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    GEOCHEMISTRY

A GRAPHIC PROCEDURE IN THE GEOCHEMICAL
INTERPRETATION OF WATER ANALYSES

By
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      This paper describes a graphical method of correlating
water analyses which has proved extremely useful in some
ground-water investigations. The first version of this paper
was duplicated by the Ground Water Branch under the date of
October 5, 1942. Subsequently a second version was published
in the Transactions of the American Geophysical Union, part 6,
pp. 914-923, 1944. Both versions contain material that is now
obselete, and Mr. Piper has therefore made appropriate modifi-
cations to bring the paper up to date. The revised paper is
presented herewith.

      General considerations--This paper outlines certain fundamental
principles in a graphic procedure which appears to be an effective tool
in segregating analytical data for critical study with respect to sources
of the dissolved constituents in waters, modifications in the character
of a water as it passes through an area, and related geochemical problems.
The procedure is based on a multiple-trilinear diagram (Fig. 1) whose
form has been evolved gradually and independently by the writer during
the past several years through trial and modification of less comprehen-
sive antecedent forms. Neither the diagram nor the procedure here des-
cribed is a panacea for the easy solution of all geochemical problems.
Many problems of interpretation can be answered only by intensive study
of critical analytical data by other methods.

      Most natural waters contain relatively few dissolved constitu-
ents, with cations (metals or bases) and anions (acid radicles) in chemi-
cal equilibrium with one another; commonly the waters contain some silicon,
iron, and aluminum but these constituents are usually assumed to occur in
the colloid state as oxides and not to be in chemical equilibrium with the
ionized constituents. Ordinarily the most abundant cation constituents

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PROPERTIES

WATER-ANALYSIS DIAGRAM

Area of circle indicates concentration in parts per million, thus:
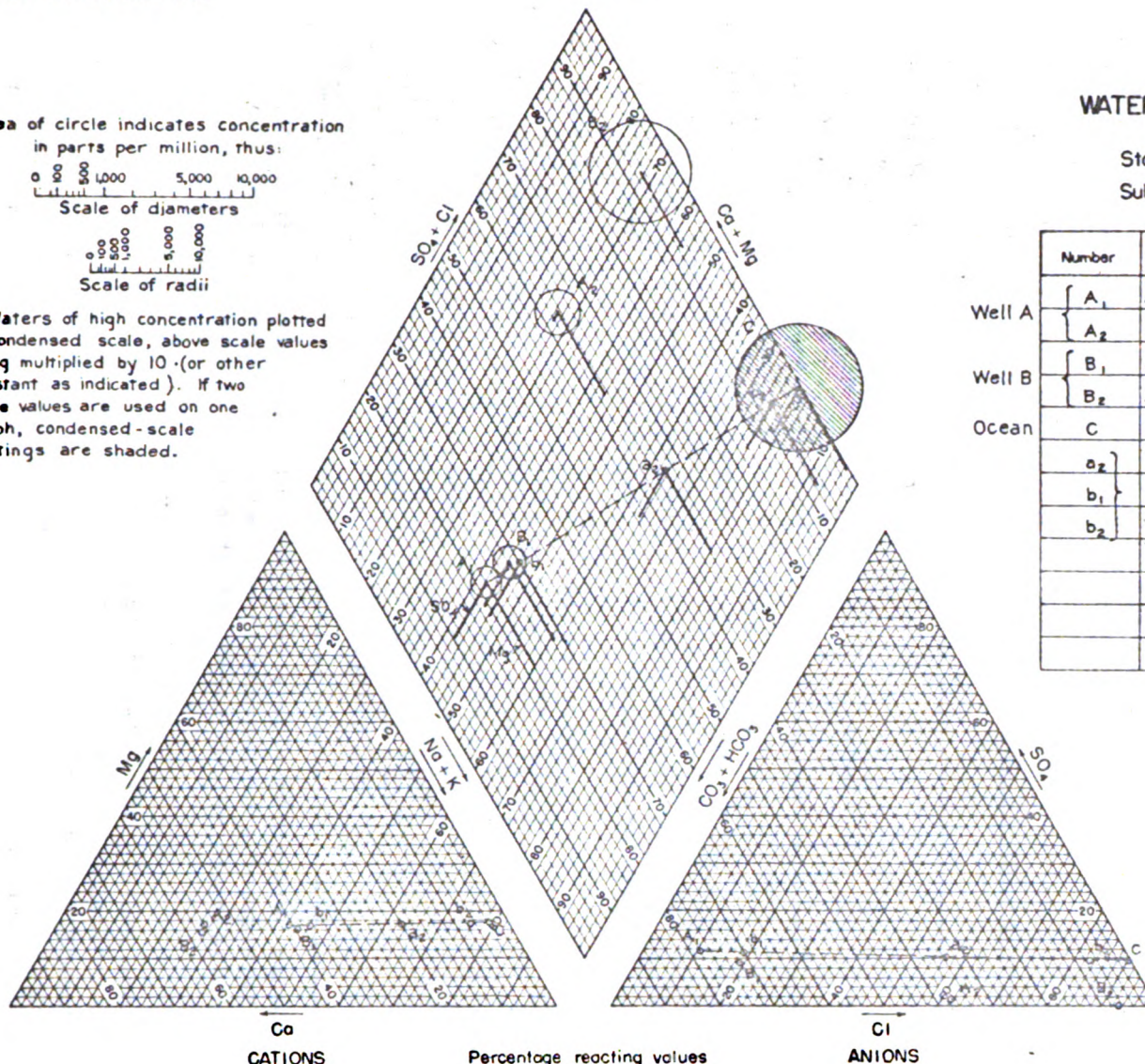
0   1,000   5,000   10,000
Scale of diameters

