See also

Air–Sea Transfer: Dimethyl Sulphide, COS, CS_2, NH_4, Non-methane Hydrocarbons, Organo-halogens; N_2O, NO, CH_4, CO. Atmospheric Input of Pollutants. Breaking Waves and Near-surface Turbulence. Bubbles. Surface Films. Surface, Gravity and Capillary Waves.

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AIR–SEA TRANSFER

Dimethyl Sulphide, COS, CS_2, NH_4, Non-methane Hydrocarbons, Organo-halogens

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The oceans, which cover 70% of Earth's surface to an average depth of 4000 m, have an immense impact on the atmosphere's dynamics. Exchanges of heat and momentum, water and gases across the sea surface play major roles in global climate and biogeochemical cycling. The ocean can be thought of as a vast biological soup with myriad processes influencing the concentrations of gases dissolved in the surface waters. The quantities of mass flux across the surface interface, though perhaps small on a unit area basis, can be very important because of the extent of the ocean surface and the properties of the gases or their decomposition products in the atmosphere.

Gas exchange across the sea–air surface depends, in part, on differences in partial pressures of the gases between the ocean surface and the atmosphere. The partial pressure of a gas in the gas phase can be understood in terms of its contribution to the pressure in the gas mixture. So the partial pressure of O_2, for example, at 0.21 atm means that at 1 atmosphere total pressure, O_2 is present as 21% of the gas, or mixing, volume. Trace gases are present in the atmosphere at much lower levels, usually expressed as parts per million (10^{-6} atm), parts per billion (10^{-9} atm) or parts per trillion (10^{-12} atm, pptv). Dimethylsulfide (DMS), when present at 100 pptv, accounts for about 100 molecules per 10^{12} molecules of mixed gas phase, or about 10^{-10} of the gas volume.

In solution, a dissolved trace gas in equilibrium with the atmosphere would have the same partial pressure as the gas in the air. Its absolute concentration in terms of molecules or mass per unit volume of water depends on its solubility. Gas solubility varies over many orders of magnitude depending on the affinity of water for the gas molecules and the volatility of the gas. Gases range widely in their solubility in sea water, from the permanent gases like nitrogen (N_2), oxygen (O_2), nitrous oxide (N_2O) and methane (CH_4) that have a low solubility in sea water to the moderately soluble carbon dioxide (CO_2) and dimethylsulfide $(CH_3)_2S$, to highly soluble ammonia (NH_3 and its ionized form NH_4^+) and sulfur dioxide (SO_2). Sulfur dioxide is more than 10^6 times more soluble than O_2 or CH_4. Using the example above of an atmospheric DMS concentration of 100 pptv, the equilibrium concentration of DMS in surface water would be about $0.07\,\mathrm{nmol\,l^{-1}}$. Generally the solubility of any individual gas increases at cooler water temperatures, and solubility of gases in sea water is somewhat less than for fresh water because of the so-called 'salting out' effect of dissolved species in sea water.

At any moment the partial pressure difference between surface water and the atmosphere depends on an array of variables. The gases in this article are biogenic, meaning that their mode of formation is the result of one or more immediate or proximate biological processes. These dissolved gases may also be consumed biologically, or removed by chemical processes in sea water, or they may flux across the sea surface to the atmosphere. The rates at which the source and sink processes occur determines the concentration of the dissolved gas in solution as well as the turnover, or residence time, of each compound. Similarly, there can be several source and sink processes for the gases in the atmosphere. Long-lived compounds in the atmosphere will tend to integrate more global processes, whereas short-lived compounds are concentrated near their source and reflect relatively short-term influences of source and sink. In this sense, carbonyl sulfide is a global gas. At it has a residence time of several years in the atmosphere, its concentration does not vary in the troposphere to any appreciable degree. On the other hand, the concentration of DMS varies on a diel basis and with elevation, with higher concentrations at night when atmospheric oxidants (most notable hydroxyl) are relatively depleted.

