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1 Introduction

A complete working model of the earth is still a rather distant goal. HOLLAND 1978

Those magnificent pictures of Earth from space, among the most humanly important and evocative results from the placing of manned and unmanned satellites in orbit during the last several decades, have shown us that underneath the clouds most of the world is blue. Viewed in this way from space, Earth is seen as a planet covered mostly by water, and most of that water is seawater.

This thin layer of water, covering 71% of Earth, affects or controls much of its climate and chemistry. The blue color of the sea tells us that most sunlight on the ocean is absorbed and not reflected. The absorption of sunlight warms the planet. The warmth evaporates water, especially from the tropical ocean. Water in the atmosphere is a greenhouse gas, and this also helps to warm Earth. Some atmospheric water forms clouds; by reflecting sunlight clouds help to cool Earth. The balance between heating and cooling is always changing as clouds form and dissipate, so exact calculation of the balance is difficult. The water in the ocean is a vast reservoir of heat, which buffers and slows global change. Both the currents in the ocean and the winds in the atmosphere carry heat from low latitudes, where there is a net input, towards the poles, where there is a net loss of heat to space. These vast currents, simple in general concept, immensely complex in detail, profoundly affect both the local distribution of climate and the overall climate of Earth. The ocean contains an enormous reservoir of carbon dioxide; it both absorbs and releases this greenhouse gas from and to the atmosphere. Living things in the sea influence both the release and the uptake. Physical, chemical, biological, and geological processes interact with each other in the ocean and with the atmosphere to create the environment of the planet we live on.

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Table 1.1 Volumes of water on Earth

	Volume; units of 10 ³ km ³	%
Seawater	1 332 000	97.4
Sea ice	20	—
Continental ice	27 800	2.0
Lakes and rivers	225	—
Groundwater	8062	0.6
Vapor (liquid volume)	13	_
TOTAL	1 386 120	100

From Baumgartner and Reichel 1975, Menard and Smith 1966, and Table J-6. The water chemically combined in crustal rocks and in the mantle is not included in these estimates.

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Earth surface area	$510 \times 10^6 \text{ km}^2$
Land surface area	$148 \times 10^6 \text{ km}^2$
Ocean surface area	$362 \times 10^6 \text{ km}^2$
Ocean surface area, % of Earth area	71%
Ocean: total volume	$1.33 \times 10^9 \text{ km}^3$
	$1.33 \times 10^{18} \text{ m}^3$
Average depth	3680 m
Temperature range	\sim -2 to \sim 40 °C
Pressure range	~ 1 to ~ 1000 atm
	~ 1 to ~ 1000 kg cm ⁻²
	~ 1 to ~ 1000 bar
	~ 100 to $\sim 10^5$ kPa

Menard and Smith 1966, Charette and Smith 2010. For more detail, see Appendix J.

1.1

Scope of chemical oceanography

Most of the water on Earth is within the province of the marine chemist, because over 97% of all (non-mineral-bound) water on Earth is in the sea, and most of the rest is frozen (Table 1.1). The maximum depth is a little over 10 km, so the pressures encountered in the ocean range from 1 atmosphere to about 1000 atmospheres, or roughly 100 to 100 000 kPa (Table 1.2). The marine chemist must therefore consider the effects of pressures within this range on chemical reactions within the sea.

The temperature of the open sea ranges normally from about $-2 \degree C$ to about $30 \degree C$, although in tropical bays and tide pools the temperature may reach or exceed $40 \degree C$, so most of the time a marine chemist must be concerned with temperature effects only within this range. The salt content varies from 0 (when, of course, it is not really seawater) to 41 g/kg (grams per kilogram = parts per thousand by weight, usually

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Table 1.3 Mean and percentile distribution^(a) of potential temperature^(b) and salinity in the major oceans of the world

Ocean	Mean	P _{5%}	P _{25%}	P _{50%}	P _{75%}	P95%	
Potential temperature, °C							
Pacific	3.36	0.8	1.3	1.9	3.4	11.1	
Indian	3.72	-0.2	1.0	1.9	4.4	12.7	
Atlantic	3.73	-0.6	1.7	2.6	3.9	13.7	
World	3.52	0.0	1.3	2.1	3.8	12.6	
Salinity, S							
Pacific	34.62	34.27	34.57	34.65	34.70	34.79	
Indian	34.76	34.44	34.66	34.73	34.79	35.19	
Atlantic	34.90	34.33	34.61	34.90	34.97	35.73	
World	34.72	34.33	34.61	34.69	34.79	35.10	

^(a) Percent of the total volume of the ocean with temperature and salinity characteristics falling below the indicated values ($P_{5\%}$, $P_{25\%}$, etc.). From Montgomery 1958.