0   1,000   5,000   10,000
Scale of radii

Waters of high concentration plotted at condensed scale, above scale values being multiplied by 10 (or other constant as indicated). If two scale values are used on one graph, condensed-scale plottings are shaded.

State_____

Subarea_____

	Number	Date	Parts per million	Depth or perforations
Well A	A_1	June 3, 1931	240	
	A_2	Dec. 20, 1940	566	
Well B	B_1	June 6, 1931	260	
	B_2	June 30, 1936	2,230	
Ocean	C	May 18, 1941	34,100	
	a_2			
	b_1	Hypothetical mixtures; see		
	b_2	text and table 2.		

$SO_4 + Cl$

$Ca + Mg$

$Na + K$

$CO_3 + HCO_3$

Mg

SO_4

Ca
CATIONS

Percentage reacting values

Cl
ANIONS

Plotted by_____

Checked by_____

Date_____

GPO 06-083910

FIG. I--WATER-ANALYSIS DIAGRAM

are two "alkaline earths", calcium (Ca) and magnesium (Mg), and also one "alkali", sodium (Na). Potassium (K) also occurs commonly, but ordinarily is much less abundant than sodium. Still other cation-constituents occur in appreciable quantities in highly concentrated natural waters and in some waters of unusual composition. For the graphic methods treated in this paper all these less abundant constituents are summed with the major three constituents to which they are respectively related in chemical properties, as indicated by the two ranks of entry in Table I. The most common anion-constituents are one "weak acid"; bicarbonate (HCO_3); also two "strong acids", sulfate (SO_4) and chloride (Cl). Less common anion-constituents are listed in Table I; for plotting, these are summed with the major three anions to which they are respectively related. Thus, for much of the graphic methods here described, a natural water is treated substantially as though it contained only three cation-constituents and three anion-constituents.

Table I--Common and minor constituents of natural waters

Cations	Reciprocal of combining weight	Anions	Reciprocal of combining weight
Alkaline earths		Weak acids	
Calcium (Ca^{++})	0.04990	Bicarbonate (HCO_3^-)	0.01639
Barium (Ba^{++})	0.01456	Carbonate ($CO_3^=$)	0.03333
Strontium (Sr^{++})	0.02282	Tetraborate ($B_4O_7^=$)	0.01288
Magnesium (Mg^{++})	0.08224	Orthophosphate ($PO_4^{\equiv}$)	0.03157
Alkalies		Strong acids	
Sodium (Na^+)	0.04348	Sulfate ($SO_4^=$)	0.02082
Potassium (K^+)	0.02558	Chloride (Cl^-)	0.02820
Caesium (Cs^+)	0.00752	Iodide (I^-)	0.00788
Rubidium (Rb^+)	0.01170	Bromide (Br^-)	0.01251
Lithium (Li^+)	0.14409	Fluoride (F^-)	0.05263
Ammonium (NH_4^+)	0.05543	Nitrate (NO_3^-)	0.01613
		Nitrite (NO_2^-)	0.02174

Notes: Of the second-rank constituents only potassium, carbonate, fluoride, and nitrate are commonly determined in a "complete" analysis. Reciprocals of combining weights are based on the international atomic weights of 1938.

In substantially all natural waters the cations are in chemical equilibrium with the anions. Accordingly, if the concentrations of the several dissolved constituents are measured in terms of "reacting value" or "equivalents per million" the totals of cations and anions are necessarily equal in principle. A difference between these totals is then a measure of analytical error. [The concentration of a constituent in equivalents per million (milligram equivalents per kilogram) is computed by multiplying its concentration in parts per million by the reciprocal of its combining weight.]