The extent of disequilibrium between the partial pressures of a gas in the surface water and in the atmosphere determines the thermodynamic gradient which drives gas flux. The kinetics of flux ultimately depend on molecular diffusion and larger-scale mixing processes. Molecular diffusivity is generally captured in a dimensionless parameter, the Schmidt number (ratio of viscosity of water to molecular diffusivity of gas in water), and varies widely between gases depending primarily on the molecular cross-section. From moment to moment, the flux of any particular gas is dependent on interfacial turbulence which is generated by shear between the wind and the sea surface whereby higher wind speed causes increasing turbulence and thus stimulating the onset of waves and eventually the production of bubbles and sea spray. There are considerable uncertainties relating gas exchange to wind speed. These arise due to the various sea-state factors (wave height, swell, breaking waves, bubble entrainment, surfactants, and others) whose individual dependencies on actual wind speed and wind history are not well quantified. The fluxes of gases across the air–sea interface are usually calculated using a wind-speed parameterization. These estimates are considered to be accurate to within a factor of 2 or so.

This article summarizes the characteristics of several important trace gases – dimethylsulfide, carbonyl sulfide, carbon disulfide, nonmethane hydrocarbons, ammonia and methylhalides – focusing on their production and fate as it is determined by biological and chemical processes.

Dimethylsulfide

Natural and anthropogenic sulfur aerosols play a major role in atmospheric chemistry and potentially in modulating global climate. One theory holds that a negative feedback links the emission of volatile organic sulfur (mostly as DMS) from the ocean with the formation of cloud condensation nuclei, thereby regulating, in a sense, the albedo and radiation balance of the earth. The direct (backscattering and reflection of solar radiation by sulfate aerosols) and indirect (cloud albedo) effects of sulfate aerosols may reduce the climatic forcing of trace greenhouse gases like CO_2, N_2O and CH_4. The oxidation products of DMS which also contribute to the acidity of rain, particularly in marine areas, result from industrialized and/or well-populated land.

Dimethylsulfide (DMS) is the most abundant volatile sulfur compound in sea water and constitutes about half of the global biogenic sulfur flux to the atmosphere. Studies of the concentration of DMS in the ocean have shown that average surface water concentrations may vary by up to a factor of 50 between summer and winter in mid and high latitudes. Furthermore, there are large-scale variations in DMS concentration associated with phytoplankton biomass, although there are generally poor correlations between local oceanic DMS concentrations and the biomass and productivity of phytoplankton (due to differences between plankton species in ability to produce DMS).

The nature and rates of the processes involved in the production and consumption of DMS in sea water are important in determining the surface concentrations and the concomitant flux to the atmosphere. The biogeochemical cycle of DMS (**Figure 1**)

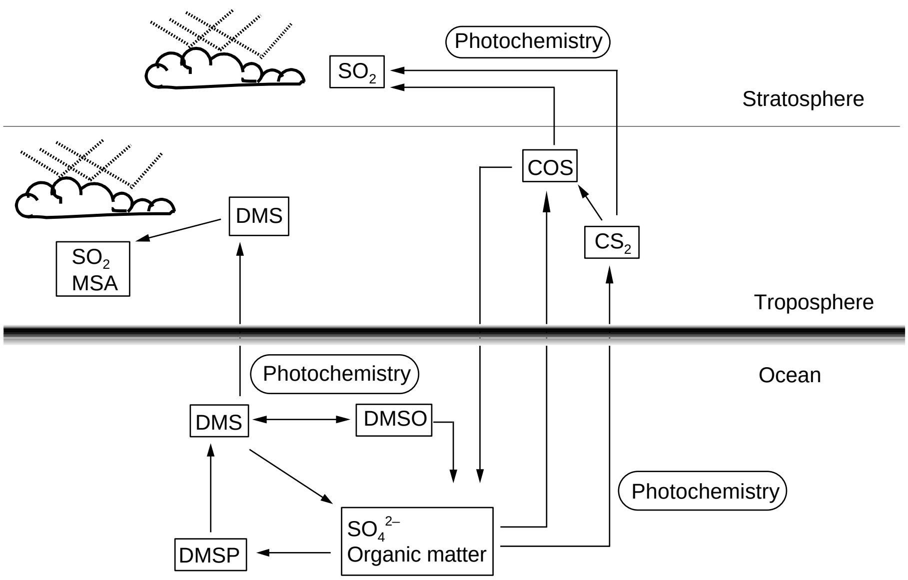

Figure 1 Fate and production of dimethylsulfide (DMS), carbonylsulfide (COS) and carbon disulfide. DMSO, dimethylsulfoxide; DMSP, dimethylsulfoniopropionate; MSA, methane sulfonic acid.