^(b) When water is placed under pressure it is slightly compressed and its temperature rises; conversely, when it is decompressed its temperature drops. The temperature of seawater from various depths is usually expressed as the "potential temperature." This is the temperature it would have if brought to the surface without gain or loss of heat (i.e. it is decompressed adiabatically). Similarly, the potential density is its density when brought to the surface. (For special purposes the potential density is sometimes specified as that at some other standard depth.)

abbreviated as ppt or ‰; later we will learn a more precise definition of salinity); sometimes one may have to consider the more saline products of evaporation in closed basins. Other special circumstances are occasionally encountered, the most interesting and important being the occurrence of extremely hot seawater (sometimes greater than 400 °C) circulating through hot basaltic rocks exposed at the oceanic spreading centers and other regions of submarine volcanism.

Even the narrow temperature ranges, noted above as characteristic of the ocean, may give a misleading impression about the relative constancy of typical seawater (Table 1.3). Some 90% of the entire ocean volume is found between 0 and 12.6 °C, with a salt content ranging from 34.33 to 35.10‰. One-half of the entire volume of the ocean is characterized by temperatures between 1.3 and 3.8 °C and concentrations of salt between 34.61 to 34.79‰. The mean values for the world ocean are 3.52 °C and 34.72%. The observation that 90% of the total volume of seawater is found within a range in the concentration of salt only $\pm 1.1\%$ from the overall mean value posed an extraordinary challenge to analytical chemists during the early days of oceanography. A great wealth of information resides in the real patterns of differences in the content of salt and other substances from place to place throughout the mass of seawater, but that information could only be revealed by chemical measurements of extraordinary sensitivity and precision. The majority of analysts in most other fields are satisfied to achieve a two-standard-deviation accuracy and

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precision of $\pm 1\%$ of the measured value. However, in evaluating distributions of salt throughout the water masses of the oceans, data with only this precision would be essentially useless. In the ocean we need measurements of salinity that are at least 100 times better than this, with respect to both precision and accuracy.

The field of chemical oceanography is primarily concerned with the chemical and physical nature of seawater under the conditions and within the limits described above, with the chemical reactions which proceed in the medium, with the processes which control the composition of seawater, and with the processes by which the sea affects the atmosphere above and the solid earth below. To this end it is important to study the chemical nature of seawater itself, and the interactions of seawater with the atmosphere, with the sediments, with hot basaltic rocks, and with the organisms that live in the sea and to a great extent also control its chemistry.

We may look at the ocean in a variety of ways, and ask a variety of questions:

- Why is the ocean full of seawater? Where did it come from? How long will it last?
- Is the ocean the wastewater and salt of the whole world, accumulated for millions or billions of years, and is this question a useful approach to understanding the sea?
- Seawater is a very concentrated solution of some substances, and an extremely dilute solution of some others; in each case, what are the chemical consequences?
- The ocean is the environment for much of the life on Earth, and is probably where life began; to what extent is its chemistry today the product of life processes, and to what extent does the chemical environment in the sea control these processes?
- The oceans are the chemical environment through which and under which most of the world's sediment accumulation takes place. In addition, there are very important exchanges between hot basaltic rocks and seawater at the undersea spreading centers. To what extent are the chemistry of the sea, the nature of the sediments, and even the chemistry of the mantle, influenced or controlled by these chemical exchanges?
- The ocean is an integral part of the processes affecting the climate on Earth; to what extent does it both record and modulate those processes which tend to change the climate?

While, as Holland suggested, we may never have a complete working model of the Earth, it is nevertheless clear that many pieces of such a model are in place and pretty well understood, and the same is true of the sea. We know the sizes of some geochemical reservoirs very well, and most of the rest well enough. Many of the exchange processes are known, but in many cases the rates and what controls them are poorly known. We have little knowledge of changes in reservoir sizes through time, even less do we understand changes in the rates of important processes through time and with changing climate.

