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THE EXCHANGE OF OXYGEN ACROSS THE SEA SURFACE¹

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In temperate latitudes the oxygen content of sea water near the surface varies with the season. This results because the solubility of oxygen in the water changes with its temperature and because of the unequal production and utilization of oxygen by organisms at different seasons. Both factors cause an exchange of oxygen across the sea's surface. The present paper is an attempt to determine the magnitude of this exchange as it occurs in the Gulf of Maine and to evaluate the factors responsible.

The solution of the problem depends upon the assumption that the column of water underlying a square meter of sea surface and extending to a depth beyond which seasonal changes in chemical composition are insignificant may be treated as a closed system except for exchanges with the atmosphere. If Q_o represents the quantity of dissolved oxygen observed to be present in the column and ΔQ_o its change between two periods of observation, ΔQ_E the quantity entering through the sea surface, and ΔQ_x the net increase in oxygen resulting from biological activity during the time, then the equation representing the net exchange across the sea surface is

$$\Delta Q_E = \Delta Q_o - \Delta Q_z. \tag{1}$$

Negative values of ΔQ_o and ΔQ_x represent decreases in the dissolved oxygen in the column and negative values of ΔQ_B represent oxygen escaping to the atmosphere.

EVALUATION OF ΔQ_{σ} AND ΔQ_{z}

The oxygen content of the column, Q_o , may be obtained by integrating the area enclosed by a curve representing the station data for

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oxygen concentration and depth. From the difference in Q_o obtained at successive surveys ΔQ_o is estimated.

The net change in oxygen content due to biological activity, ΔQ_z , may be deduced from observed changes in inorganic phosphate content of the column, PO₄. In the process of photosynthesis a quantity of oxygen is set free proportional to the phosphate withdrawn from the water. Conversely, the oxidation of organic matter in metabolism sets free a quantity of phosphate corresponding to the oxygen consumed (Redfield, 1934; Fleming, 1941; Edmondson and Edmondson, 1947). In order to convert changes in phosphate into the quantities of oxygen set free or absorbed in the accompanying biological processes, use is made of an equivalence based on the observation (made in the waters of the Sargasso Sea) that the disappearance of 1 cc O₂ accompanies the appearance of 0.373×10^{-3} mg-at of phosphate phosphorus (Redfield, 1942)². Correspondingly, the disappearance of 1 mg PO₄ is accompanied by the appearance of 28.3 cc O₂, or

$\Delta Q_x = 28.3 PO_4.$

Some uncertainty exists regarding the exactness of this relation in its present application, since it depends upon the assumption that in decomposition the process has gone to completion. Actually significant quantities of phosphorus are present in sea water at certain seasons in the form of dissolved organic compounds (Redfield, Smith and Ketchum, 1937). These do not contribute to the measurement of phosphate concentration; yet it cannot be doubted that their liberation was attended by some oxidation of associated organic matter. Increases in inorganic phosphate may consequently lag behind the attendant consumption of oxygen at the times and places where decomposition is occurring. Moreover, the value of the relation depends upon the average elementary composition of the organisms responsible, which may differ somewhat with time and place. Finally, the methods for determination of inorganic phosphate are not wholly satisfactory. Estimations of ΔQ_x are consequently subject to much greater errors than those of ΔQ_{α} .

DATA

The data examined were collected in the course of a survey of the Gulf of Maine made by the ATLANTIS in 1933-34. Eleven cruises were completed in the course of 15 months.

The area chosen for examination was the western basin of the Gulf,

² This equivalence is based on phosphate data uncorrected for salt error. See Cooper (1938). a region of 200 to 250 m in depth lying between Cash's Ledge and the Massachusetts Coast. Within an area 35 miles square, lying between the latitudes 42° 15' and 42° 50' N and the longitudes 69° 10' and 69° 45' W, there were available from two to four sets of station data for each cruise³.

The data were collected as part of a general hydrographic survey, and consequently no special provision was made to secure measurements designed to meet the special requirements of the present study.

In evaluating the quantity of oxygen and phosphate in the column under a square meter of surface at each period of observation, the data from the several stations occupied in the area were averaged. Oxygen data were available from three stations for each cruise except for January and August, when two stations were occupied, and for December, when four stations were occupied in the area. Phosphate data were available from three stations in September 1933, from two in July and December 1933 and in March and May 1934, and from one in January, April, June and September 1934. No data were available for October 1933 and August 1934.