Thus, to the extent that a natural water can be treated in terms of three cation-variables and three anion-variables, (I) the relative concentrations of constituents can be expressed conveniently as a percentage of total reacting value, and (2) the essential chemical character of the water can be indicated graphically by single-point plottings of cations and anions on trilinear coordinates. For convenience, the subtotal of all cation reacting values is taken as the ICO-percent base for computing percentage reacting values of the several cation-variables; likewise for the several anion-variables. This procedure balances analytical errors automatically. Such is the basis of the diagram herein described.

Methods of plotting

The diagram herein described combines three distinct fields for plotting--two triangular fields at the lower left and lower right, respectively, and an intervening diamond-shaped field. All three fields have scales reading in 100 parts (see Figs. I and 2-A). In the triangular field at the lower left, the percentage reacting values of the three cation-groups (Ca, Mg, Na) are plotted as a single point according to conventional trilinear coordinates. The three anion-groups (HCO_3, SO_4, Cl) are plotted likewise in the triangular field at the lower right. Thus, two points on the diagram--one in each of the two triangular fields-- indicate the relative concentrations of the several dissolved constituents of a natural water.

The central diamond-shaped field is used to show the over-all chemical character of the water by a third single-point plotting, which is at the intersection of rays projected from the plottings of cations and anions as indicated on Figure 2-A. Using the scales of Figure I, the position of this plotting indicates the relative composition of a water in terms of the cation-anion pairs that correspond to the four vertices of the field. This central-field plotting can also be taken directly from the analytical data according to the vectors shown along the outer margins of the field on Figure 2-A. For such plotting only one cation-variable and one anion-variable need be used--either alkaline earths or alkalies with either weak acids or strong acids; the two percentage reacting values selected from the analytical data are doubled to suit the numerical scales of Figure I.

The three trilinear plottings just described will show the essential chemical character of a water according to the relative concentration of its constituents, but not according to the absolute concentrations. Because the absolute concentrations commonly are decisive in many problems of interpretation, it is convenient to indicate the plotting in the central field by a circle whose area is proportional to the absolute concentration of the water. Figure I shows such plottings for several dissimilar waters.

A very few natural waters contain free acid in substantial quantity--that is, hydrogen is present as a cation. The chemical character of such waters cannot be fully represented on the diagram.
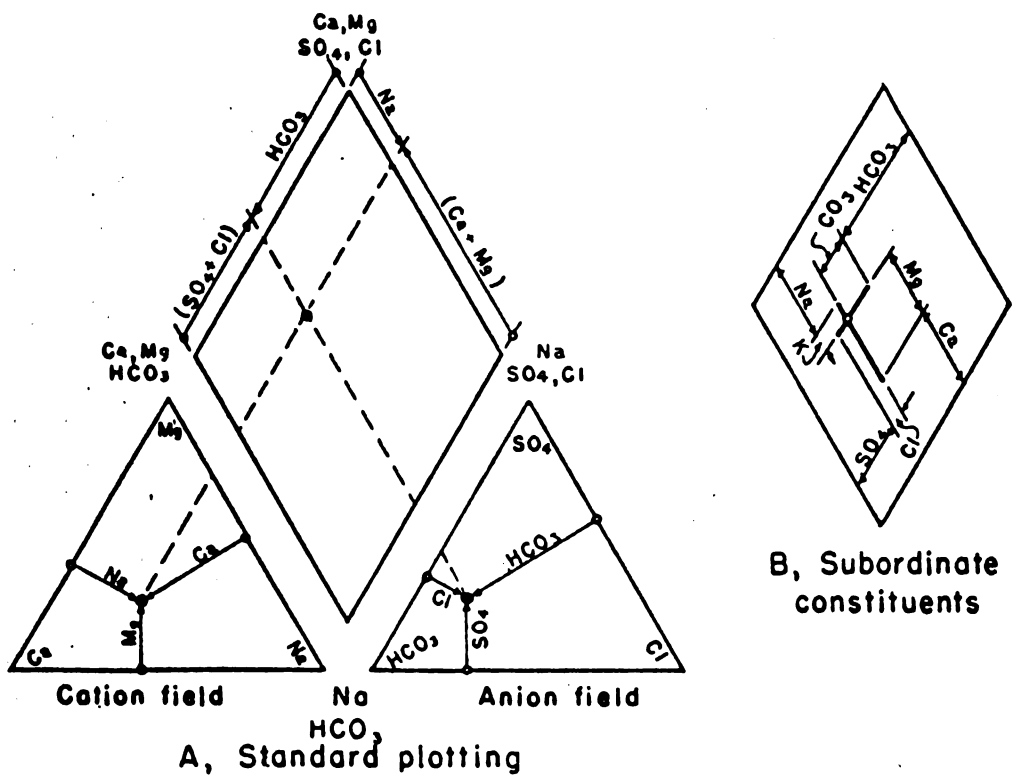
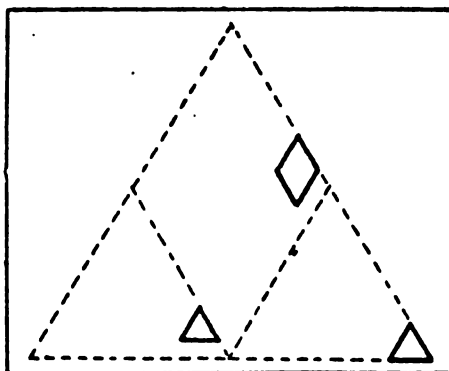**FIG. 2 -- PLOTTING KEY FOR WATER-ANALYSIS DIAGRAM**

