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Review Article

A Review on Advanced Studies of the Sulfur Biogeochemical Cycle in the Coastal Wetlands

Ayyaraju Middepogu¹, Gousiya Begum Shaik¹, Suresh Kumar Chitta¹, Anuradha Chevva Moremmagari*², and Daulin Du³

¹Department of Biochemistry, Sri Krishnadevaraya University, Anantapur-515003, Andhra Pradesh, India

Abstract

The biogeochemistry of sulfur is the most important mechanism in the earth science community due to its impact on many different biogeochemical processes such as, like carbon (C) Nitrogen (N) and heavy metals. The role of sulfur cycle at coastal wetlands and, role in complex network of transformation processes and the long-term stability of wetland systems are still not sufficiently understood. So in this review paper we are providing many of the advances in coastal wetland sulfur biogeochemical research of the last several years. This review paper includes (1) the sulfur mechanism and the action in coastal wetlands (2) Physico-chemical processes, such as., mineral precipitation and dissolution, biologically catalyzed redox reactions such as assimilatory and dissimilatory sulfate reduction, as well as oxidation/ reduction reactions (3) the interactions between sulfur transformations with other sediment minerals (4) Sulfur gas emissions to atmosphere from wetlands (5) microbial sulfur cycle (6) indicators for wetland biological assessment. Hence this review attempt has been to explain various aspects of the sulfur ecological mechanism in coastal wetlands.

Keywords: Coastal wetlands, Sulfur ecological mechanism, Assimilatory and dissimilatory sulfate reduction

Introduction

Sulfur (S) is the tenth most common element by mass in the universe and the fifth most common on Earth. It has comprised many vitamins, proteins hormones and it has played critical roles in mankind's life, both climate and in the earth of various ecosystems. It is an essential component for making amino acids (cysteine, methionine), thioesters and proteins. S is noted in the Bible as brimstone, which means burning stone (Greenwood and Earnshaw. 1997). After nitrogen (N), phosphorus (P) and carbon (C) elements S is the fourth important nutrient and it has been played an important role in the growth of plants; which is participate in the composition of protein aminophenol, photosynthesis and respiration, etc., (Verhoeven. 2009). And also S has been important for the functioning of proteins and

enzymes in plants and animals. Many plants are absorbing organic sulfur when it is dissolved in water (Hu. et al., 2002). Animals consume these plants and take up enough sulfur to maintain their health. If the plant system lacks sulfur, it will decompensate, stunt and finally die. Coastal wetlands are varying extensively due to differences in soil texture, climate, landscape, hydrology, water quality, and flora and fauna from one region to the other region. The wetland is an important ecosystem; it has multiple various functional mechanisms that are main interest to study the contemporary ecology and S plays an important role in maintaining a healthy ecosystem (Yang. et al., 2002). The biogeochemical sulfur cycle in the wetland ecosystem has attracted wide attention (Wu. Roberta. al., 2013; et al., 2001).

²Department of Biotechnology, Sri Krishnadevaraya University, Anantapur-515003, Andhra Pradesh, India

 $^{^3}$ School of the Environment safety and Engineering, Jiangsu University, Zhenjiang, China

Biogeochemical cycles are regulated by the oxidation and reduction reactions that occur within sediments (Pezeshki and DeLaune. 2012). The coastal biogeochemical cycles (N, P, C, and S) are dealing with the transformation of chemical speciation of elements and flow of materials between biotic and abiotic compartments of coastal and marine environments. Table 1 showing sulfur compounds at global emissions of biosphere, atmosphere, marine and anthropogenic sources. However, some researchers have examined the sulfur cycle in soil-plant systems of the wetlands in China. Zhang & his group reported the S accumulation and cycling in a mangrove ecosystem (Robert and

Andjean. 1988). The S exists from the wetland soils in a variety of oxidation states and present in gaseous, soluble, and solid forms. The sulfur biogeochemical interactions in wetland sediments are complex and have distinct consequences for the biota of the wetland and for the quality of the water flowing through it (Li. et al., 2015, 2016 and 2017). In this review, 1) The sulfur mechanism and action in coastal wetlands, 2) Various levels at sulfur transformations in wetlands, 3) Sulfur Physico-chemical processes, 4) sulfur-reducing bacteria (SRB) investigation combined with the genetic level at coastal wetlands 5) SRB interaction within sulfur cycle at coastal wetlands.

