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DISSOLVED SILICATE AND RELATED PROPERTIES OF SOME WESTERN NORTH ATLANTIC AND CARIBBEAN WATERS¹

By

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"As marine plants are the ultimate source of food for much of the animal life in the sea, the role played by silicon is of fundamental importance".

T. G. Thompson and R. J. Robinson, 1932

ABSTRACT

Dissolved silicate, phosphate, nitrate, and oxygen as well as salinity and temperature were observed in several regions around the Antilles Arc and in the Woods Hole-Bermuda area. It is shown that silicate is generally in low concentration in surface waters, where its ratio to phosphate, nitrate and oxygen consumption is variable. At intermediate depths, silicate, phosphate and nitrate regeneration and oxygen consumption (AOU) proceed in the ratios, by atoms, of $\Delta \text{AOU} : \Delta \text{Si} : \Delta \text{N} : \Delta \text{P} = 270 : 16 : 16 : 1$. In some of the water masses, silicate, nitrate and phosphate occur in these same ratios but are in excess of the amount of regeneration expected from the amount of oxygen consumption. However, silicate is clearly and linearly related to biological processes. In other water masses, notably in deep water of the North Atlantic, the Puerto Rico Trench and the eastern Caribbean Sea, and in anaerobic water of the Cariaco Trench, there are higher concentrations of silicate than would be expected from the amounts of phosphate present. These may originate from *in situ* re-solution of skeletal remains of diatoms or from re-solution of bottom deposits followed by upward diffusion.

Introduction. The wide variability of silicate in the ocean should make it a valuable characteristic for tracing water masses, and its biological involvement in the sea is of special interest. Although silicate is used in relatively large quantities by only a few groups of organisms, principally diatoms, these occur in large numbers and are important primary producers of organic matter. This special relationship between silicate, the diatoms, and organic production sets silicon apart from nitrogen and phosphorus, which are used by all organisms. Some of the silicate which diatoms fix concurrently with

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their organic matter is redissolved in the water column upon their death and thus re-enters the ecological cycle. The balance eventually becomes incorporated in bottom sediments, there to become a part of the geological record and to represent a phase in the geochemical migration of silicon. This paper relates the distribution of silicate in several oceanic areas to the biological cycle through nitrogen, phosphorus and dissolved oxygen and to hydrography through its distribution and temperature-salinity correlations.

Studies of seasonal variations in silicate have demonstrated a close relationship between diatom growth and depletion of silicate from the water, and conversely, between regeneration of silicate and a decrease in the diatom population (Rabin, 1905a, b, 1910, 1914; Brandt, 1920; Atkins, 1923a, b, 1926a, b, 1928, 1930; Thompson and Johnson, 1930; *et al.*). Studies of silicate and its distribution have been made by Clowes (1938), by Graham and Moberg (1944), and by others. The data of Atkins (1928, 1953, *inter alia*), Cooper (1933, *et seq.*), and others of the laboratory of the Marine Biological Association at Plymouth, England, show seasonal variations in silicate and phosphate which are more or less parallel; Cooper's paper on factors affecting the distribution of silicate shows (1952: fig. 1) an approximately linear relationship between phosphate and silicate between about 25 and 600 m at a station in the Bay of Biscay.

It has been stated that silicate in the ocean increases continuously with depth (Sverdrup, *et al.*, 1942; Graham and Moberg, 1944), but some of the DISCOVERY data (Discovery Committee, 1941: Sts. 1162-1184) show silicate maxima at slightly less than 1000 m in the equatorial and South Atlantic. This type of vertical distribution was the rule in a series of stations occupied by ATLANTIS in the equatorial Atlantic in 1952. The maxima were clearly associated with the phosphate maxima and oxygen minima. These observations made it clear that some features of the silicate distribution resulted from closely correlated biological phenomena.

Observations. Silicate was determined in samples from the eastern Venezuelan Basin and the Cariaco Trench in the Caribbean Sea; the Atlantic region east of the Windward Islands from Tobago to Martinique; the Puerto Rico Trench-Anegada Passage region (Table I); and the area between Woods Hole and Bermuda. Observations between Woods Hole and Bermuda were made on R/V BEAR Cruise 185, May 12 to 16, 1958 and R/V CRAWFORD Cruise 19,