begins with its precursor, β-dimethylsulfoniopropionate (DMSP). DMSP is a cellular component in certain species of phytoplankton, notably some prymnesiophytes and dinoflagellates. The function of DMSP is unclear, although there is evidence for an osmoregulatory role as its cellular concentrations have been found to vary with salinity. It is generally thought that healthy algal cells do not leak either DMSP or DMS, although mechanical release into the surrounding sea water can lead to DMS production during cell senescence and grazing by zooplankton or as a consequence of viral attack on phytoplankton cells. Oceanic regions dominated by prolific DMSP-producing phytoplankton tend to have high DMS and DMSP concentrations.

Breakdown of DMSP, presumably after transfer from the particulate algal (pDMSP) form to a dissolved (dDMSP) form in sea water, can proceed in different ways, mostly depending on microbiological conditions. One major pathway involves cleavage of DMSP to DMS and acrylic acid. Bacterial metabolism of dDMSP may be a major mechanism for DMS production in sea water, with acrylic acid residue acting as a carbon source for heterotrophic growth. Sulfonium compounds are vulnerable to attack by hydroxide ion; the resulting chemical elimination reaction occurs rapidly and quantitatively in strong base but only slowly at the pH of sea water.

DMS in sea water has many potential fates. The volatility of DMS and the concentration gradient across the sea–air interface lead to the ocean being the major source of DMS to the atmosphere. Estimates of the annual sulfur release (as DMS) vary from 13–37 $\mathrm{Tg\,s\,y^{-1}}$ (Kettle and Andreae, 1999). However, whereas the absolute flux of DMS from sea to air may be large on a global scale, sea–air exchange may represent only a minor sink for seawater DMS. It has been estimated that DMS loss to the atmosphere is only a very small percentage of the DMS sink, but this undoubtedly depends on the biogeochemical conditions in the water column at the time. Photochemical oxidation of DMS, either to dimethylsulfoxide (DMSO) or to other products, occurs via photosensitized reactions. The amount of photochemical decomposition depends on the amount of light of appropriate wavelengths and the concentration of colored organic compounds in solution to convert light energy into reactive radicals. Light declines exponentially with depth; the distribution of colored dissolved organic materials exhibits depth and seasonal variability. Microbial consumption of DMS, although extremely variable in both time and space in the ocean, appears to be a significant sink for oceanic DMS. The residence time of DMS is probably of the order of a day or two in most seawater systems.

Since the atmospheric residence time of DMS is about a day or two, the atmospheric consequences of DMS flux are mostly confined to the troposphere. In the troposphere, DMS is oxidized primarily by hydroxyl radical. The main atmospheric oxidation products are methane sulphonic acid, SO_2 and DMSO.

Carbonyl Sulfide

Carbonyl sulfide (COS, OCS) is the major sulfur gas in the atmosphere, present throughout the troposphere at 500 pptv. COS has a long atmospheric residence time ($\sim$ 4 years). Because of its relative inertness COS diffuses into the stratosphere where it oxidizes to sulfate particles and contributes in reactions involving stratospheric ozone chemistry. Unlike DMS which is photochemically oxidized in the troposphere, the major sink for COS is terrestrial vegetation and soils. COS is taken up by plants by passing through the stomata and subsequently hydrolyzing to CO_2 and H_2S through the action of carbonic anhydrase inside plant cells. There is no apparent physiological significance to the process; it appears to just occur accidentally to the normal physiology of plants.

