Chapter 1

Biogenic Sulfur Emissions

A Review

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Biogenic sulfur emission rates are reviewed for important components of the sulfur cycle. A summary of emission estimates is provided for vegetation, coastal and wetland ecosystems, inland soils, and oceanic environments. One area which is briefly reviewed, emissions from plants, may play a significant role in global sulfur cycling and very little work has been reported covering this subject. An important trend is that estimates of biogenic emissions are being lowered for terrestrial and wetlands areas. This, coupled with decreased wetland acreage, may significantly decrease local estimates of biogenic sulfur to acid precipitation.

Sulfur in the atmosphere originates either from natural processes or anthropogenic activity. The natural biogenic sources are thought to constitute a large fraction (estimates as high as 50 % have been reported) of the atmospheric sulfur burden (1-8). As such, these natural sources may have a substantial impact on global sulfur cycling.

Biogenic sulfur, that is sulfur compounds which result from biological processes, are only one component of the natural sulfur cycle. The first measurements of biogenic sulfur fluxes were those of Aneja and co-workers (9,10). Numerous studies have been published in the last decade which add to our understanding of the natural sulfur emissions (11-29). This research has provided experimental data, which is helpful in refining the biogenic emission estimates necessary to refine the global sulfur cycle. This data base, although sparse for detailed quantitative estimates, is helpful for estimating emissions from both terrestrial and oceanic environments.

In the initial attempts at developing global sulfur budgets, biogenic emissions were usually obtained from the amount of sulfur necessary to balance the cycle. This resulted in considerable scatter in the biogenic estimates, from 34 Tg S yr^{-1} (4) to 267 Tg S yr^{-1} (7), where Tg = 10^{12} g. It is possible with the existing data to begin to make estimates of biogenic emissions based on direct measurements. However, additional data are necessary to assess biogenic sulfur emissions independent of other portions of the global sulfur cycle.

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The objectives of this overview chapter are:

1. To review the extant data base of biogenic sulfur emissions for terrestrial and oceanic environments and to summarize direct estimates of emissions where possible.
2. To integrate the other chapters of this section into the discussion.

Biogenic Sulfur Emissions from Vegetation

It has been recognized for some time that sulfur is essential for plant growth. It is used in amino acids, and many other biochemicals. The biological transformation of sulfur compounds in natural ecosystems is closely coupled to the formation of living biomass and to the subsequent decomposition and remineralization of the biomass. Plants contain an average sulfur content of 0.25 % (dry weight basis) (30). Hence sulfur may be released directly from vegetation or during the process(es) of decomposition of the organic matter. Data on sulfur released by vegetation is scanty and the mechanism(s) of release are not known in any detail. A summary of available data is provided below and a more detailed discussion follows in the next chapter (31).

Sulfur compounds are known to be volatilized from living plant leaves (32), and from decaying leaves (33). It has been estimated that sulfur emission rates from decaying leaves are about 10 to 100 times higher than those from living leaves of the same species (33). Many fungi and bacteria release sulfur compounds (34) during plant decomposition. Some plants are known to emit H_2S (11,29,31,35,36). Emission rates of H_2S from several lawns and from a pine forest on aerobic soils, in France, ranged from 0.006 to 0.25 g S m^{-2} yr^{-1} (35). However, in the Ivory Coast, West Africa, emission rates of H_2S from humid forests ranged between 0.24 and 2.4 g S m^{-2} yr^{-1} (36).

Some plants are also known to emit dimethyl sulfide, DMS, (13,37), carbonyl sulfide, COS, (29), and carbon disulfide, CS_2, (13,37-41), and possibly ethyl mercaptan (40,41). A study conducted in a tropical rain forest which focused on *Stryphnodendron excelsum* is trteated in more detail in a following chapter (41). It is quite possible that additional studies as described in Chapter 5 (41) will lead to the discovery of other terrestrial "hot spots" which may be important in biogenic sulfur cycling.

Emission rates of sulfur from crops including corn, soybeans, oats, alfalfa, and miscellaneous vegetables have been measured (27-29). The flux from crops range from 0.008 to 0.3 g S m^{-2} yr^{-1}. H_2S and DMS are the two primary sulfur species being emitted by crops. Emission rates of sulfur from a variety of plants are summarized in Table I.