TABLE I. STATIONS IN THE ANTILLES ARC REGION

| <i>Date</i> | <i>Station Number</i> | <i>North Lat.</i> | <i>West Long.</i> | <i>Bottom Depth (m)</i> | <i>Depths at which eq. (1) is Satisfied</i> |
|---------------------------|-----------------------|-------------------|-------------------|-------------------------|---|
| VENEZUELAN BASIN | | | | | |
| 1955 | | | | | |
| 12. II | 5272 | 13°30' | 65°24' | 3860 | Phosphates not available |
| 14. II | 5273 | 14 14 | 63 25 | 1464 | Phosphates not available |
| 6. III | 5282 | 13 09 | 62 15 | 2885 | 192-482; 867 |
| 7. III | 5283 | 14 13 | 62 55 | 1483? | 195-781 |
| 9. III | 5284 | 16 03 | 66 02 | 4301 | 267-806 |
| ANEGADA-JUNGFERN PASSAGES | | | | | |
| 12. III | 5285 | 17°54' | 65°51.5' | 1955 | 188-1038; 1623-1818 (d.s.) |
| 14-15. III | 5286 | 17 53 | 65 20 | 3820 | 266-1030; 1850-3440 (d.s.) |
| 15. III | 5287 | 17 54.5 | 64 16 | 1975 | 279-854 |
| PUERTO RICO TRENCH REGION | | | | | |
| 17. III | 5288 | 18°52' | 63°05' | 5880 | 455-2898 |
| 19. III | 5289 | 19 56 | 62 50 | 6050 | 406-1461 |
| 20-21. III | 5290 | 19 54 | 64 32 | 7260 | 385-2395 |
| 22. III | 5291 | 19 52 | 66 35 | 7690 | 352-1964 |
| EAST OF THE WINDWARDS | | | | | |
| 19. II | 5274 | 14°80' | 60°42' | 1089 | 140-488 |
| 21. II | 5275 | 13 54 | 59 47 | 717 | 147-495 |
| 22. II | 5276 | 13 25 | 58 19 | 2880 | 197-2750 (d.s.) |
| 23. II | 5277 | 13 50 | 56 53 | 4812 | 189-2387 |
| 24. II | 5278 | 12 36 | 58 34 | 1851 | 195-1744 (d.s.) |
| 26. II | 5279 | 12 41 | 59 40 | 1020 | 181-561 |
| CARIACO TRENCH REGION | | | | | |
| 1957 | | | | | |
| 1. XI | 5596 | 10°52' | 66°00' | 185 | 54-161 |
| 1. XI | 5597 | 10 06 | 66 05 | 260 | 200-230 (not in Trench) |
| 2. XI | 5597 A | 10 48 | 65 52 | 252 | 150-245 |
| 2. XI | 5598 | 10 44 | 65 54 | 412 | 200 |
| 4. XI | 5599 | 10 19 | 65 32 | 58 | none |
| 4. XI | 5600 | 10 28 | 65 43 | 554 | 200 only |
| 4. XI | 5601 | 10 37 | 65 32 | 722 | none sampled |
| 5. XI | 5602 | 10 52 | 65 33 | 845 | 197, 342 |
| 6. XI | 5603 | 11 15 | 65 07 | 784 | 174-498 (not in Trench) |
| 6. XI | 5604 | 10 54 | 65 08 | 58 | none |
| 6. XI | 5605 | 10 38 | 65 08 | 933 | 228 only |
| 8. XI | 5606 | 10 32 | 64 40 | 1343 | 186 only |
| 8. XI | 5607 | 10 32 | 64 41 | 1337 | 196 only |

d.s. = deepest sample.