COS is produced in the ocean by photochemical oxidation of organic sulfur compounds whereby dissolved organic matter acts as a photosensitizer. The aqueous concentration of COS manifests a strong diel cycle, with the highest concentrations in daytime (concentration range on the order of 0.03–0.1 $\text{nmol}\,\text{l}^{-1}$). COS hydrolyzes in water to H_2S at rates dependent on water temperature and pH. The flux of oceanic COS to the atmosphere may represent about one-third of the global COS flux.

Carbon Disulfide

Concentrations in surface water are around $10^{-11}\,\text{mol}\,\text{l}^{-1}$. Although a number of studies have indicated that the ocean forms an important source for atmospheric CS_2, the underlying biochemical cycles still remain poorly understood. CS_2 is formed by photochemical reactions (possibly involving precursors such as DMS, DMSP and isothiocyanates). CS_2 formation has been observed to occur in bacteria in anoxic aquatic environments and in cultures of some marine algae species.

The residence time of CS_2 in the atmosphere is relatively short (about one week). Although CS_2 might contribute directly to SO_2 in the troposphere, its main significance is in the formation of COS via photochemical oxidation which results in the production of one molecule each of SO_2 and COS per molecule of CS_2 oxidized. The resulting COS may contribute to the stratospheric aerosol formation.

Concentrations around 14 $\text{pmol}\,\text{l}^{-1}$ of carbon disulfide in the mid-Atlantic Ocean were first observed (1974); higher concentrations have been found in coastal waters. More than a decade later CS_2 concentrations in the North Atlantic were found to be comparable to the earlier observations. However, in coastal waters CS_2 concentrations were found to be a factor 10 lower, respectively 33 and 300 $\text{pmol}\,\text{l}^{-1.}$ The global CS_2 flux has been estimated on 6.7 $\text{Gmol}\,\text{S}\,\text{y}^{-1}$, and it has been concluded that the marine emission of CS_2 provides a significant indirect source of COS, but it forms an insignificant source of tropospheric SO_2.

Nonmethane Hydrocarbons

Nonmethane hydrocarbons (NMHCs) are important reactive gases in the atmosphere since they provide a sink for hydroxyl radicals and play key roles in the production and destruction of ozone in the troposphere. NMHCs generally refer to the C_2–C_4 series, notably ethane, ethene, acetylene, propane, propene, and n-butane, but also the five-carbon compound isoprene. Of these, ethene is generally the most abundant contributing 40% to the total NMHC pool in sea water. Published data of concentrations of NMHCs in sea water vary widely sometimes exceeding a factor 100. For example, in one extensive study, ethene and propane were found to be the most abundant species in the intertropical South Pacific, with mixing ratios of 2.7 to 58 and 6 to 75 pptv, respectively; whereas in the equatorial Atlantic these species showed mixing ratios of 20 pptv and 10 pptv, respectively.

The water-column dynamics of NMHCs are poorly understood. NMHCs have been detected in the surface sea and with maxima in the euphotic zone and tend to be present at concentrations in sea water at around $10^{-10}\,\text{mol}\,\text{l}^{-1}$. Evidence suggests that photochemical oxidation of dissolved organic matter results in the formation of NMHCs. There can be very little doubt that the physiology of planktonic organisms is also involved in NMHC formation. Ethene and isoprene are freely produced by terrestrial plants where the former is a powerful plant hormone but the function of the latter less well understood. It is likely that similar processes occur in planktonic algae. NMHC production tends to correlate with light intensity, dissolved organic carbon and biological production. A simplified scheme of marine NMHC production is shown in **Figure 2**.

The flux of NMHCs to the atmosphere (with estimates ranging from $< 10\,\text{Mt}\,\text{y}^{-1}$ to $50\,\text{Mt}\,\text{y}^{-1}$) is minor on a global scale, but has a potential significance in local atmospheric chemistry. Although oceans are known to act as sources of NMHCs, the sources of individual NMHCs in the marine boundary layer are not always clear. Those NMHCs with a life time of more than a week (e.g., ethane, ethyne, propane, cyclopropane) show latitudinal gradients consistent with a continental source, whereas variations of NMHCs with life times shorter than a week (all alkenes and pentane) are more consistent with a marine source.