It is also possible that plants emit volatile sulfur containing compounds which are not easily analyzed by current gas chromatographic methods. Thus, the use of other analytical methods may reveal compounds as yet unidentified which serve as a source of volatile biogenic sulfur compounds.

Biogenic Sulfur Emissions from Wetlands

The tidal flats of marine environments are areas of extreme complexity and biological activity. They serve as both sources and sinks of a wide variety of compounds and materials. They are in a constant state of mass, energy and momentum flux with the surrounding environment. In these areas sulfur plays a major role in biological processes, principally because of the relatively high concentration of sulfate ion in marine waters.

Sulfate ion is the major electron acceptor for respiration in anoxic marine sediments and may account for 25 % of the total sediment respiration in near shore sediments, 0.3 to 3 g C m^{-2} day^{-1}. In salt marsh sediments, where total

Table I. Biogenic Emissions of Sulfur From Vegetation

Plant	Mean Sample Temperature (°C)	Primary Sulfur Species	Emission Rate g S m^{-2} yr^{-1}	Ref.
Spartina				
alterniflora, N.C.	30	DMS	0.66	(12)
S. alterniflora, N.C.	25	CS_2	0.20	(13)
Stryphnodendron				
excelsum, Costa Rica	NR*			(41)
4 meters from tree trunk		CS_2	1.5	
		H_2S	0.022	
16 meters from tree trunk		CS_2	0.15	
		H_2S	not detected	
Lawn, France	22	H_2S	0.24	(35)
Pine Forest, France	10	H_2S	0.023	(35)
Humid Forest,				
Ivory Coast	25	H_2S	0.88	(36)
Crops				
Oats (with soil), IA	35.4	DMS, H_2S	0.016	(29)
Corn, IA, OH	28.9	DMS, H_2S	0.032	(29)
Soybeans, IA	32.8	DMS, H_2S	0.066	(29)
Alfalfa, WA	22.4	DMS, H_2S	0.056	(29)
Trees				
Deciduous, IA, OH, NC	29.5	DMS, H_2S, COS	0.007	(29)
Coniferous, NC	29.2	COS	0.005	(29)
Crops				
Soybeans, IA	25.5		0.0018	(27)
Carrots, OH	22		0.113	(27)
Onions, OH	22		0.104	(27)
Grass, IA	25.5		0.018	(27)
Crops				
Soybeans,	30	DMS	0.037	(28)
Oats,	30	DMS	0.023	(28)
Orchard Grass,	30	DMS	0.008	(28)
Purple Clover,	30	DMS	0.007	(28)
Corn,	30	DMS	0.273	(28)

*NR = not reported

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respiration rates may be 2.5 to 5 g C m^{-2} day^{-1}, sulfate ion respiration may account for up to 90 % of the total (e.g. 19,23). Detailed discussions of the processes and variables affecting H_2S emissions may be found in other papers (e.g. 19,23,26,42-46), and in other chapters of this volume (47-50).

There have been a number of studies of biogenic emissions of sulfur gases other than H_2S reported in the literature. Most of these have been concerned with high productivity sources, such as salt marshes and tidal areas and are summarized in Table II.

There are a number of factors which affect the emission rates of biogenic sulfur from wetlands. In a recent study these have been investigated for wetlands in Florida, USA, (57-59) and are summarized in a chapter in this volume (60). These factors are divided into spatial, seasonal, diel and tidal components. In addition, other variables which affect emissions are temperature, insolation, and soil inundation. When these factors are taken into account in estimating emissions, and emission rates are obtained by integrating over the appropriate cycle, the emission estimates are up to two orders of magnitude lower than earlier estimates. However, using these methods results in large uncertainties in the emission estimates, and considerable additional data are required to better refine and extend emission estimates to other environments.

Biogenic Emissions From Land

Sulfur flux measurements from various locations are summarized in Table III. In three inland soils in France the H_2S flux data ranged from 0.19 to 0.24 g S m^{-2} yr^{-1} (35,61). In a broad and diverse inland study area in the U.S. 27 soils were examined, and total sulfur flux reported from 0.013 to 0.33 g S m^{-2} yr^{-1}. The primary sulfur species was H_2S and the flux ranged from undetectable to 0.16 g S m^{-2} yr^{-1}. However, several other gases, DMS, CS_2 and COS were observed in some locales.