Table 1: Global emissions of sulfur compounds natural, marine and anthropogenic sources

Sl No	Sources	Intensity (kg/year)	Reference
1	Hydrosphere		
	- Sea	1.3 x 10 ¹⁸ kg	Savoie. 1984; Hu. et al., 2018
	– Freshwater	$3.0 \times 10^{12} \mathrm{kg}$	
2	Atmosphere	4.8 x 10 ⁹ kg	Wu. et al., 2013
3	Pedosphere		
	-Soil	2.6 x 10 ¹⁴ kg	Chou. 2012; Zopfi. et al., 2008
	-Soil organic matter	$0.1 \times 10^{14} \mathrm{kg}$	-
4	Biosphere	8.0 x 10 ¹² kg	Zopfi. et al., 2004

The Sulfur Mechanism and Action in Coastal Wetlands

The coastal ocean is a crucial link among land, the ocean and the atmosphere. Coastal wetlands played an important role and responsibility for a series of important processes in the wetland ecosystem, such as carbon mineralization, water acidification, pyrite formation, tantalum cycling, etc., (Thamdrup. et al., 1994; Mandernack. et al., 2000; Wen. et al., 2019). Some researchers (Thomas. et al. 2009, Stebbins. et al., 2018) are identified anaerobic carbon mineralization in the deep ocean as a significant biological alkalinity source. Sulfate (SO₄²-) and nitrate compounds serve as (NO_{3}) electron acceptors for anaerobic respiration in the deep ocean. The majority of S is present as reduced inorganic sulfur minerals (pyrite, monosulphides and S0), organic forms, or SO₄²-. The majority of the Earth's S is stored underground in rocks and minerals including

as SO₄²- salts buried deep within ocean sediments. The coastal wetland ecosystems are exposed to very high SO₄²⁻ concentrations of up to the 2,700 ppm found in seawater (Schlesinger. 1991), and consequently sulfide (S2-) helps plant structure community in various systems (Chambers. et al., 1998, Koch. et al., 2007). In contrast, S supply to freshwater ecosystems is more heterogeneous. Usually the S cycle starts with in both atmospheric and terrestrial portion of the environment. The terrestrial portion, the cycle begins with the weathering of rocks and releasing the stored S to the water (H₂O) through the soil later this S comes into contact with air where it is converted into SO₄²-. This SO₄²- is taken microorganisms plants and (Desulfovibrio), and converted into organic forms. Animals consume these organic forms through foods, thereby moving the S through the food chain. Because of this organic S, organisms die decompose and some of the S is again released as SO₄²- and enters the cells of microorganisms. The S cycle of marine sediments is primarily driven by dissimilatory SO₄²-sulfate reduction (DSR) to S2- by anaerobic microorganisms (Jorgensen and Kasten. 2006; Jorgensen. et al., 2019) (Fig 1). This process links with the food web complex of organic matter degradation to the terminal organic carbon oxidation to CO₂. Most of the \overline{S}^{2-} is ultimately reoxidized back to SO₄²- via diverse S intermediates geochemical or microbial reactions that involve O2, nitrate, manganese [Mn(IV)], iron [Fe(III)], and other potential oxidants (Rickard., 2012). A fraction of the S2precipitates with iron and other metals react with organic matter and are buried deeply microbial the seabed. The transformations affect the isotopic composition of SO₄²⁻ and S²⁻ and the resulting isotope fractionation is thereby diagnostic for both process rates and pathways of the S cycle (Canfield., 2001). Different natural sources S compounds directly into atmosphere, including volcanic eruptions, the breakdown of organic matter in swamps and tidal flats, and the evaporation of water (Andreae. et al., 1992). (Li. et al. 2016) reported the SO₄²⁻ applications had a significant influence on the geochemical cycling of Fe and P in the coastal sediments. In the wetland bed, the spatial and temporal micro-scale gradients of oxygen (O2) concentrations and redox states established close to root surfaces enable the development of microbial biofilms of functionally different microorganisms. Those microorganisms can simultaneously mediate processes such as nitrification, denitrification, and mineralization of organic carbon, methanogenesis, SO₄²⁻ reduction, and S2- oxidation on a small spatial scale (Lee. et al., 1999; Holmer and Storkholm. 2001).

