below 350 $^{\circ}\text{C}$ (Clayton et al., 1968). The chert field (upper part, Fig. 2) is mainly in the range of $\delta^{18}\text{O}$ of 22 to 30 $^{\circ}/\text{oo}$, corresponding to 10 to 40 $^{\circ}\text{C}$ formation temperatures as crystallization (Knauth and Epstein, 1976) occurred at various stages of shallow burial. The South African chert SC3 with $\delta^{18}\text{O}$ of 14.6 $^{\circ}/\text{oo}$ shows field, morphological 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 ($\delta^{18}\text{O} = 11 \pm 1 \text{ }^{\circ}/\text{oo}$) and sand grains of sandstones without secondary overgrowths; however, overgrowths of chemically precipitated quartz extend the range to $\delta^{18}\text{O} = 16 \text{ }^{\circ}/\text{oo}$ (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 μm 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 μm 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 (Savin and Jackson, 1975). Occurrence as aggregates of fine grain size permit chert particles to extend up into the 50- μm diameter and larger sizes (AR Baxter, Fig. 2; Savin 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 (Syers 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.

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 μm 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 ± 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 μm dusts, Miami R. of Ohio and Afganistan 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.

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 μm 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.
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1 Variation with latitude of earth

2 Northern Hemisphere. An impressively narrow range of $\delta^{18}\text{O}$ values
3 (17 to 21 ‰) for the long range aerosol size quartz particles of dusts
4 (Table 1) and sediments/has been noted for North Pacific (Clayton et al., 1968, 1972;
5 Jackson et al., 1968, 1971; Rex et al., 1969), Central North America
6 (Churchman et al., 1976), and South East Asian areas (Syers et al., 1969).
7 We have found similar values for shales of Austria, loess in the central
8 part of the Peoples Republic of China, and an eolian cap on a volcanic cone
9 off the coast of Korea (unpublished data). Saharan dusts crossing the
10 tropical North Atlantic have been collected on Barbados, Cape Verde
11 Islands, and the Canary Islands and the $\delta^{18}\text{O}$ values (unpublished data) for
12 the 1-10 μm quartz fraction also fall in the broad black bar for the
13 Northern Hemisphere in Fig. 4. A scatter of values through the broad $\delta^{18}\text{O}$
14 spectrum is found for hydrothermal and chert systems, each representing
15 more homogeneous temperature and geomorphological mixing processes.

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

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 ± 0.2 to 13.5 ± 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 μm 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 ± 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 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.