It was assumed that seasonal changes in the quantity of oxygen in the water column are limited to a depth of 200 m and of phosphate to a depth of 100 m. Oxygen and phosphate contents were consequently integrated to these depths in estimating ΔQ_o and ΔQ_x .

The content of dissolved oxygen and of inorganic phosphate were both maximal in March. Consequently, it proved convenient to express ΔQ_o and ΔQ_x as the changes which have occurred or which will occur relative to the conditions obtaining at that time.

ASSUMPTIONS

The basic assumptions underlying the application of equation (1) are that exchanges of oxygen across the sides and lower surface of the water column have a negligible effect on its oxygen content and that equation (2) is valid. The latter assumption is sufficiently discussed above and in the references.

The assumption that exchanges resulting from the movement of oxygen or phosphate through the side walls of the column are negligible can be justified only by the consideration that the area of observation

[•] For data see Bulletin Hydrographique 1933 and 1934 M/Y Atlantis Stations Nos. 1699, 1703, 1721, 1747, 1748, 1749, 1856, 1857, 1858, 1864, 1865, 1875, 1876, 1909, 1910, 2022, 2023, 2024, 2074, 2075, 2076, 2168, 2169, 2170, 2220, 2221, 2236, 2258, 2259, 2273, 2274, 2275. Hydrographic observations were made by standard procedures. Oxygen was determined by Winkler's method and phosphate by the Denigès-Atkins procedure. The values for phosphate are not corrected for salt error.

is centered in a much larger region in which the horizontal distribution of components is uniform at any depth. The components which leave the column from one side by lateral mixing or flow may be supposed to be replaced almost exactly by those which enter from the opposite side. The seasonal changes in the concentration are so large compared to the gradients which are present in a horizontal direction at any time



FIGURE 1. Seasonal variation of oxygen content of successive zones of 50 m depth beneath one m² of sea surface in Gulf of Maine. that it seems unlikely that lateral transport has an important effect on the changes in the composition of the column.

The assumption that exchanges through the lower surface of the column are negligible requires

- (1) that its depth be greater than that at which seasonal effects on the components can be observed and
- (2) that the net movement of water from below by vertical mixing is insignificant.

The western basin of the Gulf of Maine is not sufficiently deep to permit the choice of a depth greater than 200 m for consideration. An examination of the samples collected periodically at greater depths shows only small fluctuation in oxygen content, and these do not appear to be related to the seasonal fluctuations observed at lesser depths. As a further check, the oxygen content of each 50-m length of the column has been estimated separately and the results plotted in Fig. 1. In the zone between 150 and 200 m the annual change in oxygen content is very small and amounts to only 10% of that of the entire 200-m column. Evidently failure to take account of seasonal changes which may occur at greater depths does not introduce significant errors.

A similar treatment of the phosphate data is shown in Fig. 2. The curve for the zone extending from the surface to 50 m shows a large seasonal fluctuation, which accounts for about 60% of the total seasonal change. The zone between 50 and 100 m also shows an increase in phosphate during the winter period. In the zones between 100 and 200 m there are fluctuations less clearly related to the general seasonal cycle and produced chiefly by low values obtained in January and April. Since there is no reason to think that phosphate concentrations would change in the deeper waters in this way, it is suspected that these fluctuations are due to systematic errors in the phosphate determinations during the cruises of January and April, such as might be occasioned by faulty standards. In order to avoid the multiplication of such errors, it has been assumed in the following consideration that the seasonal changes in phosphate below 100 m are negligible and that the best estimate of the changes in oxygen content due to biological activity are to be obtained from the phosphate content of the upper 100 m. Neglect of the changes in phosphate in the zone between 100 and 200 m reduces the estimated maximal seasonal variation of the column by only 20% and does not alter materially the general conclusions reached by this study.

Vertical mixing probably causes some exchange of oxygen and phosphate across the lower boundary. The deeper water of the Gulf has its origin through an inflow of slope water at the Eastern Channel, and

this water becomes fresher and colder as the western basin is approached, effects which can only be attributed to admixture with the overlying layers (Bigelow, 1927). It seems likely that there is a net upward movement of water as the result of this vertical exchange, in which an excess of deep water rises to escape from the Gulf in the surface currents across Georges Bank. The magnitude of this exchange cannot be evaluated and the phenomena require further study.