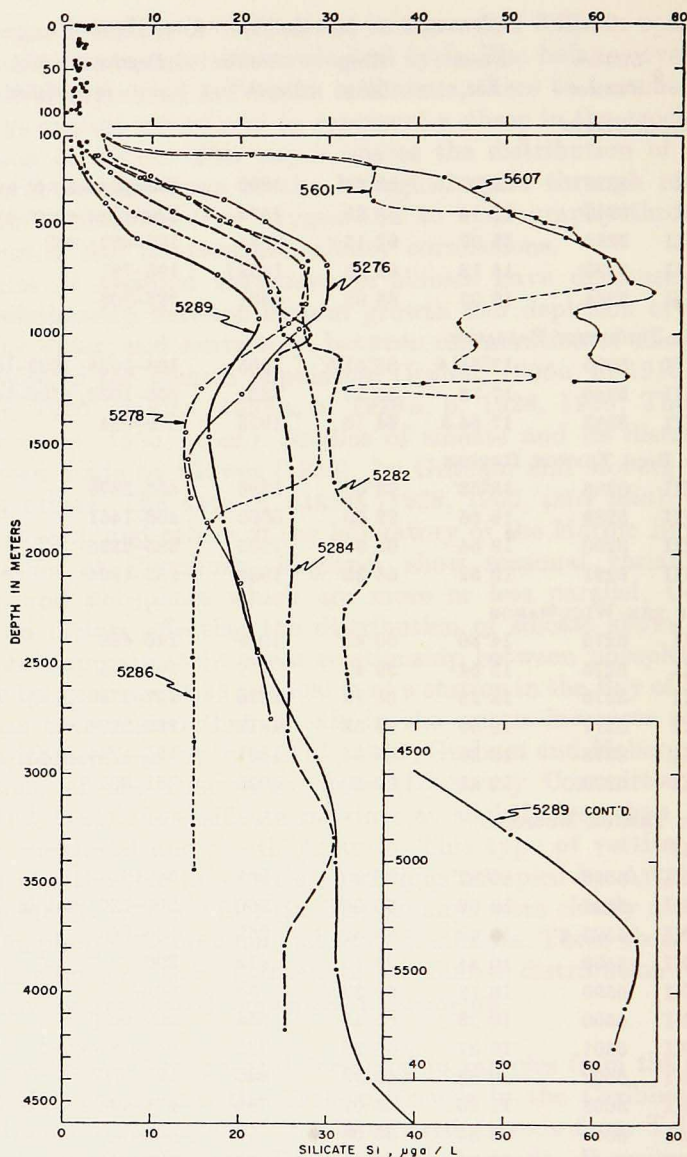


Figure 1. Silicate concentrations *vs* depth at representative stations in the Antilles Arc region. Sts. 5601 and 5607 in the Cariaco Trench; 5282 and 5284, Venezuelan Basin; 5276, 5278, Atlantic Ocean east of the Windwards; 5289, Puerto Rico Trench; 5286, Anegada-Jungfern Passage region. Open circles correspond to those in Figs. 3 and 4.