Plants

Coastal wetlands are identified some of the following plant species are present in the marsh land area, i.e., *Spartina alterniflora*, *Juncus roemerianus*, *Salicornia* spp, *Distichlis spicat*, *Limonium* spp., *Scirpus* spp., *Cladium jamaicens*, *Typha* spp., *Spartina patens and Spartina cynosuroides*. Oceans are the major

natural source of S in the troposphere and these are having nine trees, six shrubs, fourteen grasses, and one vine species, it has been listed as invasive species in the coastal area (Rejmanek and Richardson. 2013). SO₄2aubiquitous trace elements in the oceanic atmosphere and derived from sea spray. An extensive survey on SO₄²⁻ concentrations from non-marine origins over 25 regions of the world ocean concluded the average S concentration is about 0.7 µg/m³ (Savoie., 1984). Wetlands are important because it protects and improve water quality, provide fish and wildlife habitats, store floodwaters and maintain surface water flow during dry periods. Coastal wetland plants require S as a constituent of some amino acids which are essential to protein synthesis and it is also necessary for the formation of chlorophyll (Chl), vitamins, enzymes, and aromatic oils (Korb. et al., 2002) and also S is important in the bread making quality of wheat and the protein and sugar contents of forages and grains, and it increases digestibility of grasses and legumes (Wang. et al., 2002). Some researchers reported estuarine plants (salt marsh plants and mangrove swamps) detoxify the environmental S2- via sulfide oxidation (Lee. et al., 2009). Coastal wetland is used broadly to identify areas where wetland plants inhabit the coastal zone, in either freshwater or saltwater environments of the coastal zone. In the coastal zones are having vegetated environments such as salt marshes, fresh marshes. bottomland hardwood swamps, and mangrove swamps. The starting point in the marine atmosphere is S cycle is the air sea exchange of demethylsulphide (DMS) which is a function of the gas transfer velocity surface and seawater **DMS** concentration (Liss and Merlivat. 1986; Wanninkhof. et al., 2009).

Sediment and Mineral Precipitation

Depending on the natural conditions, S compounds in the environment may play the role of electron acceptor or donor in the redox processes. S containing proteins are degraded into their constituent amino acids by the action of a variety of soil organisms (bacteria and algae). The soil has formed though, big

rocks break down into smaller rocks by continuous action of wind and rain. This process takes many years for these rocks to break down into smaller particles. The S of the amino acids is converted to hydrogen sulfide (H₂S) by another series of soil microbes. In the

presence of O_2 , H_2S is converted to S and then to SO_4^{2-} by SRB. Eventually the SO_4^{2-} becomes H_2S . These H_2S compounds rapidly oxidize to gases that dissolve in water to form sulphurous and sulphuric acids (H_2SO_4) (Sheoran and Sheoran. 2006).

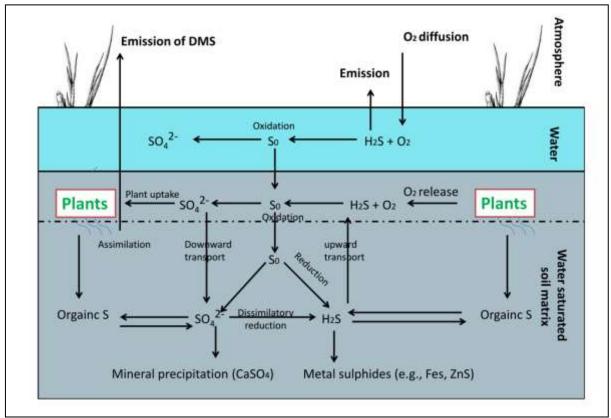


Fig 1: Sulfur cycling in coastal wetlands

These compounds contribute in large part to the acid rain this can kill sensitive aquatic organisms and damage marble monuments and stone buildings. H₂S is a common metabolic poison that is abundant in marinereducing environments (Sheoran and Sheoran. 2006). Some researchers are reported, most marine sediment concentrations (SO_3^{2-} , and $S_2O_3^{2-}$, and $S_4O_6^{2-}$) are in values not exceeding micromoles (µM) per liter (L) and also also reported elemental (S^0) is the most sulfur abundant intermediate in coastal marine sediments in Black Sea, Germany (Zopfi. et al., 2004). Mineral components can dissolve in the weathering and erosion process, it can precipitate when environmental conditions minerals good. As break compounds are separated into their ion and cat-ion components as it dissolves in water.

Changes in physical and chemical conditions (such as changes in temperature, pressure, or addition or removal of dissolved compounds like carbon dioxide (CO2) and biological activity) cause minerals to precipitate. In desert environments, H2O evaporates causing salts to precipitate on the surface of dry lake beds. The cementation is what hardens sediments into sedimentary rocks, there are common mineral cements include silica (quartz), calcite, limonite, hematite, and clay minerals (Zheng and Hoefs. 1993). Among soil microbial community, soil reducing bacteria are considered to be one of the richest and the most important groups of microbes, which play pivotal roles in participating soil organic matter (SOM) decomposition, and regulating soil C and N cycling (Jurburg. et al., 2018).