Our drive to study these matters is fueled in part by the necessity to examine the extent to which humans have caused, and continue to cause, changes in the geochemical transport rates of many substances and consequent changes in our environment. The waters of the ocean contain detectable records of some human activities over several decades, and perhaps longer. The sediments on the sea floor contain

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magnificent records of changing climate and other conditions on Earth over many tens of millions of years. We can test our understanding of natural processes by attempting to understand these records.

Because the chemistry of seawater plays such a central role in atmospheric, biological, and geochemical processes, the field of chemical oceanography overlaps many scientific disciplines and draws upon them for a variety of experimental and theoretical approaches to the study of the natural world.

History of chemical oceanography

What do we know, and when did we know it? After Howard Baker

In reading through some of the early records of observations on the chemical nature of seawater, a striking feature becomes apparent: Most of the names one comes across are already familiar because of their contributions to the foundation of the natural sciences or to the groundwork on which we base the modern study of chemistry.

For example, in the fourth century BCE, Aristotle (384-322 BCE) was interested in both the origin and the nature of seawater: "We must now discuss the origin of the sea and the cause of its salt and bitter taste." (Aristotle 1931, vol. III, p. 354^b). As we might expect, his explanations were not entirely clear. While he considered the words of Empedocles, that the sea is "the sweat of the earth," to be absurdly metaphorical, suitable for the requirements of a poem but unsatisfactory as a scientific theory, today we might still appreciate and use the metaphor, because at least some of the water in the sea must have been driven by heat out of the interior of the Earth. Aristotle did consider and use the analogy that the sweet water we drink becomes both salt and bitter as it gathers in the bladder as urine. He seemed convinced that water became salt through admixture with some earthy substance (vol III, p. 357^a). The bitter taste of sea salt or of seawater was noted again and again for well over 2000 years before the explanation was discovered. Aristotle was also aware of the relative density of seawater and fresh water. He reported a simple experiment showing that an egg would float in brine and sink in fresh water. Sailors of the time knew that ships floated higher in seawater than in fresh water, and indeed Aristotle reported that ships had for this reason sometimes come to grief upon sailing from salt into fresh water. The first recorded observations on the organic chemistry of seawater are also due to Aristotle. He noted that, especially in warm weather, "a fatty oily substance formed on the surface ... having been excreted by the sea which has fat in it." (Aristotle 1931, vol. VII, p. 932^b).

Aristotle's ideas on the hydrological and geochemical cycles were rather vague. He did, however, believe, apparently based on experimental evidence, that the vapors of sweet water arose from the sea, and fell again as rain, and he disputed the notion common at the time that there were subterranean passages by which seawater rose through the earth, perhaps straining out the salt by passing through soil, and discharging from springs over the land. In regard to the latter notion, it appears that

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a belief then current was that salt could be removed from seawater by passage through soil (we might now invoke the concepts of ion exchange). The Roman writer Pliny the Elder also stated that salt could be removed from seawater in this way. I have never seen any modern test of the suggestion, however. The notion of a subterranean return of water from sea to land was supported by the observation that in some places the springs from the ground yielded salty water. Aristotle did not dismiss the somewhat vague current notion that salt evaporated from the sea as a dry exhalation and mixed with rain to come down again. In fact, there is sea salt in rain, and one wonders if Aristotle could have had evidence of it.

Aristotle's largest contributions to marine science were his investigations in marine biology. Taking his contributions in total, however, Margaret Deacon (1971) wrote that Aristotle has a legitimate claim to the title "father of oceanography," although I am inclined to doubt that such a term has any real meaning. While certain additional facts about the chemistry of seawater were recorded by other ancient authors (such as Pliny the Elder), there were no real advances made for some 2000 years following Aristotle.

The next prominent person associated with the development of knowledge in this field was Robert Boyle (1627–1691). While Robert Boyle has been called the "father of chemical oceanography," he was also one of the founders of the Royal Society, the first scientific society, and he is more commonly called the "father of modern chemistry." Boyle added considerable weight to the notion that an element is "that which has not been decomposed." Robert Boyle is best known for his work in pneumatics and for the simple law relating the pressures and volumes of a gas at constant temperature $(P_1V_1 = P_2V_2)$, known as Boyle's law.