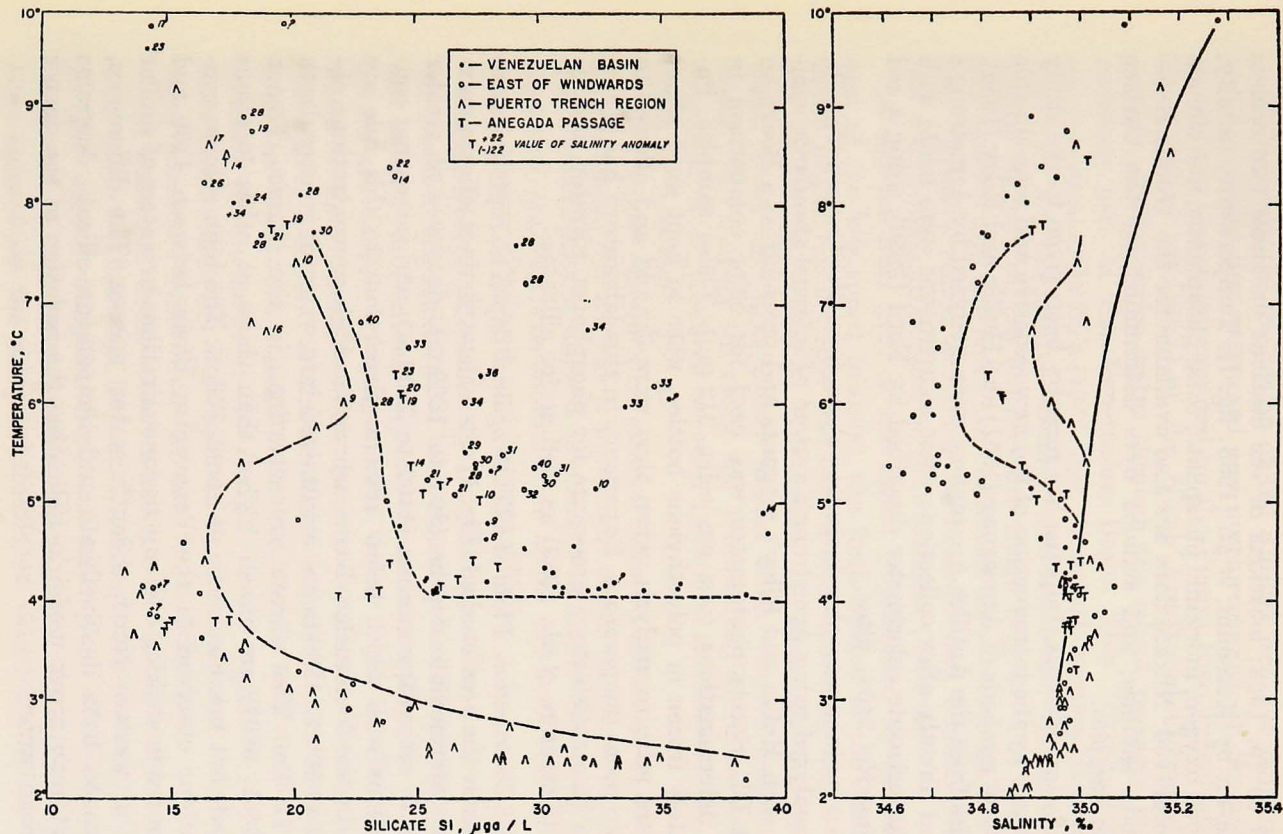


Figure 2. Temperature-silicate and temperature-salinity correlations. The solid curve is Iselin's (1936) correlation for western Sargasso Sea waters. Numbers on the temperature-silicate diagram are salinity anomalies and are negative unless otherwise indicated. Points having no numbers have salinity anomalies less than ± 5 . Question marks indicate no salinity values are available. The short dashes envelope the samples from the Venezuelan Basin on the T/Si diagram, and those from the Puerto Rico Trench and from east of the Windwards on the T/S diagram. The long dashes envelope the samples from the Puerto Rico Trench on both diagrams. All samples from between Woods Hole and Bermuda having more than $20 \mu\text{g-at silicate-Si/l}$ (CRAWFORD Cruise 19) come from depths greater than 2500 m.

July 7 to 19, 1958; locations of the stations on these two cruises are shown by Ketchum, *et al.* (1958: fig. 1). Temperature, salinity, dissolved oxygen, inorganic phosphate, total phosphorus, and silicate were observed. Nitrate data are also available for the Woods Hole-Bermuda samples, and sulfides were determined for the Cariaco Trench samples.

Analytical Methods. Impetus has recently been given to the study of silicates by the introduction of the new sensitive and reproducible analytical methods of Armstrong (1951) and Mullin and Riley (1955). Samples from the Antilles Arc regions were analyzed by Armstrong's method directly after collection; color comparisons were made with the photoelectric colorimeter described by Ford (1950), using a red (Corning No. 2404) filter.

The remaining samples, including those from the Cariaco Trench, were analyzed using Armstrong's system of internal standards combined with Mullin and Riley's reagents and procedures; a Beckman Model DU Spectrophotometer was used for color comparison in these determinations (one cm cells, 812 $m\mu$). These samples, immediately frozen in polyethylene bottles, were so kept until about 12 hours prior to analysis, when they were thawed and allowed to come to room temperature. Experience in this laboratory has shown this to be a satisfactory procedure for phosphates, nitrates, and nitrites (Ketchum, *et al.*, 1958) as well as for silicates.

Depth Distribution. Plots of silicate against depth at representative stations in the area studied (Fig. 1) often show distinct silicate maxima at intermediate depths (800 to 1200 m), decreases at greater depths to secondary minima (1400 to 1800 m), and increasing concentrations with depth below 1800 m. Exceptions to this rule are found in the Venezuelan Basin, where the high concentrations observed at 800 to 1200 m are maintained into water of greater depth than 1200 m. The silicate concentrations in the Cariaco Trench (Sts. 5601, 5607) are much higher than those at other locations sampled, but maxima occur at about 800 m. The high silicate concentrations observed in the Venezuelan Basin between 1100 and 2600 m are in striking contrast to concentrations in waters of similar depth in western North Atlantic water masses. The differences, obvious on both depth-silicate and temperature-silicate diagrams (Fig. 2), may prove useful in indicating the isolation of the deeper Caribbean water.

Hydrography. In the region of the Antilles Arc, the Atlantic currents generally flow westward and northwestward. In the northern part of the region (represented by the Puerto Rico Trench stations, Fig. 2), the "salt half" of the North Equatorial Current (Montgomery, 1938) carries water through the island passages into the northern part of the Venezuelan Basin. This "North Atlantic" water is close to Iselin's (1936: figs. 25 and 53) western Sargasso Sea water in its temperature-salinity characteristics, though a trifle fresher in part of the temperature range (Fig. 2). The deepest of this water which can enter the Caribbean does so through the Jungfern Passage, which, according to Frassetto and Northrop (1957), has a controlling sill depth of 1072 fathoms (1961 m).

In the southern part of the region (represented by the stations east of the Windward Islands), the fresh tongue of the North Atlantic Equatorial Current (between 5 and something above 10° C; 400 to 1000 m) contains admixtures of Antarctic Intermediate water which are significantly fresher than western Sargasso Sea water. The deepest sill along the Windwards, which according to Sverdrup, *et al.* (1942) is between Dominica and Martinique, has a depth of less than 1500 m.

The Cariaco Trench is an isolated basin to the south of the Venezuelan Basin. Its sill has a maximum depth of about 150 m, and below this depth the basin is so poorly ventilated that it has become anaerobic and contains sulfides from about 400 m to the bottom at about 1400 m (Richards and Vaccaro, 1956).