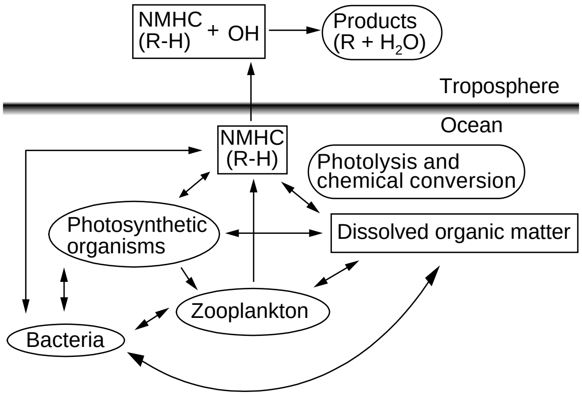

Figure 2 Simplified scheme of marine nonmethane hydrocarbon (NMHC) production. In the marine troposphere NMHC acts as a sink for hydroxyl (OH) radicals and thereby plays a key role in ozone chemistry.

Ammonia

Ammonia is an extremely soluble gas, reacting with water and dissociating into an ammonium ion at ambient pH. At pH 8.2, about one-tenth of dissolved ammonia is present as NH_3. Ammonium is also a rapidly cycling biological nutrient; it is taken up by bacteria and phytoplankton as a source of fixed nitrogen, and released by sundry physiological and decompositional processes in the food web. Anthropogenic loading of ammonium (and other nutrients) into the coastal marine environment results in increased phytoplankton growth in a phenomenon called eutrophication. Ammonium is oxidized to nitrate by bacteria in a process known as nitrification (**Figure 3**). Conversely, in anoxic environments, ammonium can be formed by nitrate-reducing bacteria.

Ammonia plays an important role in the acid–base chemistry in the troposphere where the unionized ammonia (NH_3) is converted into ionized ammonia (NH_4^+) via a reaction that neutralizes atmospheric acids as HNO_3 and H_2SO_4. This leads to the formation of ammonium aerosols such as the stable ammonium sulfate. Eventually the ammonia returns to the surface by dry or wet deposition.

Few data exist on the fluxes of NH_3 over marine environments. Evidence suggests that most of the ocean surface serves as a source of NH_3 to the atmosphere, even in regions of very low nutrients. In the North Sea, an area situated in the middle of densely populated and industrialized countries of Western Europe, air from nearby terrestrial sources may act as a source of NH_3 into surface waters. It has been estimated that the annual biogenic emission of ammonia from European seas is around $30\,\mathrm{kt\,N\,y^{-1}}$, which is comparable to the emissions of smaller North European countries, leading to the