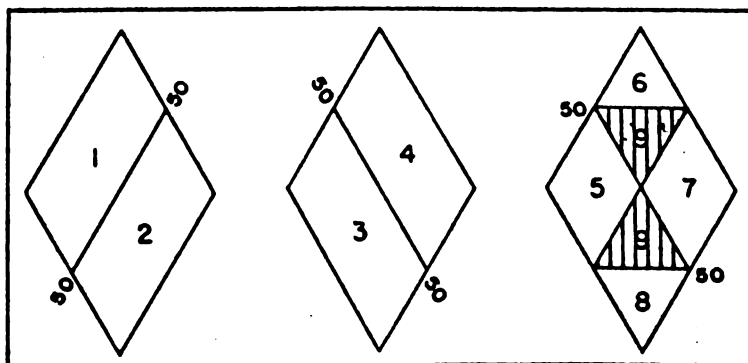**FIG. 3 -- SCHEME OF PLOTTING BY EXTENDED SCALES**

FIG. 4 -- SUBDIVISIONS OF THE DIAMOND-SHAPED FIELD

Obviously, the plottings in the diamond-shaped field do not bring out critical ratios between certain constituents, such as the ratios between sodium and potassium, calcium and magnesium, carbonate and bicarbonate, or sulfate and chloride. When pertinent, these ratios can be indicated graphically by vectors that indicate four of the common eight constituents, as shown by Figure 2-B.

With respect to the sources of dissolved constituents, or to progressive changes in chemical character within a particular area, many problems involve waters which differ only slightly in character or in which significant differences are masked by some common but preponderant constituents. Under such circumstances, the small differences in character can be emphasized by using the full diagram to represent proportionate subdivisions of the standard three plotting fields, and plotting the constituents with extended scales. For example, among brines similar to ocean water the minor constituents can be differentiated by plotting at five times the standard scale and using the full diagram to represent the small areas outlined in Figure 3. If desired, minor or accessory constituents can be so emphasized by vectors in the diamond-shaped field, as described in the preceding paragraph.

Differentiation of water-types

Certain distinct types can be quickly discriminated by their plottings in certain subareas of the diamond-shaped field, as indicated by Figure 4 and the following explanation: Area 1, alkaline earths exceed alkalies; Area 2, alkalies exceed alkaline earths; Area 3, weak acids exceed strong acids; Area 4, strong acids exceed weak acids; Area 5, carbonate hardness ("secondary alkalinity" after Palmer) exceeds 50 percent--that is, chemical properties of the water are dominated by alkaline earths and weak acids; Area 6, noncarbonate hardness ("secondary salinity") exceeds 50 percent; Area 7, noncarbonate alkali ("primary salinity") exceeds 50 percent--that is, chemical properties are dominated by alkalies and strong acids--ocean water and many brines plot in this area, near its right hand vertex; Area 8, carbonate alkali ("primary alkalinity") exceeds 50 percent--here plot the waters which are inordinately soft in proportion to their content of dissolved solids; Area 9, no one cation-anion pair exceeds 50 percent.

These subareas might serve as a basis for numerical or other symbols to designate specific classes, types, and subtypes of water. Symbols for this purpose have been introduced by Palmer (1911, pp. 13, 14) and Hill (1940, p. 47); however the writer feels that inflexible classifications of this sort tend to confuse by over-emphasizing differences in composition that may not be significant to the problem under consideration.