FIGURE 2. Seasonal variation of inorganic phosphate content of successive zones of 50 m depth beneath one m³ of sea surface in Gulf of Maine.

It is likely, however, that such exchanges are small compared to the seasonal effects under consideration.

RESULTS

Table I records the results of the computations. The values obtained for ΔQ_o , ΔQ_x , and ΔQ_B are shown plotted against time in Fig. 3.

TABLE I. OXYGEN AND PHOSPHATE CONTENT OF WATER BENEATH ONE SQUARE METER OF THE SURFACE OF THE GULF OF MAINE AND THE SEASONAL CHANGES ESTIMATED TO OCCUR RELATIVE TO THE CONDITIONS OBTAINING IN MARCH

	1	2	3	4	5	6
Period of	Q.	ΔQ.	PO.	APO.	10-	10-
Observation	104 cc	104 cc	9	g	104 cc	10º cc
July 6-10, '33	122.2	-9.3	7.79	-4.86	13.7	-23.0
Sept. 4, '33	115.9	-15.6	8.17	-4.48	12.6	-28 2
Oct. 28-29, '33	110.4	-21.1			(10 0)	(-31 1)
Dec. 2-4, '33	113.8	-17.7	10.17	-2.48	7.0	-24 7
Jan. 9, '34	126.1	-5.4	11.27	-1.38	3.9	-0.3
March 22, '34	131.5	0	12.65	0	0	0
April 18, '34	130.1	-1.4	9.18	-3.47	9.8	-11 2
May 22, '34	131.3	-0.2	7.77	-4.88	13.8	-14.0
June 26-July 1	125.1	-6.4	8.44	-4.21	11.9	-18.3
August 11, '34	119.9	-11.6			(12.5)	(-24 1)
Sept. 19, '34	113.0	-18.5	7.89	-4.76	13.4	-31.9

Column 1 Q., oxygen content of column 1 m² \times 200 m deep.

2 ΔQ_{\circ} , change in Q_{\circ} relative to March.

3 PO₄, inorganic phosphate content of column 1 m² \times 100 m deep.

4 ΔPO_4 , change in PO₄ relative to March.

5 ΔQ_3 , change in oxygen content, corresponding to ΔPO_4 , attributed to biological activity. It is assumed that ΔPO_4 and ΔQ_3 are negligible below 100 m.

 $\delta \Delta Q_B$, exchange of oxygen across 1 m² sea surface, relative to March.

Values in parentheses obtained by interpolation from curves in Fig. 3.

The annual change in the oxygen content of the water (maximum numerical value of ΔQ_o) is about 20 \times 10⁴ cc per m² of sea surface. The content is maximal in the latter part of March and minimal at the end of October.

The net annual change in oxygen content attributable to biological processes (maximum value of ΔQ_x) is about 13×10^4 cc per m² of sea surface. Oxygen production exceeds consumption during April and May. The processes are nearly balanced from June to September. Consumption exceeds production between October and March.

The annual exchange of oxygen across the sea surface (maximum numerical value of ΔQ_E) is about 30×10^4 cc per m². This quantity leaves the sea water during spring and summer and re-enters during the winter seasons. The turning points occur in the latter parts of October and March. The quantity of oxygen involved in the exchange is equal to that in a layer of air 1.5 m in depth.

FIGURE 3. Seasonal values of ΔQ_s , the observed change in oxygen content, and ΔQ_x , the estimated change in oxygen content due to biological activity in the water beneath one m³ of sea surface in Gulf of Maine, relative to the content observed in March. The curve ΔQ_s represents the quantities of oxygen estimated to have crossed one m³ of sea surface between March and the time of observation. Negative values indicate losses of oxygen from the water.

Of the annual exchange across the sea surface only two-fifths may be attributed to the net production or consumption of oxygen by organisms. The remainder presumably results from the effects of temperature changes on the solubility of oxygen in sea water.