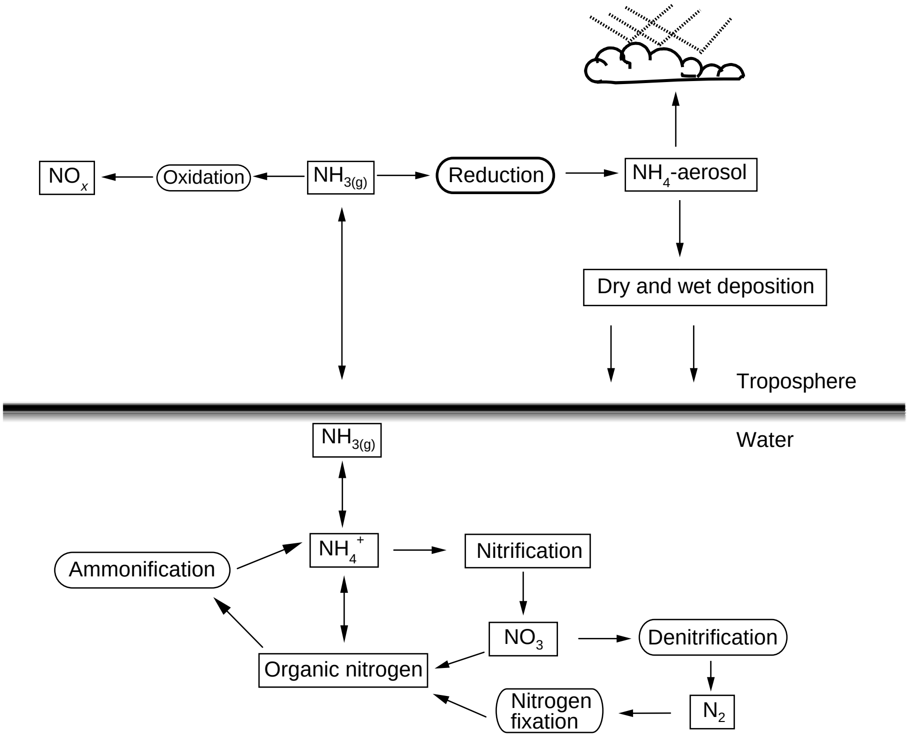

Figure 3 Simplified scheme of marine NH_x chemistry. In the marine boundary layer NO_3 acts as an initiator for the degradation of many organic compounds, in particular dimethylsulfide (DMS).

conclusion (amongst others) that seas are among the largest sources of imported ammonium for maritime countries. The net emission of ammonia from coastal waters of the north-east Pacific Ocean to the atmosphere has been shown to be in the order of $10\,\mu mol\,m^{-2}\,d^{-1}$.

Organohalogens

Halogenated compounds, such as methyl chloride (CH_3Cl), methyl bromide (CH_3Br) and methyl iodide (CH_3I) are a major source of halogens in the atmosphere, and subsequently form sources of reactive species capable of catalytically destroying ozone. Among these CH_3I is likely to play an important role in the budget of tropospheric ozone, through production of iodine atoms by photolysis. Due to their higher photochemical stability methyl chloride and methyl bromide are more important in stratospheric chemistry; it has been suggested that BrO species are responsible for losses of tropospheric ozone in the Arctic (**Figure 4**).

Atmospheric methyl halides, measured over the ocean by several cruise surveys, have been shown to have average atmospheric mixing ratios of: CH_3Cl, 550–600 pptv; CH_3Br, 10–12 pptv; CH_3I, 0.5–1 pptv. Their temporal and spatial variations are not well understood, neither is their production mechanism in the ocean known. Measurements of atmospheric and seawater concentrations of CH_3Cl and CH_3I have indicated that the oceans form natural sources of these methyl halides. In contrast, CH_3Br appears to be undersaturated in the open ocean and exhibits moderate to 100% supersaturation in coastal and upwelling regions, leading to a global atmosphere to ocean flux of $13\,Gg\,y^{-1}$. Coastal salt marshes, although they constitute a minor area of the global marine environment, may produce roughly 10% of the total fluxes of atmospheric CH_3Br and CH_3Cl and thus contribute significantly to the global budgets.

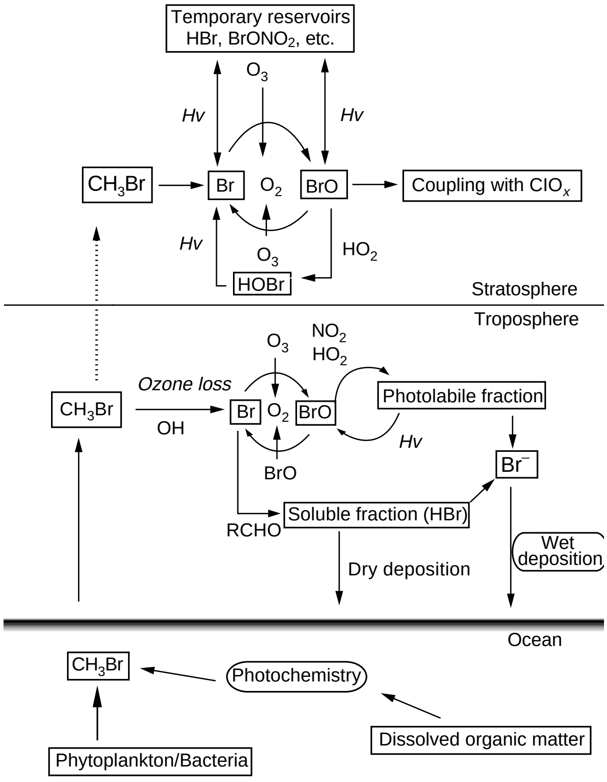

Figure 4 Schematic illustration of the circulation of methyl bromide.