To serve most needs for classifying waters by types the writer proposes--in lieu of symbols such as those introduced by Palmer, Hill and others--to designate a water by a binomial symbol written in the form of a decimal fraction, whose two terms are (1) the percentage of hardness-causing constituents among the cations and (2) the percentage of bicar-

bonate (and carbonate, if present) among the anions. For example, the symbol 64.80 would indicate a water in which the hardness-causing constituents (Ca + Mg) amount to 64 per cent of all the cations, in terms of reacting values (equivalents); also in which the weak anions (CO_3 + HCO_3) amount to 80 percent of all the anions in like terms. The two terms of this symbol can be taken directly from analytical data in terms of percentage reacting value, or can be scaled from the diagram. This form of symbol has the distinct advantage of indicating the general character of a water specifically, without the disadvantage of implying that two waters have distinctly different characters merely because their analyses plot on either side of a boundary between arbitrary subdivisions of any water-analysis diagram. For treatment in a text, waters can be grouped according to limiting values for the two terms of the symbol here proposed, and those limits can be varied at will to suit the discussion of the problem at hand.

This decimal-fraction symbol indicates numerous characteristics of a water simply but specifically. Thus, the more common type of natural water contains chiefly calcium, magnesium, and bicarbonate; its symbol approaches 100.100 as a limit. For its fairly common opposite, the alkali-carbonate water, the symbol approaches 0.100 as a limit. The first term of the symbol indicates relative hardness in percentage of total equivalents. If the second term exceeds the first, all the hardness is carbonate or "temporary" hardness. However, if the second term is smaller, some of the hardness is noncarbonate or "permanent" and the relative amount of noncarbonate hardness is indicated by the numerical difference between the two terms. The first term of the symbol is the percentage complement of the "per cent sodium" introduced by Scofield (1933, pp. 22-23) to measure the effect of a water on the physical properties of a soil when applied for irrigation. Thus, if this term is greater than about 50 the physical condition of the soil is not likely to be impaired seriously, but if the term is less than about 40 such impairment may result.

Mixtures of waters

Many hydrologic problems involve apparent mixtures of natural waters, which the investigator seeks to confirm or disprove. The solution of such problems is facilitated by use of the diagram as described beyond.

Mixtures of two waters in all proportions, if all products remain in solution, plot in the three fields on the respective straight lines that join the points representing the respective chemical characters of the two waters mixed. Thus, in Figure 5-A the straight lines AB will include the plottings of every possible mixture of two waters whose chemical characters are represented by points A and B, respectively. Point M represents a possible mixture in one particular proportion.

There is an obvious application of this procedure in demonstrating a cause for deterioration of water quality in a coastal area--whether or not due to simple invasion by ocean water. If so, chemical analyses of the progressively deteriorated water must, within reasonable limits of