Metabolic processes and solubility effects due to temperature supplement one another in producing the surface exchange. However, their relative importance differs with the season. In April and May practically all surface exchange is accountable by oxygen produced in photosynthesis. From June through September the metabolic processes are balanced and the continued surface exchange of oxygen accompanies the warming of the water. During the winter both processes contribute to maintaining the exchange; two-fifths of the oxygen absorbed from the atmosphere is consumed by organic activity and three-fifths held in solution by the cooling water.

THE OXYGEN PRESSURE HEAD ACROSS THE SEA SURFACE

Oxygen may be presumed to move across the sea surface as a result of the difference between the partial pressure of oxygen in the atmosphere and the tension, or escaping tendency, of the dissolved oxygen in the surface water.

If it is assumed that the air in contact with the sea surface is at the temperature of the water and is saturated with water vapor, the oxygen pressure of the atmosphere, P, will be 0.21 (1 - w) atmospheres where w is the tension of aqueous vapor. The oxygen tension of the water, p, is equal to the oxygen pressure of the atmosphere when the water is in equilibrium with the atmosphere or is "saturated." Under other conditions it follows from Henry's law that $p = P C_x/C_o$, where C_o is the oxygen content of sea water in equilibrium with one atmosphere of moist air and C_x is the oxygen content of the sample in question. The head of oxygen pressure across the sea surface is consequently given by

$$P - p = P(C_o - C_x)/C_o,$$
 (3)

when C_x is the oxygen content of water in the sea surface.

In order to evaluate P - p, data from the samples of water collected at a depth of one meter during each cruise have been averaged and the results are entered in Table II. In the direct estimation of Pfor application to the right-hand member of equation (3), allowance for the tension of water vapor in the atmosphere is inconsequential, since it reduces the value of P by only one or two per cent. In the estimation of $C_o - C_z$, however, it is important to take account of the water vapor in determining the value of C_o . Its value is proportional to P and will be reduced correspondingly as the vapor tension increases,

Period of	Temp.	Salinity	Р	С.	C.	P - p
Observation	°C	°/00	atm	cc/l	cc/l	atm
July 6-10. '33	15.18	31.96	0.206	5.83	6.12	-0.010
Sept. 4. '33	19.55	31.56	0.205	5.38	5.72	-0.013
Oct. 28-29, '33	11.02	32.18	0.207	6.32	6.26	0.002
Dec. 2-4, '33	7.11	32.86	0.208	6.84	6.65	0.006
Jan. 9. '34	5.24	32.96	0.208	7.14	6.69	0.103
March 22, '34	2.92	33.07	0.208	7.54	7.47	0.002
April 18, '34	4.72	32.75	0.208	7.24	7.65	-0.012
May 22, '34	9.19	31.65	0.208	6.59	7.27	-0.021
June 26-July 1, '34	14.95	31.93	0.207	5.89	6.28	-0.014
August 11, '34	18.45	31.96	0.205	5.49	5.89	-0.015
Sept 10 '34	17 65	32 12	0.206	5.56	5.88	-0.012

TABLE II.	ESTIMATES OF P - p, THE PRESSURE HEAD OF OXYGEN ACROSS SURI	ACE,
AND AVERAG	ED DATA FOR WATER AT 1 METER DEPTH ON WHICH ESTIMATE IS BA	SED

the reduction amounting to as much as 2% at the warmer seasons. Since the difference between C_o and C_x does not exceed 10% and at most seasons is 6% or less, neglect of the correction for vapor tension will lead to substantial error in estimating $C_o - C_x$.

To determine C_o , values in the table of Fox (1909) for the quantity of oxygen dissolved in sea water in equilibrium with dry air have been corrected to take account of the reduction in partial pressure of oxygen in an atmosphere saturated with water vapor. An alignment chart constructed from these values has been used to estimate C_o from the observed temperature and salinity of the water samples for which C_s was measured.

The estimate of the values P - p for the period of survey is plotted in Fig. 4. During the winter the pressure is positive, favoring a movement of oxygen from the atmosphere into the water. During the summer the pressure is reversed. The maximum positive value of 0.013 atmospheres was found in January, while the maximum negative value of 0.021 atmospheres occurred in May. The changes in direction come in October and March. These relations are consistent with the movements of oxygen across the sea surface deduced from measurements of oxygen and phosphate in the water column beneath the surface.