Silicate-Temperature-Salinity Relationships. The water masses of the Antilles Arc region have been characterized by plotting silicate and salinity, respectively, against temperature (Fig. 2). These diagrams are useful, at temperatures below 7° C, in distinguishing water of four areas: (a) The eastern Venezuelan Basin; (b) The Atlantic Ocean east of the southern islands of the Antilles Arc, *i. e.*, the Windward Islands from Martinique to Tobago; (c) The Atlantic Ocean north of the Antilles in the region of the Puerto Rico Trench; and (d) The region of the Anegada and Jungfern Passages between Puerto Rico and the Virgin Islands.

The waters of these areas colder than 10° C have been compared with Iselin's (1936: figs. 25 and 53) temperature-salinity diagrams for the western Sargasso Sea, and salinity anomalies have been computed. Where these have a value greater than ± 5 , they have been entered on the temperature-silicate diagram (Fig. 2).

Significant negative salinity anomalies, indicating admixture of Antarctic Intermediate water, are accompanied by correspondingly high silicates. Water of this type is found in the 6 to 7° C range east of the Windwards, at the Anegada-Jungfern Passage stations, and in the Venezuelan Basin. It extends down to 5° C east of the Windwards, but the 5 to 6° C water in the Anegada-Jungfern Passage region and in the Venezuelan Basin tends to be somewhat more saline, the latter being more like the Antarctic Intermediate water than the former.

At temperatures colder than 5° C there are no significant salinity anomalies in the Antilles Arc region, *i. e.*, all of the water masses have about the same temperature-salinity correlations. It is in the 4 to 5° C range that silicate concentrations are distinctive and clearly distinguish the silicate-rich Caribbean waters from the silicate-poor Atlantic waters, with the Anegada-Jungfern Passage samples being of an intermediate character. Water colder than 4° C is absent from the Caribbean, but such waters have similar silicate and salinity characteristics in both of the Atlantic regions concerned. The still colder water in the depths of the Puerto Rico Trench continues to increase in silicate to the greatest depths sampled.

Biochemical Relationships. The requirement of diatoms for large quantities of silicate, the marked correspondence between diatom outbursts and silicate depletion, and the preponderance of diatoms among photosynthetic marine plants lead one to inquire into the possibility of relating the distribution of silicate in the ocean to biological phenomena. Redfield (1934), Fleming (1940), Cooper (1938), Gilson (1937) and others have demonstrated statistical correlations among inorganic phosphate, nitrate, carbon dioxide, and dissolved oxygen in the water and the composition of marine organisms (see Richards and Vaccaro, 1956, for a review). In the regions studied, the concentration of silicate is also shown to be linearly related to the regeneration of phosphate and nitrate and to oxygen consumption in much of the water column. On the other hand, there are, in addition, hydrographic sources of silicate, not demonstrably related to biological fixation and regeneration, which account for significant proportions of silicate, particularly in shallow and very deep waters.

Silicate-Phosphate Relationships. Plots of silicate as a function of inorganic phosphate (Fig. 3) show that, at the intermediate depths