A, Two-component mixture

B, Three-component mixture

FIG. 5 -- PRELIMINARY CONFIRMATION OF MIXTURES

A, Separation of calcium sulphate in solid phase

B, Reduction of sulphate

C, Base-exchange softening

D, Chemical reaction and separation of solid phase
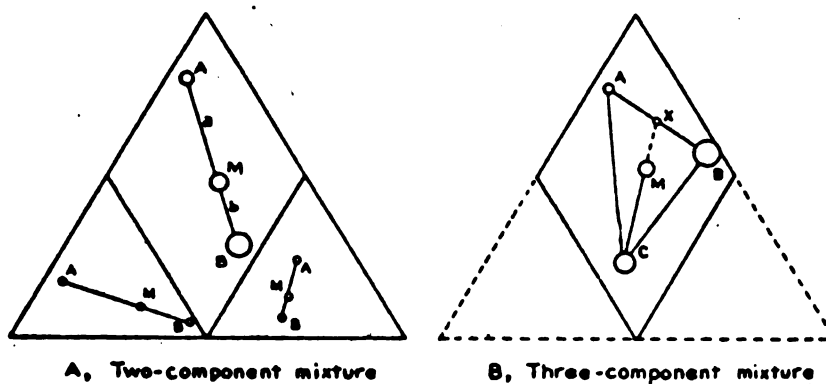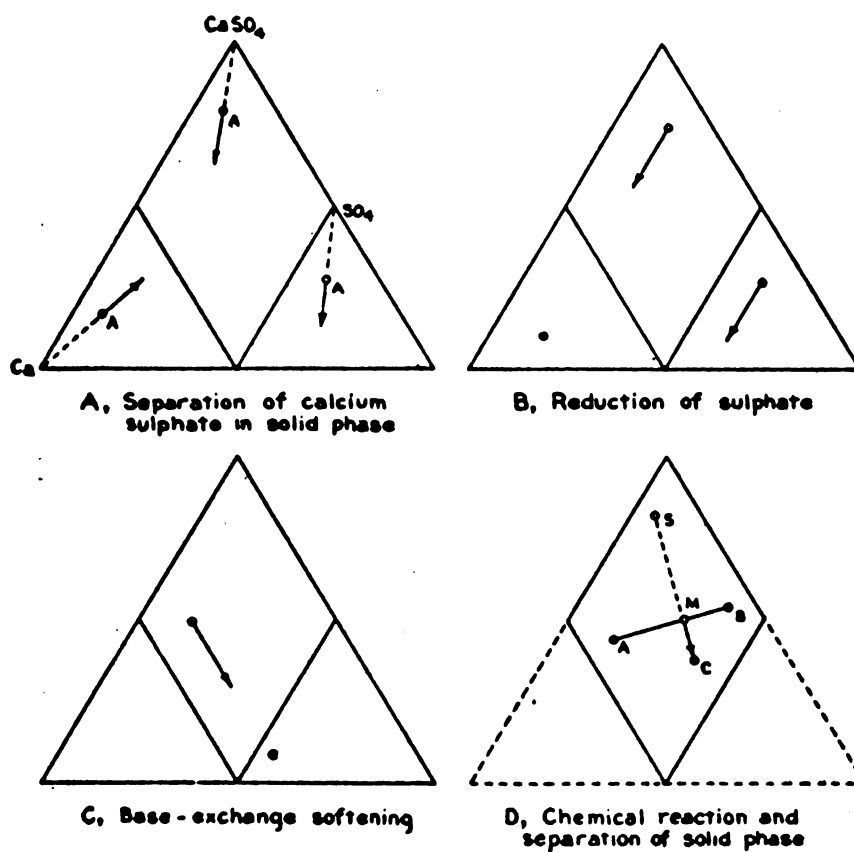
FIG. 6 -- VECTORS CHARACTERISTIC OF CERTAIN GEOCHEMICAL PROCESSES

error, plot on a set of three vectors directed toward the composition of ocean water. If the analyses do not so plot, simple admixture of seawater is not a valid and adequate explanation of the deteriorations. For example, on Figure 1 there are plotted the chemical characters of five natural waters from a longshore area in whose ground waters, at certain places, the content of chloride and certain other constituents has changed very substantially in recent years. The five waters include: Two from well A, in 1931 and 1940, respectively; two from adjacent well B, in 1931 and 1936, respectively; and one from the ocean a few miles away. From the plottings on Figure 1 it is obvious that this deterioration in ground-water quality could not have been caused by a simple intermingling of the fresh ground-water with ocean water.

To demonstrate conclusively that a certain water is a quantitative mixture of two other waters--neither diluted, concentrated, nor chemically modified after the mixing--one graphic criterion and one graphic-algebraic criterion must be satisfied. First, by the graphic criterion, in all three fields of the diagram the apparent mixture must plot on straight lines between the plottings of its two inferred components. Also, the area-concentration plottings in the central field must conform to the principle that the concentration of a mixture is necessarily greater than the least, but less than the greatest, of the several concentrations of its components. This graphic criterion alone is not decisive because it involves only percentage reacting values and does not involve absolute concentrations. Neither is any other simple graphic construction on the diagram decisive. The second and decisive criterion requires satisfaction of the following equations:

With reference to Figure 5-A let: V_a = proportionate volume in mixture M of water having composition A; V_b = proportionate volume of water having composition B; E_a = concentration of water A, in equivalents; E_b = concentration of water B, in equivalents; E_m = concentration of the mixture, in equivalents; a = intercept between the plottings of A and M, measured in any of the three fields of the diagram and at any convenient scale; and b = intercept between the plottings of B and M. Then it follows and can be shown that

$$(a/b) = (V_b \cdot E_b / V_a \cdot E_a) \qquad (1)$$

$$(V_a / V_b) = (b \cdot E_b / a \cdot E_a) \qquad (2)$$

$$E_m = \left[E_a \cdot E_b (a + b) \right] / (a \cdot E_a + b \cdot E_b) \qquad (3)$$

$$V_a = b \cdot E_b / (a \cdot E_a + b \cdot E_b) \text{ and } V_b = a \cdot E_a / (a \cdot E_a + b \cdot E_b) \qquad (4)$$

Further, with respect to any particular chemical constituent, let C_a = concentration in component water A, in equivalents or parts per million as desired; C_b = concentration in component B; and C_m = concentration in the mixture. Then

$$C_m = (C_a \cdot V_a) + (C_b \cdot V_b) \qquad (5)$$

Decisive proof of a quantitative mixture is accomplished when, for the water of intermediate composition, agreement is shown between analytical data and corresponding values computed from equation (3) for total concentration and from equations (4) and (5) for concentration of individual chemical constituents. Equation (3) is numerically equivalent to a corresponding equation derived by Langelier and Ludwig (1942, pp. 350-351), but is expressed in a form that facilitates the necessary computations.