Atmosphere

The S found in the atmosphere and enters into the atmosphere through both natural and human sources. Natural recourses occur; instance of volcanic eruptions, bacterial processes, evaporation from water, decaying organisms. When S enters the atmosphere through human activity, this is mainly a consequence of industrial processes where sulfur dioxide (SO₂) and H₂S gases are emitted on a wide scale. When SO₂ enters the atmosphere it will react with O2 to produce sulfur trioxide gas (SO₃), or with other chemicals in the atmosphere, to produce S salts. SO₂ may also react with water to produce H₂SO₄. This H₂SO₄ may also be produced from DMS, which is emitted to the atmosphere by plankton species (Andreae. et al., 1992; Larry. et al., 2014). The S gases are both foul smelling and noxious. Researchers examined various studies, wetlands emit the range of S gases and they vary according to factors such as salinity, wetting drying regime, soil type and diurnal cycles. Three main types of S gases can be emitted by wetlands; which are., H2S, volatile organic sulfur compounds (VOSC) and SO₂. These S compounds are differing in the way these are produced and their odour characteristics smell threshold and toxicity. The human nose can detect some of these compounds at very low concentrations and also other various types of S compounds, H₂S, Carbonyl sulfide (COS), Carbon disulfide (CS2), Methanethiol (MT) (CH₃SH), DMS ((CH₃)₂S), Dimethyl disulfide (DMDS) ((CH₃)₂S₂), SO₂. All these particles are settled back onto earth, or react with rain and fall back onto earth as acid deposition. The particles are absorbed by plants again and are released back into the atmosphere, so that the sulfur cycle will start over again (Amend. et al., 2004).

Sulfur Physico-Chemical Processes Sulfur Oxidation and Desulfonylation

Sulfur oxidation involves the oxidation of reduced S compounds such as H_2S , inorganic sulfur (S_0) and thiosulfate ($S_2O_2^{-3}$) to form H_2SO_4 , an example of a sulfur oxidizing bacterium is *Paracoccus*. Generally, the oxidation of sulfide occurs in various

stages, with inorganic S being stored either inside or outside of the cell until needed. The two step process occurs because S²- is a better electron donor than inorganic sulfur or S₂O₂-3; this allows a greater number of protons to be translocated across the membrane. Sulfur oxidizing microorganisms generate reducing power for CO₂ fixation via the calvin cycle using reverse electron flow an energy requiring process that pushes the electrons against their thermodynamic gradient to produce Nicotinamide adenine dinucleotide (NADH). Biochemically reduced compounds are converted to sulfite (SO₂-3) and, subsequently, SO₂-4 by the enzyme sulfite oxidase. Some organisms, however, accomplish the same oxidation using a reversal of the APS reductase system used by SRB. In photosynthetic reactions, the energy liberated is transferred to the electron transport chain for ATP and NADH production. In addition to aerobic S oxidation, some organisms (Thiobacillus denitrificans) use nitrate (NO⁻³) as a terminal electron acceptor and grow anaerobically. S2- is a product of bacterial DSR by using organic compounds as electron donors. Elemental sulfur is a product of S²- oxidation, which may be performed by abiotic oxidation and/or biological oxidation by using different electron acceptors, such as oxygen, nitrite and nitrate (Zheng., 2007; Zheng and Cai., 2007) (Fig 2). There are several chemical reactions leading to the removal of a sulfonyl group from organic compounds i.e. desulfonvlation reactions. As the sulfonyl functional group is electronwithdrawing, methods for cleaving the Scarbon bonds of sulfones are typically reductive nature. Olefination in replacement with hydrogen be accomplished using reductive desulfonylation methods (Alonso and Ájera., 2009). The wetland system can influence the sulfur cycling by the releasing of organic carbon compounds and/or oxygen from the plant roots to enhance the sulfate reduction or reoxidation of the reduced sulfur compounds. Moreover, the processes of sulfur transformations, such as SO₄²- reduction can influence the conditions for biochemical processes (Leon. et al., 2002; Geurts. et al., 2009). Oceanic DMS emissions to the atmosphere are potentially important to the Earth's radioactive balance. Since these emissions are driven by the surface seawater concentration of DMS, it is important to understand the processes controlling the cycling of sulfur in surface seawater (Bates. *et al.*, 1994).