In 1673, Robert Boyle published a tract entitled *Observations and Experiments about the Saltness of the Sea*, in which he related experiments on the chemistry of seawater. As a result of some of the first real chemical tests, Boyle learned that even so-called fresh waters – river and lake waters – contain small amounts of salt, and therefore the salt in the oceans must be, at least in part, the accumulated washings from the land. In this he displayed an elementary perception of the geochemical cycle. No report on the matter having been published for 2000 years, Boyle demonstrated once again that sweet water could be obtained by evaporating seawater, and suggested that this could be of use to mariners who are short of water while at sea.

Boyle reported that seawater was of similar saltness at the surface and at depth. He recorded this observation after hearing reports from divers that the water tasted just as salty at depth as it did on the surface. He also analyzed "deep" samples collected in the English Channel with what must have been the first oceanographic sampling bottle, devised by Robert Hooke. Boyle was the first perceptive analyst of seawater, but he could not get reliable estimates of the salt in seawater by drying it and weighing the residue. As we will later see, this fact of nature is still true, and has caused an endless amount of trouble for oceanographers. He did report an approximate value of 30 g in a kilogram of seawater (in our units), nearly correct for that location. Some years after, Boyle was responsible for introducing the silver nitrate test for the saltness of water and we will later see what an important role that this chemical reaction has had in the development of modern oceanography. Boyle found that direct measurements of

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specific gravity were more reliable and consistent than measurements of the total salt content, by whatever method. During Boyle's time chemical knowledge was not sufficiently advanced that it was possible to determine the individual salts in seawater. Boyle used the word "saltness" to mean the concentration of salt in seawater, and this may be the first use of one word for that purpose.

Edmond Halley (1656–1742) was an English natural scientist of remarkable accomplishments. He was a mathematician, scientist, and navigator, and he became the Astronomer Royal of Great Britain. A friend of Isaac Newton, he urged Newton to publish the Principia Mathematica, edited the manuscript, and paid for its publication. His name lives on attached to "Halley's Comet," which he predicted would return each 76 years, and it did most recently in 1985–1986. Halley (1687) was the first to attempt a calculation of the hydrological balance for a part of the ocean. He measured the rate of evaporation of water from open pans and estimated that the Mediterranean must lose 53×10^9 tons of water per day in the summer and gain 1.8×10^9 tons per day from the inflowing rivers and rain. The difference between these numbers led him to comment that he might pursue further the question of what happens to the vapor, and the relation of this to the fact, known to mariners, that the current always sets inward through the Strait of Gibraltar; however, he never returned to the topic. His comments on this and other topics stimulated others to think about the world in rational and quantitative ways. Halley was perhaps the first person to think that the concentration of salt in the sea might be increasing with time, and that this increase might eventually be measurable. He urged that samples of seawater from various places be analyzed and the results recorded for comparison with analyses in future years (Halley 1715). Halley was concerned with the terribly difficult problem of determining the longitude of ships at sea, and commanded a vessel, the Paramore, on three voyages between 1698 and 1701 to plot the variations in the magnetic field over much of the North Atlantic. He wished to investigate whether the observed variations of magnetic north relative to the bearing to the pole star might provide a suitable method for determining longitude. These voyages were the first carried out purely for scientific studies of Earth.

Halley's influence extends on in other ways through more or less vague historical connections. He was concerned with making accurate measurements of the distance from Earth to the Sun, and in 1716 he wrote a paper calling attention to the value of making observations, at several locations around Earth, of the transits of Venus across the Sun, which he calculated were due in 1761 and 1769, long after he would have died. The British Admiralty fitted out the Transit of Venus Expedition in 1769 and sent it to the Pacific to make observations in the area around Tahiti. The name of the ship was the *Endeavour*, and the captain was James Cook. Some years later the *Endeavour* was sold into commercial service, and was sunk in the harbor of Newport, RI. The University of Rhode Island's research vessel, *RV Endeavor*, is named after Captain Cook's ship and carries as a talisman a piece of the wood originally believed to be from the original *Endeavour* (though now believed to be from Cook's other ship, *Resolution*, also sunk in Narragansett Bay [Abbass 1999]), and a sliver from *that* piece was carried by the space shuttle *Endeavour*.