15 Some land areas in the Southern Hemisphere were part
16 of a frigid South Polar supercontinent, Gondwanaland, and associated seas
17 in late Paleozoic time. Glacial and fluvial erosion of predominantly
18 igneous and metamorphic rocks, with low $\delta^{18}\text{O}$ values of quartz from the
19 mountains of continental sutures, carried silts onto continental shelves
20 during the late Carboniferous, Permian, and Triassic periods (le Roux
21 et al., 1977). These continental shelves were uplifted into continents as
22 plate motions carried the Southern Hemisphere continents northward into
23 their present positions, along with the low $\delta^{18}\text{O}$ quartz in their silty
24 sediments. Of course, large portions of the sediments of these continents
25 were formed before and after the Gonwanaland glacial periods and some areas
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 ± 1 ‰ for coarse sands to intermediate values for silts to 25 or
6 30 ‰ for fine silt and clay size of chert ($\sim 1\text{--}5$ μm 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 ($1\text{--}10$ μm 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 ± 2 ‰ in Southern Hemi-
19 sphere mixed detrital sediments and about 19 ± 2 ‰ in those of the
20 Northern Hemisphere (for constant size, the $1\text{--}10$ μm 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	30	20.7 \pm 1.8	Churchman <u>et al.</u> , 1976
	"	ws*	31	19.1 \pm 2.2	" " " "
SW. USA	Soils	1-10	13	19.6 \pm 0.3	Sridhar <u>et al.</u> , 1977
		1-10	18	19.0 \pm 0.2	" " " "
	Dusts	1-10	21	18.4 \pm 0.8	Gillette <u>et al.</u> , 1977
E. Cent. TX, AZ	Soils	1-10	5	17.7 \pm 0.6	Sridhar <u>et al.</u> , 1977
Hawaii (4 Islands) [†]	Soils	1-10	5	18.3 \pm 0.2	Sridhar <u>et al.</u> , 1977
		ws	25	17.6 \pm 0.5	Jackson <u>et al.</u> , 1971
N. Pacific	Pelagic	ws [§]	8	17.5 \pm 0.5	Clayton <u>et al.</u> , 1972
S. Pacific	Pelagic	2-5	7	15.1 \pm 1.5 [#]	" " " "
Australia, Victoria	Loess in basaltic soils	2-5	10	14.9 \pm 0.5	Mokma <u>et al.</u> , 1972
		5-20	11	14.5 \pm 0.2	" " " "
SW. USA	Soils	ws	8	14.7 \pm 0.9	Sridhar <u>et al.</u> , 1977
New Zealand					
N. Island	In till	5-20	5	13.6 \pm 1.0	Mokma <u>et al.</u> , 1972
S. Island	" "	5-20	7	13.2 \pm 0.2	" " " "
S. Island	" "	2-5	7	13.3 \pm 0.1	" " " "
S. Island	Glacial dust bands	ws	3	13.5 \pm 0.4	" " " "
S. Africa	Soils and sediments	1-10	28	12.2 \pm 2.1	le Roux <u>et al.</u> , 1977

* Whole quartz from samples.

[†] Including for 1-10 μm two lake sediments from on top of Mauna Kea, and excluding hydrothermal area for ws.

[§] Weighted average calculated from size fractions, excluding Gulf of Alaska.

[#] 9° 13' to 58° 26' S. latitude.

Stratosphere

----- 10 to 20 km

Troposphere

Jet stream Aerosolic dust ...