Fig 2: Oxidation and Reduction reactions

Assimilatory and Dissimilatory Sulfate Reduction

Assimilatory sulfate reduction (ASR) is a pathway used by prokaryotes, fungi and photosynthetic organisms to inorganic SO₄²⁻ to S²⁻, which is further incorporated into carbon skeletons of amino acids to form cysteine (Cys) or homo-Cys. In this pathway, SO₄²⁻ is first activated by (adenosine triphosphate) ATP sulfurylase (ATPS) forming adenosine 5-phosphosulfate (APS). In higher plants, APS is reduced by APS reductase (APR) to sulfite, which is further reduced to the level of S2- by sulfite reductase (Suter. et al., 2000). APS can also be phosphorylated by APS kinase phosphoadenosine 5-phosphosulfate, which is utilized for synthesis of a wide range of sulphated compounds in reactions catalyzed by a variety of sulfo transferases. DSR is a form of anaerobic respiration that uses SO₄²as the terminal electron acceptor. This metabolism is found in some types of bacteria and archaea which are often termed sulfur reducing microorganism (SRM). DSR occurs in three steps (a) Conversion (activation) of sulfate to Adenosine 5-phosphosulfate (APS), (b) reduction of APS to sulfite (c) reduction of sulfite to S2-. This process requires the consumption of a single ATP molecule and the input of 8 electrons (e-) (Larry. et al., 2014; Grein. et al., 2013). The protein complexes are responsible for these chemical conversions (-Sat, Apr and Dsr-) are found in all currently known organisms that perform dissimilatory sulfate reduction (Pereira. 2011). Energetically SO₄²⁻ is a poor electron acceptor for microorganisms as the sulfatesulfite redox couple is $E^{0'}$ -516 mV, which is negative to allow reduction by NADH or ferrodoxin that are the primary intracellular electron mediators (Muyzer and

Stams., 2008). To overcome this issue, SO₄²⁻ is first converted into APS catalysed by the sulfurylase (Sat), utilized a enzyme ATP single ATP molecule. The APS sulfite redox couple has a E^{0} of -60 mV, which allows APS to be reduced by either NADH or reduced ferrodoxin using the enzyme adenylyl-sulfate reductase (Apr), which requires the input of 2 electrons (Muyzer and Stams., 2008). In the sulfite reduced final step, is the dissimilatory sulfite reductase (Dsr) to form S²-, requiring the input of 6 electrons (Grein. et al., 2013). Thus, APR is a key step in sulfate assimilation and as such, the enzyme is highly regulated, e.g. by light, S and nitrogen supply, heavy metals, or chilling (Koprivova. et al., 2000).

Sulfur Reducing Microorganisms

SRB are anaerobic microorganisms that use SO₄²- as a terminal electron acceptor, for degradation example, the of organic compounds. These organisms are ubiquitous in anoxic habitats, where they have an important role in both the S and carbon (C) cycles. SRB can cause a serious problem for industries, such as the offshore oil industry, because of the production of S2-, which is highly reactive, corrosive and toxic. A classic example of a sulfur-oxidizing bacterium (SOB) is Beggiatoa grows chemoorgano heterotrophi-cally oxidizing by compounds to CO_2 in the presence of O_2 , though high concentrations of oxygen can be a limiting factor (Pester. et al., 2012). Organic compounds are also having carbon source for biosynthesis of this organism. Some species may oxidize H₂S to S⁰ as a supplemental energy, is this S intracellularly. Some species have the ability chemolithoautotrophic growth, sulfide oxidation for energy and CO2 as a source of carbon for biosynthesis (Ng. et al., 2010). In this metabolic process, internal stored nitrate is the electron acceptor and ammonia reduced to (NH_3) . Marine autotrophic Beggiatoa species are able to oxidize intracellular S to sulfate. reduction of So frequently occurs when O2 is lacking. S is reduced to sulfide at the cost of stored carbon or by added hydrogen gas. This may be a survival strategy to bridge periods without O2. This reaction process follows as below.

Sulfide oxidation: $2H_2S + O_2 \rightarrow 2S + 2H_2O$

However, these organisms can also be beneficial by removing SO₄²⁻ and heavy metals from waste water streams. In addition, SRB have been studied for new molecular biological and genomic techniques (Hu. *et al.*, 2018). Various SRB's (like *Desulfovibrio*, *Desulfotomaculum* and *Desulfomonas*) are used in reduction and oxidation reactions.