It may not be possible to agree on who was the first true oceanographer, but one person who could claim the title was Count Luigi Ferdinando Marsigli (*aka* Louis

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Ferdinand Marsilli, 1658–1730). In 1725, he published the first book entirely devoted to oceanography: *Histoire Physique de la Mer*. This book was the first to include sections on the sea's basin, on the water in it, on the movements of the water, and on marine plants and animals. He had often measured the saltness of the water from various locations, and this book contains the first recorded tabulated data of such characteristics. He collected water from various depths to determine the saltness (he used the word "salure"), and also found a way to measure the temperature down to some tens of meters. His was the first evidence for a seasonal thermocline. Marsigli was the first person to apply color tests to determine the acid or basic quality of seawater, and reported that seawater was basic. He also extended Boyle's observations on the unreliability of measurements of the salt content of seawater made by weighing the residue left after evaporation. Marsigli was concerned about the problem of the bitter taste of seawater and sea salt and, like many at that time, believed it was due to the "bitumin" material present in seawater.

As a young man Marsigli had accompanied the Venetian envoy to the Ottoman court at Constantinople. There he interested himself in many aspects of that exotic place, including the two-layer flow of water in the Bosporus, which he learned about from talking to fishermen and ship captains. In 1681, he produced the first clear explanation of this important phenomenon (Soffientino and Pilson 2005, 2009). Interesting aspects of his life are given in Stoye (1994).

Antoine Laurent Lavoisier (1743–1794) was the first chemist to discard the phlogiston theory and to substitute for it the concepts of oxidation and reduction; he was the first chemist to write real chemical equations, and his experiments were superbly quantitative. Lavoisier can be regarded as the person chiefly responsible for setting chemistry on its modern path. About a hundred years after Boyle's pioneering work, Lavoisier (1772) attempted to analyze the major components of sea salt. He evaporated seawater, and extracted the dried residue with alcohol and with alcohol–water mixtures and recrystallized the salts from the resulting solutions. By this approach he was able to identify several products (expressed here in modern terms):

CaCO ₃	calcium carbonate
CaSO ₄ ·2H ₂ O	gypsum, calcium sulfate
NaCl	sodium chloride
$Na_2SO_4 \cdot xH_2O$	Glauber's salt, sodium sulfate
MgSO ₄	Epsom salt, magnesium sulfate
MgCl ₂	magnesium chloride

This early attempt to analyze such a mixture of salts showed the complexity of seawater as a chemical system and defined the approximate amounts of the major components. The following quotation (Lavoisier 1772) suggests both his sense of the geochemical cycle and the complexity of seawater as a chemical system:

The water of the sea results from a washing of the entire surface of the globe; these are in some ways the rinsing of the grand laboratory of nature; one therefore expects to find mixed together in this water all the salts which can be encountered in the mineral kingdom, and that is indeed what is found.

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Lavoisier was the first person to realize clearly that the bitter taste of seawater and of sea salt was not due to some bituminous substance, but rather to the presence of MgSO₄ or MgCl₂ ("sel d'Epsom" or "sel marin a base de sel d'Epsom").

Lavoisier also made significant contributions to geology, agriculture, public administration, and the creation of the metric system, and was perhaps the greatest scientist of the time. He was a political moderate, and supported the early goals of the French Revolution. Unfortunately, he was born to wealth, he had inherited a minor noble title purchased by his father, and he was a partner in the Ferme Général, a private company that had collected certain taxes for the government under the regime of Louis XVI. During the last excesses of the Revolution he was guillotined. At the time his friend, Joseph-Louis Lagrange, said: "It took but a moment to cut off that head, though a hundred years perhaps will be required to produce another like it."

Joseph Louis Gay-Lussac (1778–1850) is perhaps best known for Gay-Lussac's law (for every degree temperature increase, gases expand 1/273 of their volume at 0 °C), which made possible the early attempts at devising an absolute scale of temperature. Gay-Lussac is responsible for the observation that oxygen and hydrogen react to form water exactly in the proportions of one volume of oxygen to two volumes of hydrogen; careful measurements of this sort were fundamental to the development of the atomic and molecular theories. He introduced the verb "to titrate" and developed volumetric methods of standardizing acids and titrating solutions of silver salts. This latter development was of exceedingly great importance in subsequent analyses of seawater.