Several recent studies have reported additional data for the emission rates of biogenic sulfur species. Lamb and co-workers (29) measured emissions rates in several regions of the U.S. and observed H_2S, COS and DMS, during the summer of 1985. The total flux of the sulfur species can be summarized for two soils, mollisol and histol, averaging 0.008 and 0.114 g S m^{-2} yr^{-1}, respectively.

Goldan and co-workers (28) measured sulfur fluxes from bare soils at two mid-continent sites, also during the summer of 1985. The principal sulfur species were COS, H_2S, DMS, and CS_2, all of which were strongly correlated with air temperature. The emission rate of the sulfur species ranged from 0.003 to 0.008 g S m^{-2} yr^{-1}.

An important trend is becoming evident as additional data is being published. The trend is that recent terrestrial emission estimates appear to be greater than twenty times lower than reported in earlier studies (15,16,37,63). This trend is discussed in more detail in Chapter 2 (64), as are some of the possible explanations for the differences.

Lower terrestrial and coastal emission estimates combined with the increasing loss in wetlands (65-68), although it may not significantly impact the global sulfur cycle, may be an important consideration in local contributions of natural emissions to acid precipitation.

Biogenic Emissions for Oceanic Environments

This topic is included in order to complete a discussion of biogenic sulfur emissions. The reader is referred to more comprehensive reviews in this book and other sources for more details (e.g. 69-73).

Table II. Biogenic Emissions of Sulfur Compounds from Coastal Ecosystems

Source	Month of	Emission Rate ($g\ S\ m^{-2}\ yr^{-1}$)						Ref.
	Year	H_2S	DMS	DMDS	CH_3SH	CS_2	COS	
Salt Marsh NY	10/11	0.55	0.15	0.018	0.064	-	-	(9,10)
Swamps and tidal flats, Denmark		0.044	-	-	-	-	-	(17)
Coastal area Denmark		~19	-	-	-	-	-	(51)
Salt marsh N. Carolina	7/8/9	0.5	0.66	-	-	0.2	0.03	(12,13,52)
Salt Marshes N. Carolina	5/7/10	0.033	0.538	0.0005	0.00026	0.035	0.012	(15,16)
Delaware	8	0.096	0.48	0.00053	-	0.07	0.012	
Massachusetts	8	-	0.06	0.006	-	0.028	0.004	
Virginia		-	1.87	0.04	0.22	1.38	0.03	
Salt marsh Virginia	8	9.5	-	-	-	-	-	(53)
Salt marsh Virginia	8/9	0.0013	-	-	-	-	-	(54)
Salt marsh Massachusetts	1 yr.	2.05	2.88	0.42	-	0.16	0.3	(55)
Salt marsh N. Carolina	8	0.5	-	-	-	-	-	(26)
Salt marsh N. Carolina	8	0.33	0.083	0.00064	-	0.0017	0.052	(29)
Salt marsh N.Carolina		0.05	0.1	-	0.0037	0.0032	0.0042	(28)

Table III. Biogenic Emissions of Sulfur Compounds from Inland Soils

Source	Month of	Emission Rate ($g\ S\ m^{-2}\ yr^{-1}$)						Ref.
	Year	H_2S	DMS	DMDS	CH_3SH	CS_2	COS	
Equatorial forest Ivory Coast	1/10 11/12	0.07	-	-	-	-	-	(35)
Lawn, France	4/5/6/11/12	0.044	-	-	-	-	-	(35)
Dry inland soil N. Carolina	8/9/10	<0.01	<0.01	<0.05	<0.05	<0.05	<0.01	(62)
Mollisol, Iowa	7	0.15	0.0032	-	-	0.017	0.017	(15,16)
Histosol, Ohio	7	0.047	0.0032	-	-	0.006	0.012	(15,16)
Inceptisols, Ohio	7	0.04	0.002	0.0007	0.001	0.01	0.003	(15,16)
Mollisol, Iowa	7	0.0003	0.0005	-	-	0.0003	0.0015	(28)
Histisol, Ohio	7	0.0024	0.0003	-	-	0.0007	0.0036	(28)
Mollisol, Indiana	7	0.0002	0.0005	-	-	0.00023	0.0029	(29)
Histisol, Ohio	7	0.009	0.0004	-	-	0.00004	0.015	(29)

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Oceans and the marine environment are the major source of biogenic sulfur. The reasons for this are the generally abundant phytoplankton in surface oceans and the areal extent of these waters. A summary of the emission rates of DMS, the major biogenic sulfur species in marine environments, is presented in Table IV. Estimates are included which were determined directly and from model calculations.