Rain ↘

Winds (local) → Dunes, Flottsand, Loess

Earth

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

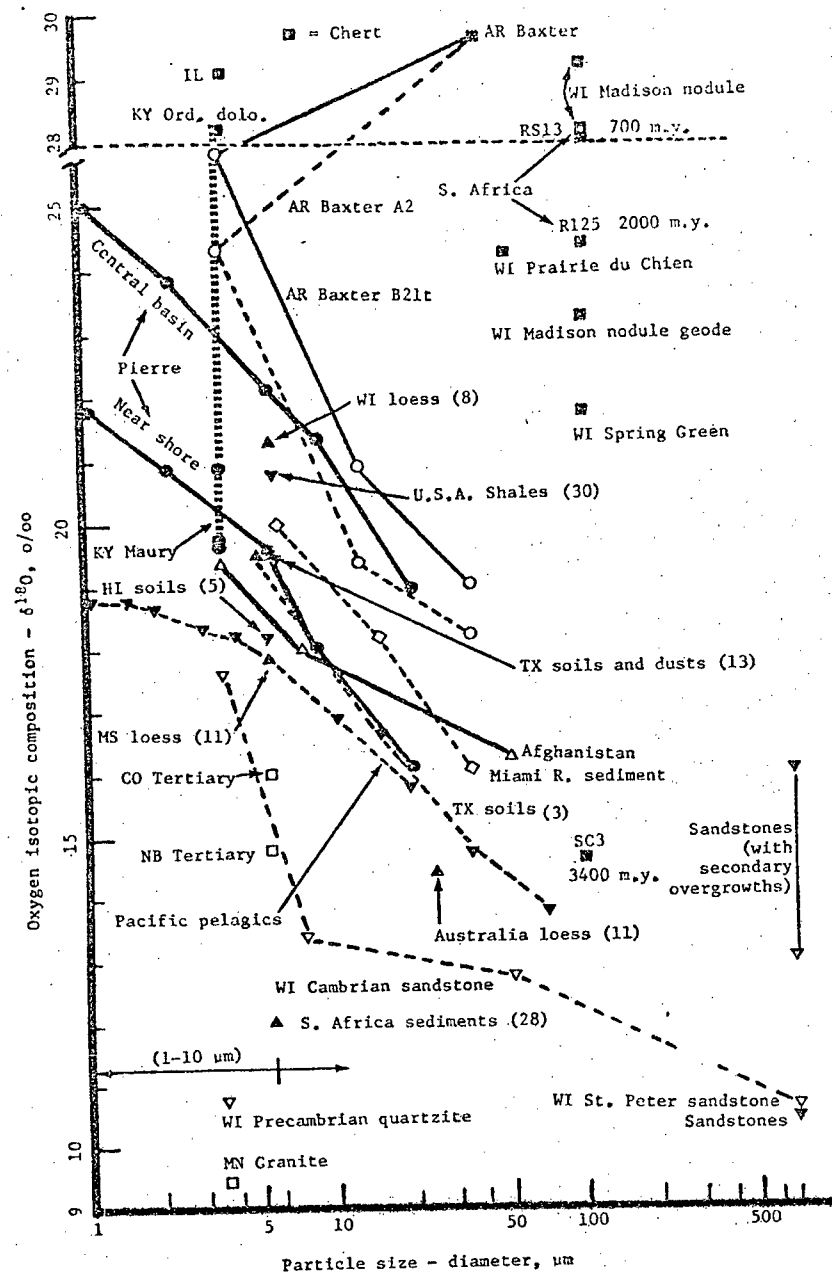


Fig. 2. Variation in quartz oxygen isotopic composition with particle size and provenance of detrital sediments. Chert and high temperature rock and Maury and Baxter soil data from Syers et al. (1969), Savin and Epstein (1970), Sayin and Jackson (1975), and Sridhar et al. (1975); North Pacific pelagic 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.