Gay-Lussac investigated the concentration of total salt in the ocean, and showed that, while variations may exist in near-shore waters, and small variations in the open ocean, in general the concentration of total salt is remarkably constant throughout the Atlantic. He understood that such small variations as there were must be due to variations in river runoff and to differences between evaporation and precipitation. He also understood that the ocean currents would tend to mix the water and minimize differences.

The idea of a "column" of seawater, so universally used today in the phrase "water column," seems to have been his: "la salure d'une colunne d'eau de l'Océan" (Gay-Lussac 1817). He also considered whether settling of the salts might cause deep water to be more saline than surface water, and addressed the matter experimentally by keeping salt solutions in 2-meter-high glass tubes for 6 to 20 months, and then analyzing the top and bottom (Gay-Lussac 1817, 1819). He found no difference.¹

¹ It is a minor curiosity of nature that salts could indeed be slightly more concentrated at the bottom of such a tube, where at equilibrium the gravitational tendency to settle is balanced against the effects of molecular diffusion. Pytkowicz (1962) calculated that the effect is about 0.18 km⁻¹, or 1.8×10^{-4} m⁻¹. If Gay-Lussac had started with seawater having a salinity of 35.000, and if he had been able to keep the tubes absolutely unstirred and with no convection for several years, the top of the tube might have had a salinity of 34.994 and the bottom a salinity of 35.006, a difference he could not have been able to measure. In the ocean the general water motions are much too great to allow the effects of molecular settling to become apparent, even in the most isolated basins.

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During the middle of the nineteenth century there was a remarkable development of analytical technique and of chemical understanding. For some investigators, seawater, because of its complexity, was even then regarded as a challenge to the analyst, so a considerable number of analyses were published. Alexander Marcet (1819), having analyzed samples from the North and South Atlantic, and the Mediterranean and other arms of the Atlantic, reported: "... all specimens of seawater which I have examined, however different in their strength, contain the same ingredients all over the world, these bearing very nearly the same proportions to each other; so that they differ only as to the total amount of their saline contents." This statement was "probably the first suggestion of the relative constancy of composition of seawater" (Wallace 1974), and the so-called Principle of Constant Composition is often called Marcet's Principle. In 1851, the German chemist Ernst von Bibra published analyses of five samples from the Atlantic, three from the Pacific and one from the North Sea (von Bibra 1851). All samples showed essentially the same relative proportions of the component substances.

The first book describing the physical characteristics of the ocean that achieved a large popular audience was Matthew Fontaine Maury's *Physical Geography of the Sea*, first published in 1855, widely read at the time, and influential in spreading oceanographic knowledge. The book was influential in part because of the fame of the author, who was well known and honored for his pioneering work (while superintendent of the US National Observatory) in compiling and publishing his wind and current charts of the oceans and accompanying sailing directions. These were of immense benefit to seamen of all nations, and to all sea-going commerce.

In his book Maury stated, probably on the evidence from von Bibra, that "if we take a sample of ... the average water of the Pacific Ocean, and analyze it, and if we do the same by a similar sample from the Atlantic, we shall find the analysis of the one to resemble that from the other as closely as though the two samples had been from the same bottle after having been well shaken." (Maury 1855, p. 152). He drew from this evidence the conclusion that the ocean currents must be sufficient to move and mix the whole ocean throughout the world, and expressed the idea succinctly as: "seawater is, with the exceptions above stated, everywhere and always the same, and ... it can only be made so by being well shaken together." (Maury 1855, p. 151.)

Since that time the notions of the general near-constancy of composition of seawater, of ocean mixing, and of the importance of ocean currents in carrying water around the world, have become generally appreciated. The total salt content and specific gravity were also known to vary regionally, but only to a very small extent. This principle of constant composition, however, was really established on a secure foundation by Johan Georg Forchhammer, a prominent Danish geologist and a superb chemical analyst (Forchhammer 1865). He believed that to understand the formation of sediments on the sea floor it was necessary to have an accurate knowledge of the chemical components in the water. He analyzed about 260 samples of seawater from all the major regions of the ocean and even the Caspian Sea. He introduced the practice of reporting a ratio of the concentration of each of the major elements to that of chloride, and demonstrated the remarkable constancy of these ratios. He introduced the use of the word "salinity" to characterize the total salt