Macrophytic and phytoplanktonic algae produce a wide range of volatile organohalogens including di- and tri-halomethanes and mixed organohalogens. There is evidence for the involvement of enzymatic synthesis of methyl halides, but the metabolic production pathways are not well known. In free sea water, photochemical processes, ion substitution and, possibly the alkylation of halide ions (during the oxidation of organic matter by an electron acceptor such as Fe(III)) are also potential formation mechanisms. Sunlight or microbial mediation are not required for these reactions. In the ocean, chemical degradation of CH_3Br occurs by nucleophilic substitution by chloride and hydrolysis. Microbial consumption is also a likely sink for halogenated compounds.

Conclusions

The biogenic trace gases are influenced by the complete range of biological processes – from the biochemical and physiological to the ecological level of food web dynamics. The gases that are influenced directly by plant physiology (probably the light NMHCs and isoprene, for example) tend to be most closely related to phytoplankton biomass or primary productivity. Other gases produced during grazing and decomposition (e.g., DMS, NH_3), or gases formed by photochemical reactions in dissolved organic material show differing temporal dynamics and different dynamics spatially and with depth in the ocean. As conditions change in an apparently warming world, changes in the dynamics of surface ocean gases can be expected. The behavior of these trace gases or even the dynamics of the planktonic community are not understood sufficiently to allow good quantitative predictions about changes in trace gas flux to be made. Changes in flux of some gases could lead to an acceleration of warming, while changes in others could lead to cooling. It is, thus, important to understand the factors controlling trace gas dynamics in the surface ocean.

See also

Air–Sea Gas Exchange. Air–Sea Transfer: N_2O, NO, CH_4, CO. Atmospheric Input of Pollutants. Carbon Cycle. Chlorinated Hydrocarbons. Nitrogen Cycle.

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N_2O, NO, CH_4, CO

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Introduction

The atmospheric composition is maintained by abiotic and biotic processes in the terrestrial and marine ecosystems. The biogenic trace gases nitrous oxide (N_2O), nitric oxide (NO), methane (CH_4) and carbon monoxide (CO) are present in the surface mixed layer over most of the ocean, at concentrations which exceed those expected from equilibration with the atmosphere. As the oceans occupy 70% of the global surface area, exchange of these trace gases across the air–sea interface represents a source/sink for global atmospheric budgets and oceanic biogeochemical budgets, although marine emissions of NO are poorly characterized. These trace gases contribute to global change directly and indirectly, by influencing the atmospheric oxidation and radiative capacity (the 'greenhouse effect') and, together with their reaction products, impact stratospheric ozone chemistry (**Table 1**). The resultant changes in atmospheric forcing subsequently influence ocean circulation and biogeochemistry via feedback processes on a range of timescales. This article describes the marine sources, sinks, and spatial distribution of each trace gas and identifies the marine contribution to total atmosphere budgets. There is also a brief examination of the approaches used for determination of marine trace gas fluxes and the variability in current estimates.

Nitrous Oxide (N_2O)

The N_2O molecule is effective at retaining long-wave radiation with a relative radiative forcing 280 times that of a CO_2 molecule. Despite this the relatively low atmospheric N_2O concentration results in a contribution of only 5–6% of the present day 'greenhouse effect' with a direct radiative forcing of about $0.1\,\mathrm{W m^{-2}}$. In the stratosphere N_2O reacts with oxygen to produce NO radicals, which contribute to ozone depletion.

N_2O is a reduced gas which is produced in the ocean primarily by microbial nitrification and denitrification. N_2O is released during ammonium