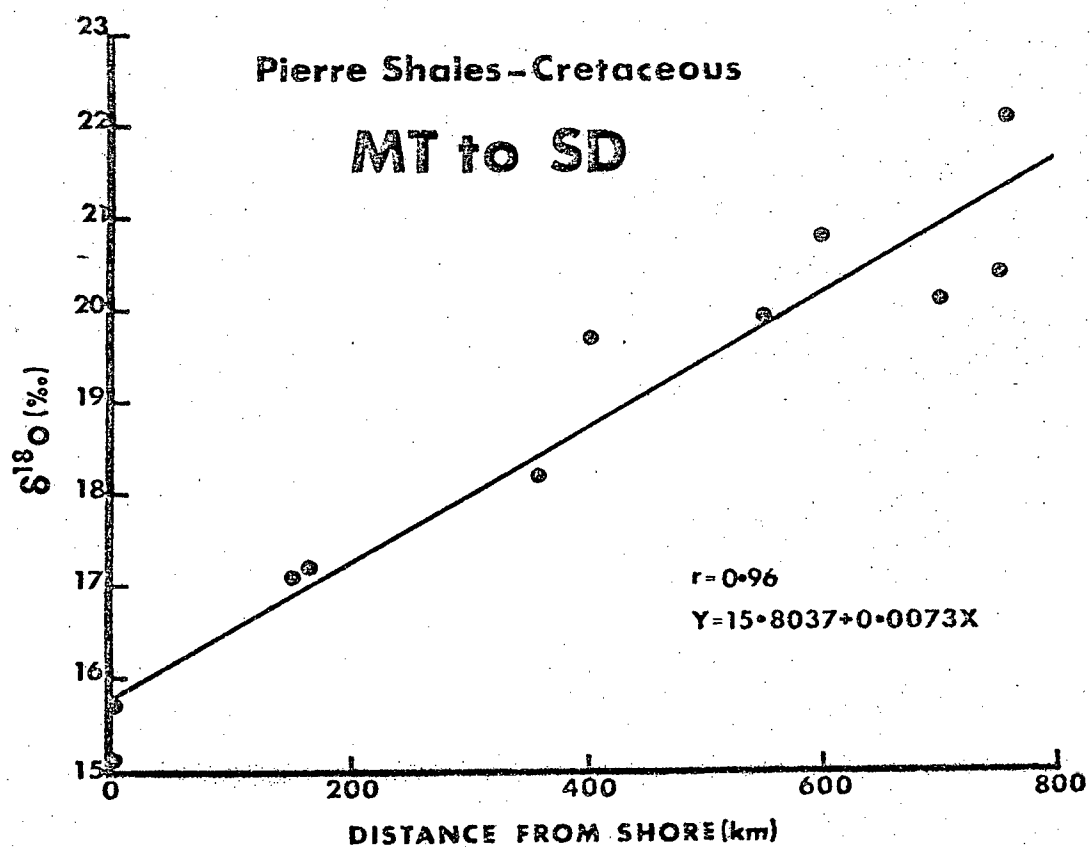


Fig. 3. Variation in quartz oxygen isotopic composition ($\delta^{18}O$) of the Pierre shale and related rocks with distance from mountainous shore of the Cretaceous Sea from Montana (MT) to South Dakota (SD). Numerical data from Churchman et al. (1976), for quartz from whole shales.

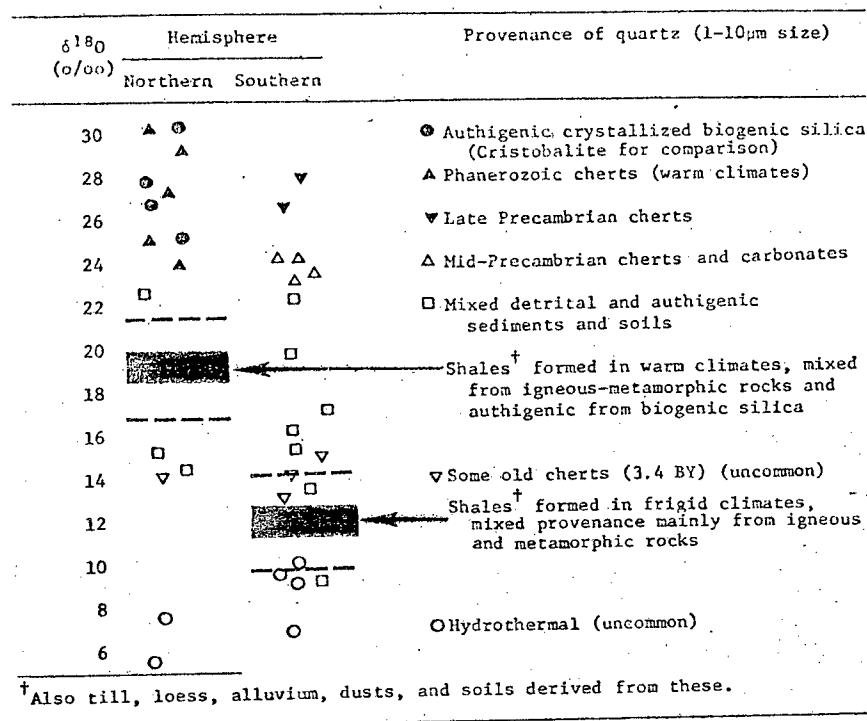


Fig. 4. Divergence in Northern and Southern Hemisphere quartz oxygen isotopic ratios ($\delta^{18}O$) of long range aerosol size (1-10 μ m) particles. ---- = sigma. Northern Hemisphere data are from Jackson et al. (Hawaiian highland eolian caps, 1968, 1971), Clayton et al. (Pacific pelagic sediments, 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).