As has been pointed out by Hill (1940, pp. 48-49), the chemical character of a mixture of waters will plot at the center of gravity of the plottings of the respective components, each having been weighted according to its concentration and its proportionate volume in the mixture. Based on this principle, graphic-algebraic criteria for decisive proof of quantitative mixtures are perhaps feasible for a three-component system but become involved for systems with more than three components. Problems involving three or more components are likely to be infrequent; accordingly, it seems most practicable to solve them by an adaptation of the two-component criteria given above. Thus, on Figure 5-B let M represent a water presumed to be a mixture of waters A, B, and C. To prove or disprove a quantitative mixture project a straight line on the diagram through C and M to intersect AB at point X, which represents the chemical composition of a hypothetical mixture of waters A and B in the same proportions that these waters would enter a mixture of composition M. From preceding equations (3), (4), and (5) determine the composition and concentration of water X. Then, treat water M as a presumed mixture of waters X and C. As desired, this procedure is readily adaptable to mixtures of more than three components.

Application to geochemical problems

The greatest utility of the diagram herein described probably is in "screening" a large number of water-analyses for critical study with respect to sources of the dissolved constituents, modifications in chemical character as a water passes through an area, and related geochemical problems.

Changes in the chemical character of a natural water by solution of progressively increasing amounts of some particular mineral must plot on a set of straight-line vectors directed in each of the three fields toward the point representing the chemical composition of the mineral. This case is analogous to mixing one water with another whose concentration is infinitely great.

A natural water may be concentrated progressively by evaporation until it becomes saturated with respect to certain constituents, which then separate out in the solid phase. Until a saturation-point is reached, the chemical character of the concentrating water is represented by a single fixed point in each of the three fields of the diagram. If the material separating in the solid phase is of constant composition then, as evaporation continues, the character of the water will be traced on the

respective fields by straight-line vectors directed away from the points
that represent the composition of the separating solid. If the solid
phase is a simple compound of one cation and one anion, the vector in
the central field will be directed away from one of the apexes of that
field, each of which represents a particular simple salt or group of
salts. In the two triangular fields, the vector will be directed away
from the respective apexes that represent the cation and anion composing
the solid phase. For example, on Figure 6-A point A represents the com-
position of a hypothetical water that is saturated with respect to cal-
cium sulfate ($CaSO_4$). As evaporation then continues and calcium sulfate
forms in solid phase, the changing composition of the liquid phase will
trace the three vectors shown. If a separating solid phase is a mixture
of salts in a constant proportion, a straight line will be traced in the
central field trending away from the point that corresponds to the con-
stant composition of the mixture; straight lines may or may not be traced
in the two triangular fields.

Certain changes in the chemical character of a water are caused
by chemical reactions which in effect substitute one cation or one anion
for another, molecule for molecule. Thus, reduction of sulfate (Revelle,
1941, pp. 595-596) is equivalent to substitution of bicarbonate (HCO_3)
for an equivalent amount of sulfate (SO_4); natural softening by reacting
with base-exchange minerals (Renick, 1925, pp. 63-68) to substitution of
sodium and potassium (Na, K) for calcium and magnesium (Ca, Mg). These
two chemical changes are traced on the diagram by straight-line vectors
parallel to the bases of the central field as shown by Figures 6-B and
6-C, respectively.

If two waters (or a water and a mineral) react chemically when
brought together and some product or products of the reaction form in
solid phase, the chemical character of the products remaining in solution
will not plot on the straight line joining the points that represent the
two reacting waters. Rather, it will plot on the extension of the
straight line drawn from (1) the point that represents the composition of
the solid phase to (2) the point that indicates the proportionate volumes
and compositions of the two reacting waters. Thus, in Figure 6-D suppose
that waters A and B are brought together in a proportion indicated by
point M on the straight line AB, and that a solid phase of composition S
results; then, the soluble products will plot on the extension of the
straight line SM, as at point C. If the precipitate is a compound of one
base and one acid, this relation between points A, B, M, S, and C will
apply likewise in the two triangular fields (not shown) and point S will
fall on one apex of each field. If the two waters are brought together
in various proportions and the product S is appreciably soluble, the com-
position of the liquid phase will traverse line AB up to the point of
saturation with respect to product S and then, beyond the point of satura-
tion, will deflect away from point S.