THE COEFFICIENT OF OXYGEN EXCHANGE

The rate of exchange of oxygen across the sea surface is defined theoretically by the equation

$$\frac{dQ}{dt} = E \cdot S \cdot (P - p), \tag{4}$$

where dQ/dt is the rate at which exygen enters the surface, S is the surface area and E is a constant, the exchange coefficient. E may conveniently be given the dimensions cc $O_2/\text{month} \times \text{m}^2 \times \text{atmosphere.}$

FIGURE 4. Seasonal variation in the pressure head of oxygen, P - p, across the surface of the Gulf of Maine. Negative values represent a pressure of oxygen in the water greater than in the atmosphere, a condition favoring the escape of oxygen from the sea water to the atmosphere.

Values of E may be estimated from equation (4), taking dQ/dt to equal the amount of oxygen entering the surface between successive times of observation divided by the time in months between the observations, and by expressing P - p by the average of its values at the beginning and end of the intervening period. These approximations will not lead to large errors for periods when ΔQ_B and P - p are relatively constant or are changing at nearly constant rates. The estimations are notreliable when the curve describing these functions are undergoing inflection, since at such times the average values may differ greatly from the properly weighted mean. Estimates of the value of E obtained in this way are given in Table III.

The three estimates made between the end of October and the latter part of March when P - p is positive, indicating a movement of oxygen into the sea water, yield an average value for E of 11.3×10^6 . The value for January 9-March 22 is suspect, because the average value of P - p is unreliable, being obtained when the curve is undergoing inflection. The two values obtained between October 29 and January 9 are in close agreement and give the most reliable estimate

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	1	2	3	4	5
Period of Observation	Δt	ΔQ	$\Delta Q/\Delta t$	(P - p)	E
July 8-Sept. 4	1.8	-5.2	-2.9	-0.0155	2.5
Sept. 4-Oct. 29	1.8	-2.9	-1.6	-0.0055	2.9
Oct. 29-Dec. 3	1.2	6.4	5.3	0.0040	13.2
Dec. 3-Jan. 9	1.2	15.4	12.8	0.0095	13.5
Jan. 9-Mar. 22	2.4	9.3	3.9	0.0075	5.2
Mar. 22-Apr. 18	0.9	-11.2	-12.4	-0.0050	24.8
Apr. 18-May 22	1.1	-3.8	-3.5	-0.0165	2.1
May 22-June 28	1.1	-4.3	-3.9	-0.0175	2.2
June 28-Aug. 11	1.5	-5.9	-3.9	-0.0145	2.7
Aug. 11-Sept. 19	1.3	-7.8	-6.0	-0.0135	4.4

TABLE III. ESTIMATES OF THE COEFFICIENT FOR THE EXCHANGE OF OXYGEN ACROSS THE SURFACE OF THE GULF OF MAINE

Column 1 Δt is interval between observations—in months.

2 ΔQ is quantity of oxygen entering surface during interval, *i. e.*, the change in ΔQ_B values of Table II. Unit: 10⁴ cc/m².

 $\Delta Q/\Delta t$ is mean rate of exchange during interval. Unit: 10⁴ cc/m² × months.

 $(P-p)_m$ is mean of values of oxygen pressure head P-p at beginning and end of interval. Unit: atmospheres.

5 E is exchange coefficient of oxygen. Unit: 10' cc/month \times m² \times atmosphere.

for E for the period when oxygen is entering the sea surface as 13×10^6 .

Of the estimates made during the remainder of the year, when P - p is negative, that for March 22-April 18 is unusually large. Its reliability may be questioned, because at that time the estimate of the oxygen exchange is based almost wholly on changes in the phosphate concentration of the upper layers of water. The resulting value for E is entirely subject to the errors in this estimation. The six remaining estimates are in fair agreement and yield an average value for E of 2.8×10^6 . The exchange coefficient for the period when oxygen is leaving the sea may be fixed approximately at this value.

These estimations indicate that the exchange coefficient of oxygen at the sea surface is greater during the winter period when oxygen is being taken up by the water than during the summer period when the movement of oxygen is in the opposite direction. The values for Efor these two periods appear to be approximately 13×10^6 and 2.8×10^6 respectively.