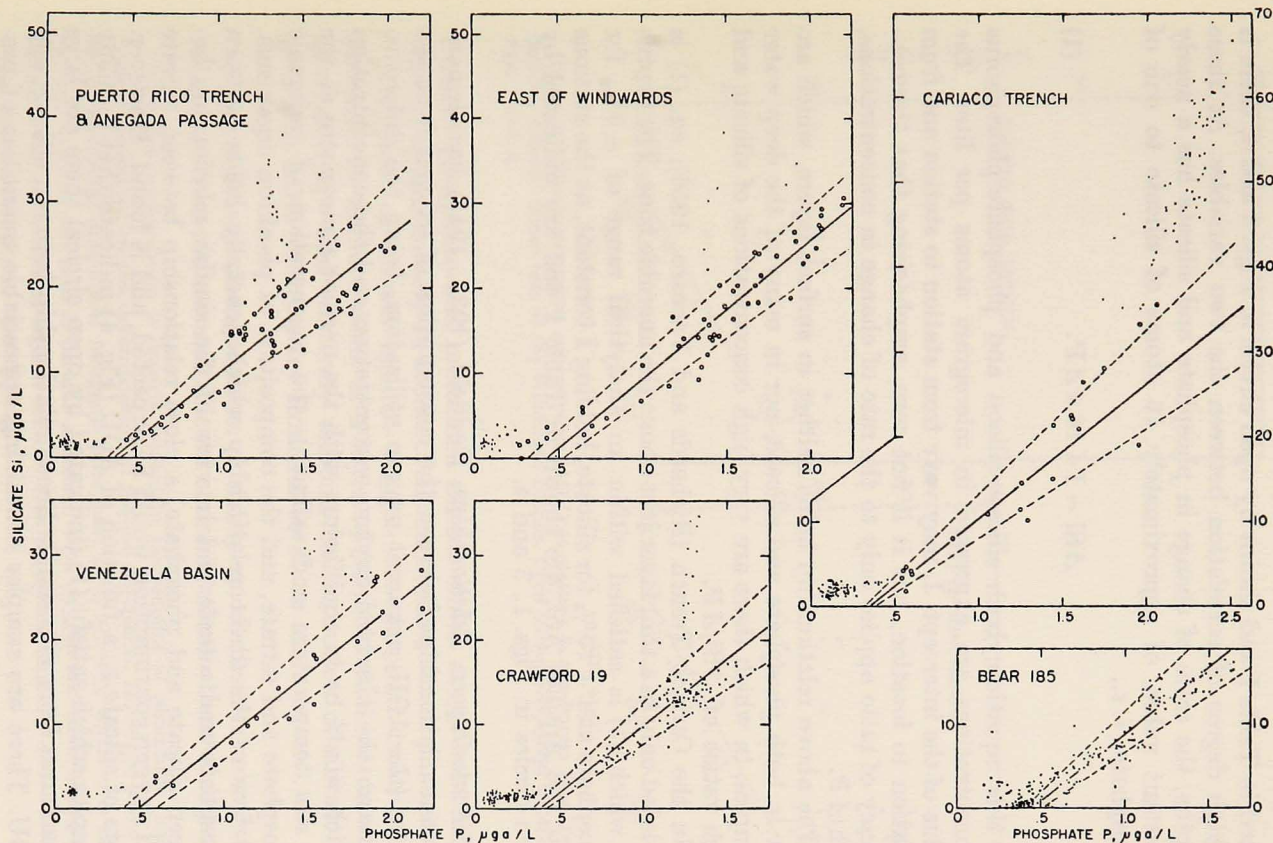


Figure 3. Silicate-phosphate relationships. The regression lines (solid) have slopes of $\Delta\text{Si}:\Delta\text{P} = 16:1$, by atoms. The broken envelope lines include all points along the regression lines with allowances for $\pm 9\%$ analytical errors in phosphate and $\pm 10\%$ in silicate. Open circles represent samples in which eq. (1) is assumed to be satisfied (see text).

listed in Table I and shown by open circles in Figs. 3 and 4, there is a high degree of correlation between the two variables. At these depths, the ratio of change in phosphate and silicate has a nearly constant value, of approximately 16 atoms of silicate to one of phosphate, *i. e.*,

$$\Delta \text{Si} = \text{I} + 16 \times \Delta \text{P}. \quad (1)$$

In this equation, both silicate-silicon and phosphate-phosphorus concentrations are expressed in microgram atoms per liter. The value of the intercept, I, may vary from station to station and from location to location, but it is not zero, emphasizing that the constancy of ratio applies only to the rate of change in concentrations, $\Delta \text{Si}/\Delta \text{P}$.

The above relationship holds neither in surface layers, which are low in both phosphate and silicate, nor in many of the deep water samples, in which there are very high concentrations of silicate and high ratios of $\Delta \text{Si}/\Delta \text{P}$.

In the Cariaco Trench (Richards and Vaccaro, 1956), eq. (1) is satisfied only in a thin layer just above the anerobic zone. The depths at which (1) is satisfied within an analytical range of $\pm 9\%$ for phosphate and $\pm 10\%$ for silicate, holding I constant, at the stations near the Antilles Arc, are shown in Table I and are indicated by open circles in Figs. 1, 3 and 4.

Silicate-Oxygen Relationships. Redfield (1934, 1942) has demonstrated relationships between the consumption of oxygen from sea water (the AOU, apparent oxygen utilization, being the difference between the observed oxygen concentration and the concentration which would be in equilibrium with the normal atmosphere at the *in situ* temperature and salinity), the regeneration of inorganic phosphate and nitrate, and the composition of plankton organisms. In view of the direct relationship which generally holds between phosphate and nitrate and in view of the similar relationship between silicate and phosphate, a close relationship between silicate and oxygen consumption would be expected, and is found. However, plots of silicate as a function of AOU (Fig. 4) indicate that in many samples which satisfy (1) (indicated by open circles) there are large departures from any simple linear relationship between silicate and AOU. These are samples containing appreciable quantities of pre-