The chemical composition of a natural water may undergo complex
changes by an interplay of several or numerous processes. The causes are
commonly obscure. However, when comprehensive chemical data are available
the diagram herein described can assist greatly in a preliminary discrimi-
nation of causes, by application of the principles just described. Doubt-

less other useful principles will be developed as this diagram and simi-lar diagrams are more widely used.

Preliminary analysis of a typical problem

The plottings on Figure 1, to which reference has been made, are typical of a problem in the saline contamination of fresh ground-water in a longshore area. Table 2 gives the corresponding numerical data.

From their analytical data one could infer that waters B_1, A_2, and B_2 represent progressive stages in the contamination of water A_1 by some unknown high-chloride source. Because these are longshore ground waters from wells only about 200 feet deep, the ocean is an obvious potential source of a high-chloride contaminant. However, Figure 1 shows conclusively that the contaminated ground waters are not simply a mixture of ocean water with uncontaminated water A_1, because their plottings do not conform to the graphic criterion for a simple mixture as already developed. Specifically: (1) In the cation-triangle, waters A_2 and B_2 do not fall between the plottings of A_1 and C, although all the plottings are in substantial alighment; (2) in the anion-triangle, B_1, A_2, and B_2 all plot below the line A_1C; and (3) in the central diamond-shaped field B_1 plots very slightly above, but A_2 and B_2 plot far above the line A_1C. If the analysis of water B_2 had not been available, these discordances would not have been obvious in the analytical data. Neither would the analysis of water B_1 have shown clearly that it represented the incipient stage of contamination.

With reference to the corresponding two hypothetical mixtures, waters A_2 and B_2 contrast sharply in two respects: (1) Their content of calcium is much greater and that of sodium is much less; in percentage reacting value the excess of calcium is substantially equal to the defi-ciency of sodium, as though the hypothetical mixtures had been hardened by an ion-for-ion exchange of bases with the water-bearing material (see Fig. 6-C). (2) Their content of sulfate is substantially deficient, as would be expected if sulfate had been reduced to bicarbonate (see Fig. 6-B). These two contrasts also appear to apply in a small measure to water B_1, thus tending to confirm the inference that this water repre-sents an incipient stage in a common process of contamination.

Table 2--Principal chemical constituents of certain longshore ground waters and of ocean water

Constituent	A_1	B_1	b_1	A_2	a_2	B_2	b_2	C
Parts per million								
Calcium (Ca)	39	40	39	102	42	466	65	393
Magnesium (Mg)	10	10	11	19	22	77	98	1,228
Sodium (Na)	47	52	56	[54	152	255	808	[10,220[a]
Potassium (K)				3.6]				353]
Carbonate (CO_3)	..	..	..	0	..	..	..	0
Bicarbonate (HCO_3)	204	207	204	203	203	166	199	139
Sulfate (SO_4)	24	21	26	6.7	49	0	207	2,560
Chloride (Cl)	16	32	32	199	199	1,346	1,346	18,360
Percentage reacting values								
Calcium (Ca)	40.4	39.3	36.8	56.0	19.9	57.2	7.0	3.4
Magnesium (Mg)	17.1	16.2	17.1	17.2	17.2	15.6	17.3	17.6
Sodium and potassium (Na + K)	42.5	44.5	46.1	26.8	62.9	27.2	75.7	79.0
Totals	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0
Bicarbonate (HCO_3)	77.9	71.7	69.9	36.7	33.4	6.7	7.1	0.4
Sulfate (SO_4)	11.6	9.2	11.3	1.5	10.2	0	9.5	9.3
Chloride (Cl)	10.5	19.1	18.8	61.8	56.4	93.3	83.4	90.3
Totals	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0

[a] Calculated

Notes: A_1 and A_2 indicate water from Well A on June 3, 1931, and December 20, 1940, respectively. B_1 and B_2 indicate water from Well B on June 6, 1931, and June 30, 1936, respectively. C indicates water from the ocean a few miles from wells A and B. a_2, b_1, and b_2 indicate hypothetical mixtures of waters A_1 and C in such proportions that their chloride contents are equal to those of A_2, B_1, and B_2, respectively.

Obviously the data here presented are not adequate fully to define this water-quality problem, in part because the analyses of waters A_1, B_1, and B_2 are approximate only. However, as an elementary example of procedure they are especially effective because they afford a striking comparison but involve only water-quality and time as principal variables. A complete solution of the problem here suggested involves data so voluminous that it is not feasible to introduce them.

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