Desulfovibrio

Desulfovibrio is a genus of gram negative sulfur reducing bacteria and this species has commonly consists in aquatic environments with high levels of organic material, as well as water logged soils. Like SRB, Desulfovibrio was long considered to be obligately anaerobic. Desulfovibrio strains have been found in a variety of habitats, including soil, the intestines and feces of animals, and both salinic and fresh water (Tarasov. et al., 2015). Desulfovibrio vulgaris Hildenborough is a model organism for studying the energy metabolism of SRB and for understanding the economic impacts of biocorrosion SRB. including of infrastructure and bioremediation of toxic metal ions (Heidelberg. et al., 2004). These types of bacteria are known as aerotolerant. Some Desulfovibrio species have in recent vears been shown to have bioremediation potential for toxic radionuclides such and reductive uranium iron by a bioaccumulation process. Because of Desulfovibrio's historical importance, two strains have already been genomically sequenced and one is currently in progress.

These strains include Desulfovibrio desulfuricans G20 (completed), Desulfovibrio vulgaris subsp. vulgaris Hildenboroug (completed), and Desulfovibrio magneticus (in progess), which were sequenced by DOE **Joint** Genome Institute, TIGR, and NITE, respectively. Both of the completely sequenced genomes showed Desulfovibrio to have one chromosome and measure over 3 Mbp in length. Both sequencings also found the number of proteins to be above 3000 (Heidelberg. et al., 2004). D. vulgaris Hildenborough is a model organism for studying energy metabolism of SRB and for understanding the economic impacts of SRB, including biocorrosion of metal infrastructure and bioremediation of toxic metal ions.

Indicators for Wetland Assessment

Wetland assessment has been a popular field and however, various indicators have been used in wetland assessment for different purposes and backgrounds (Reddy and Ronald., 2009). Wetlands are receiving millions of sewage and pollutants from agricultural industrial and effluents producing by harmful algal blooms (HABs) and which is caused human diseases & destroy aquatic systems (Van Dohla, 2000). The development of wetland monitoring and assessment strategies is necessary to explain the impact on human activities on wetland health and improve wetland management and protection. Various wetland assessment indicators have been developed employed to measure the changes ecological condition. There are primarily three types of indicators based on physical (water depth, open water area, and land uses), chemical (total P and N, sediment chemistry) and biological (plant species, composition and abundance of macroinvertebrates, algae, etc.) characteristics (Asmus. et al., 2009; Yagow. et al., 2006). Bioindication uses higher plants, animals, and microbial species to predict or indicate wetland water quality and health conditions (Liu and Sun., 2010; Sims. et al., 2013).

Concluding Remarks and Perspective

Several groups of microbes are responsible for carrying out processes involved in the S cycle. Anoxygenic photosynthetic bacteria as well as chemoautotrophic archaea and bacteria use H₂S as an **electron donor**, oxidizing it first to S^0 , then to SO_4^{2-} . This leads to stratification of H₂S in soil, with levels increasing at deeper, more anaerobic depths. Many bacteria and plants can use SO_4^{2-} as a S source. Coastal biogeochemical cycle's exhibit interlinks of different mechanisms that regulate the transfer of chemical elements in the marine ecosystem and encompass rapid complicated processes that bridge material transport between the various compartments of the Earth system, land, ocean, and atmosphere as well as human society. The external forcing factors on coastal biogeochemical cycles can be either natural or anthropogenic, as well as the combination of these two. Biogeochemistry in the coastal ocean is composed of cycles of macronutrients and trace elements between biotic and abiotic compartments driven by formation of organic matter fuelled by solar energy and the metabolisms (i.e., anabolic and catabolic processes) based on the chemical energy stored in the organisms. Since the late 1970s, it has been recognized that the microbial loop an important component biogeochemical cycles in the ocean, which is an integral part of the entire food web and considerably the pathways biogeochemical cycles in the coastal ocean. It is known that coastal biogeochemical cycles, such as sulfur cycle have strong feedbacks to the atmosphere (e.g., emission of greenhouse gases) as well as the open ocean through change in material fluxes. This review expended the biogeochemical role of S in marine sediments, such as sulfate reduction, pyrite and organic S formation and metal cycling, is an area of intense research. Sulfate reduction and burial of S in the form of pyrite and organic S have previously been observed in mangrove sediments. Little is known, however, about the actual mechanisms and control of specific processes involved in mangrove S cycling. Although S compounds generally are important for energy transfer

and element cycling in tidal sediments, pyrite formation and oxidation.

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