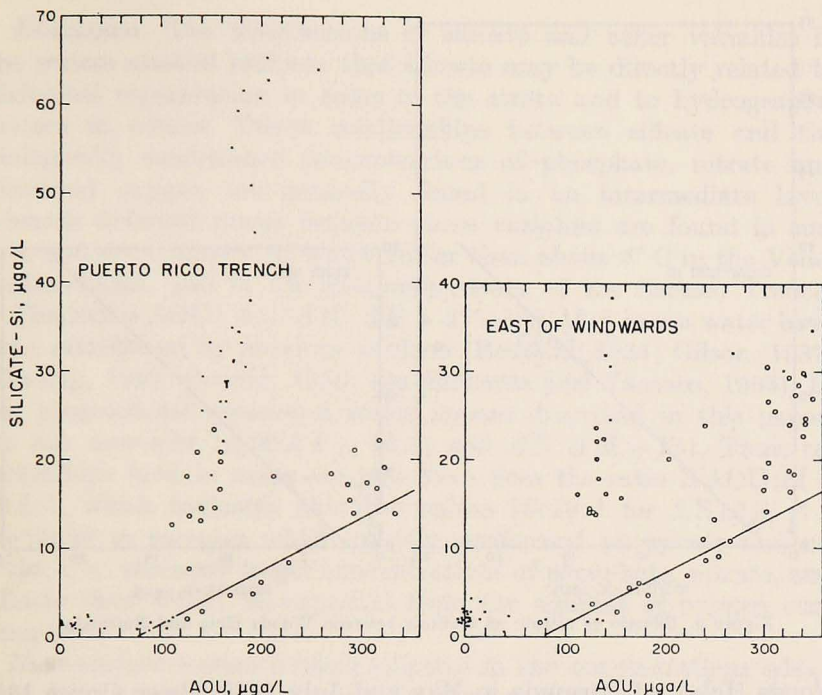


Figure 4. Silicate vs oxygen consumption (AOU) at stations in the Puerto Rico Trench and the Atlantic Ocean east of the Windward Islands. Open circles correspond to those in Figs. 1 and 3.

formed² phosphate and preformed silicate which are present in the ratio of approximately 16 atoms of silicon to one atom of phosphorus.

The plots of silicate against oxygen consumption indicate the presence, in upper waters, of a layer of variable thickness in which there is a low and relatively uniform concentration of silicate but in which AOU values vary from nil to 80 or more $\mu\text{g-at/l}$.

Silicate-Nitrate Relationships. A fairly good positive correlation exists between nitrate and silicate concentrations observed between

² Oxygen consumption is linearly related to the amount of phosphate (and nitrate) regenerated on the decomposition of marine organisms. However, there may be an additional amount of inorganic phosphate (and nitrate), which was in the surface water at the time it became saturated with oxygen and sank beneath the surface with its burden of organic matter. This phosphate (and nitrate) is referred to by Redfield (1942) as "preformed".

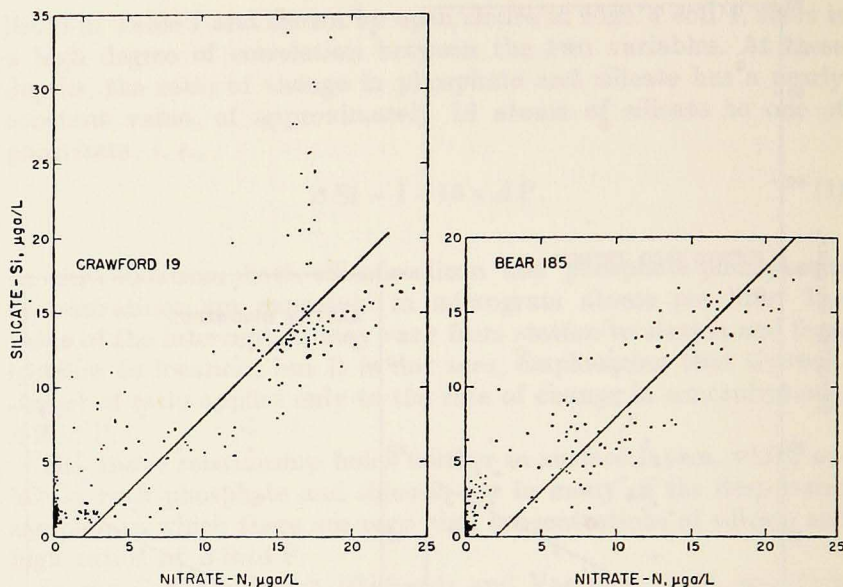


Figure 5. Silicate vs nitrate at stations between Woods Hole and Bermuda.