The first report of DMS in the ocean appeared in 1972 (33). The authors suggested that DMS might be more important than H_2S as a biogenic sulfur source for balancing the global sulfur budgets. Preliminary estimates of DMS sea-to-air flux based on the limited data were made by Liss and Slater (74).

DMS is produced in oceanic waters by benthic and to a greater extent by planktonic marine algae (78), suggesting that it is ubiquitous in the surface ocean (20,69-71,75,79,80). Its distribution has been characterized by "hot spots" with high DMS concentrations superimposed on more or less constant level of approximately 1-3 nM. These high concentrations, hot spots, may be the result of blooms of e.g. *Phaeocystis poucheti*, which are known to produce DMS. The most comprehensive survey relating DMS and its precursor DMSP (dimethyl-propionosulfonate) to marine phytoplankton appears in another section of this book (81), with over 120 phytoplankton clones having been surveyed.

Estimates of DMS flux from oceanic environments have resulted from direct measurement of DMS in surface waters (22) and from model calculations (76,77). At this time it is not possible to determine which of the numbers, if any, are correct, however, there is little doubt that the marine environment is one of the major sources of biogenic sulfur.

There is considerable debate regarding the marine environment and the possibility of it being a major source of H_2S. In a coastal area, integrated over tidal and diel cycles, a rate of H_2S of 0.10 g S m^{-2} yr^{-1}, has been reported (26).

Table IV. Biogenic Emissions of DMS from Open Oceans and the Marine Environment

Environment	Emission (g S m^{-2} yr^{-1})	Reference
Open Ocean*	0.02	(74)
Ocean	0.075	(75)
Ocean	0.106	(22)
Ocean*	0.022	(76)
Gulf of Mexico*	0.037	(77)

*Based on model calculations

Emission Flux Measurement Methods

To complete the overview on sulfur emissions, a brief discussion of methods used to estimate emissions is appropriate. There are primarily two methods that may be used to measure earth-atmosphere flux of gases. In the dynamic chamber method, an open-bottom chamber is placed over a surface of interest such as mud, soil, or water, with or without vegetation, to capture the gases

emanating from the surface. A carrier gas is introduced into the chamber and mixed with the natural gases. The carrier gas is usually, but not necessarily free of the species being detected. The effluent gas from the chamber is sampled and analyzed for the compounds of interest and the flux is estimated by mass balance. While this technique is easy to use, there are indications that care should be taken to minimize the changes which the chamber itself may exert upon the emitting surface (82).

The second method is the micrometerological method (vertical gradient). The concentration of the gas of interest is measured at various altitudes above the source along with the wind speed and direction. To determine the vertical concentration profile, samples obtained simultaneously at various elevations on a tower must be analyzed. This requires the ability to determine very precisely small differences in concentrations (at low ambient concentrations) among the vertical samples. Estimates of flux are made by applying turbulent diffusion theory to the concentration profile data. This method, although reasonably simple in concept, is very difficult in practice and requires considerable supporting micrometerological data.

Summary

The goal of developing estimates of biogenic sulfur emissions, to provide data for use in estimating regional and global fluxes of biogenic sulfur to the atmosphere, based upon direct measurements is far from being accomplished. With improved analytical methods and a better understanding of the factors affecting biogenic emissions improved estimates are being obtained. Biogenic emissions remain a major area of interest where information is required on anthropogenic impacts on ecosystems and for refining models of global sulfur cycling.

Future studies of biogenic emissions should be designed to include sufficient data for uncertainty analyses of flux estimates. It is also important to conduct intercomparisons of different sampling and measurement methods, as well as the methods used for estimating emissions, i.e. the dynamic chamber vs the micrometerological methods. Additional data are required to confirm emission estimates for those environments which have been characterized, and to extend the emission estimates to environments which have not been studied. Studies in ecosystems such as tropical rain forests should include surveys designed to identify potential terrestrial "hot spots" of volatile sulfur emissions.

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