DISCUSSION OF THE COEFFICIENT OF OXYGEN EXCHANGE

The only pertinent data on the rate of movement of oxygen across an air-water interface of which I am aware are those of Bohr, Krogh, and Adeney and Becker. In studying the absorption of oxygen by small bubbles at 37° C, Krogh (1910) obtained a value of 0.0776 cc per minute per cm² per atmosphere, which, when converted to the dimensions in which E has been expressed, equals 33.5 x 10⁶ cc per month per m² per atmosphere. Adeney and Becker (1920) measured the quantity of oxygen leaving a relatively large bubble in its passage through unsaturated water. Estimates by Dorsey (1940: 552-555) based on their data give values of about 0.017 cc per minute per cm² per atmosphere or, when converted into the presently employed units, 7.3×10^6 cc per month per m² per atmosphere. Adeney and Becker's results indicate that the coefficient is about 5% lower for sea water and that it is not materially changed by temperature.

The present estimates for the sea surface agree in magnitude with these measurements. The values of Adeney and Becker are actually spanned by the estimates made for the summer and winter periods.

The difference in the value of the exchange coefficient between summer and winter is sufficiently distinct to suggest that some real differences in the conditions near the sea surface at these seasons affect the exchange. Before accepting this conclusion it is desirable to examine the assumption that the air immediately over the sea surface is saturated with water vapor. The introduction of the correction for water vapor causes the values of P - p to be smaller in winter and larger in summer than would be the case if the atmosphere were assumed to be dry or incompletely saturated. The values of E are consequently made larger in winter and smaller in summer. Thus the application of this correction might produce seasonal differences in the estimated value of E of the sort observed.

To determine whether the assumed saturation of the atmosphere could produce the seasonal difference obtained for E its values for the periods December 2-January 9 and June 26-August 11 have been calculated, assuming the atmosphere over the sea surface to be dry. The results give values of 10.5×10^6 for the former period and 3.7×10^6 for the latter. The results obtained, assuming a saturated atmosphere, were 13.5×10^6 and 2.7×10^6 respectively. It is evident that the difference in the winter and summer values of E cannot be attributed in any major degree to possible error in this assumption.

The possibility that the difference in the value of E depends directly on the seasonal difference in temperature is ruled out by Adeney and Becker's measurements on bubbles.

Two conditions suggest themselves in explanation of the seasonal difference in E. The values are calculated on the assumption of a flat sea surface of unit area. Waves, spindrift, and the bubbles entrained by white caps, which all tend to increase the air-water interface within a given area, are much more prominent in winter, favoring higher values of E at that time. Also, conditions near the sea surface in summer are much more stable than in winter. There is greater probability that the air immediately over the surface is saturated with

water vapor and that small scale gradients of oxygen concentration exist immediately beneath the sea surface. The use of oxygen analyses from samples collected at a depth of one meter in estimating the pressure of oxygen in the surface water leaves much to be desired. In winter, when unstable conditions favor boisterous mixing, this procedure is probably adequate, but in summer it is likely that the water at the sea surface is more nearly in equilibrium with the atmospheric oxygen than the sample collected. This appears to be the most important consideration in accounting for the seasonal difference in the

estimated values of the coefficient of oxygen exchange. Taken together the several factors considered are sufficient to account for the results.

SUMMARY

1. It is estimated that about 30×10^4 cc of oxygen per m² leaves the surface of the Gulf of Maine between the end of October and the latter part of March, a like amount re-entering from the atmosphere during the alternating period.

2. The net production and consumption of oxygen by organisms is sufficient to account for two-fifths of this exchange, the remainder being attributable to the effects of temperature on the solubility of oxygen.

3. In April and May the production of oxygen in photosynthesis is sufficient to account for the entire surface exchange; during the summer the exchange is attributable chiefly to decreasing solubility resulting from the warming of the water. In late fall and winter, excess oxygen consumption and increasing solubility are both responsible for the exchange.

4. The pressure head of oxygen across the sea surface has been determined and found to agree in direction with the estimated movement of oxygen.

5. The exchange coefficient of oxygen is estimated to be about 13×10^6 cc per m² per atmosphere per month in winter. In summer a value of 2.8×10^6 is obtained.

6. The seasonal difference in the values obtained is attributed to conditions at the sea surface, such as sea state and stability, which cause errors in the measurements and estimations.

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