Woods Hole and Bermuda in May and July 1958 (BEAR Cruise 185 and CRAWFORD Cruise 19; see fig. 5), except in near-surface waters generally in both May and July and in the deep samples taken near Bermuda in July. Though there is much scatter in these data, the ratio of change in silicate-silicon to nitrate-nitrogen is close to one for one, by atoms. In deep waters near Bermuda in July (not sampled in May), the Si/N ratio of 1/1 is greatly exceeded.

The near-surface samples from this region show different situations prevailing during the two periods. The surface waters all have excess quantities of phosphate, as shown by Ketchum, *et al.* (1958), but concentrations of both silicate and nitrate are so low that one or the other or both may limit the further growth of planktonic organisms. The May samples show low nitrates ($< 2 \mu\text{g-atN/l}$) accompanied by up to $4 \mu\text{g-at/l}$ of silicate-silicon. However, in July, many of the low concentrations of silicate ($< 2 \mu\text{g-at/l}$) were accompanied by up to 3 and, in a few cases, by over $5 \mu\text{g-at/l}$ of nitrate-nitrogen. This suggests that, in May, low concentrations of nitrate were more probably limiting the growth of phytoplankton than low concentrations of silicate, while the opposite situation prevailed in July.

Discussion. The distributions of silicate and other variables in the waters studied indicate that silicate may be directly related to biological regeneration in some of the strata and to hydrographic factors in others. Direct relationships between silicate and the biologically conditioned concentrations of phosphate, nitrate and dissolved oxygen are generally found in an intermediate layer whereas different ratios between these variables are found in surface and deep waters, in water colder than about 5° C in the Venezuelan Basin, and in the anaerobic strata of the Cariaco Trench.

The ratios $\Delta \text{C} : \Delta \text{N} : \Delta \text{P} = 270 : 106 : 16 : 1$ in sea water have been established by previous workers (Redfield, 1934; Gilson, 1937; Fleming, 1940; Cooper, 1938; see Richards and Vaccaro, 1956). In the geographical areas and water masses described in this paper, we can now add: $\Delta \text{Si} : \Delta \text{P} = 16 : 1$, and $\Delta \text{N} : \Delta \text{Si} = 1 : 1$. These relationships hold in more samples than does the ratio $\Delta \text{AOU} : \text{Si} = 16.9 : 1$, which indicates that the values 16:16:1 for $\Delta \text{N} : \Delta \text{Si} : \Delta \text{P}$ are valid in samples which contain preformed phosphate and nitrate; *i. e.*, there are larger concentrations of phosphate, nitrate, and silicate than would be expected from the amount of oxygen consumption.

Near-surface waters contain silicates in low concentrations which vary from place to place and from time to time, and the ratios of silicate to nitrate and to phosphate are also variable in these waters. Between Woods Hole and Bermuda, where all three of these variables have been studied, there is evidence (Ketchum, *et al.*, 1958) that inorganic phosphate was always present in sufficiently high concentrations that growth of planktonic organisms was not limited. On the other hand, either silicate or nitrate might have been limiting, for there is a suggestion in the data that first one and then the other may be exhausted while nonlimiting concentrations of the other are still present.

In many of the waters studied, particularly the deeper samples, there are additional increments to the concentration of silicate, independent not only of the amount of oxygen consumption but also of the amounts of phosphate (and nitrate) regenerated. It appears that (1) silica re-solution may continue in deeper water after the regeneration of phosphate (and nitrate) has gone to completion in shallower strata, or (2) there may be re-solution of silica from bottom sediments with subsequent upward diffusion. These high silicate concentrations were observed in:

a. The Venezuelan Basin and Anegada Passage, where, between 4 and 5° C, they may arise from *in situ* re-resolution of siliceous organic remains;

b. The Cariaco Trench, a special locus of isolated water in which there has been extensive accumulation of silicate from re-resolution of siliceous materials; and

c. Deep Atlantic water; some of the increment in the deeper part of the Puerto Rico Trench may also be of local, *in situ* origin.

The high silicate concentrations in the deep water of the Venezuelan Basin constitute the most strikingly different water-mass characteristic which has been observed to distinguish Caribbean and western North Atlantic water. However, Caribbean water is also more than 0.5 ml/l poorer in oxygen and somewhat richer in phosphate than the Atlantic water directly to the north. These are the kinds of chemical differences which distinguish the stagnant or very poorly renewed deep water of the Cariaco Trench and which one would expect to find in any stagnant or poorly flushed marine basin. For these reasons, it seems that, contrary to Sverdrup, *et al.* (1942: 640), the renewal of the deep water of the Venezuelan Basin must be a relatively slow, not a relatively rapid, process. Worthington's (1955) observations have pointed toward these same conclusions, which the silicate observations tend to corroborate.

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