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Extreme sulfur-cycling in acid brine lake environments of Western Australia

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ABSTRACT

Regional-scale extreme acid brines in southern Western Australia are an example of sulfur-rich end-member continental waters and are amongst the most physically and chemically dynamic settings in the world. Here, aqueous sulfur compounds, sulfur minerals, and sulfur-interacting microorganisms are abundant and diverse and sulfur cycling is intense. Lake waters and groundwaters of the Yilgarn Craton are Na–Cl–SO_4–Mg-rich with pH as low as 1.7, salinity as high as 32% total dissolved solids, and sulfuric acid as the dominant acid. Measured concentrations of dissolved total S range from 500 to 19,800 ppm, and SO_4 ranges from 692 to 35,169 ppm. The difference between total S and SO_4 suggests that other S species are present in these waters, particularly in the most acid brines. Rainwaters, which have previously been suggested as a source of sea spray aerosol SO_4 to the lake systems, yielded 7 to 8 ppm SO_4. Sulfur isotope analysis shows that $\delta^{34}S$ for lake waters is 17.0 to 19.1‰, for groundwaters is 17.0 to 18.6‰, for gypsum is 20.2 to 20.4‰, and for alunite is 18.9‰. These values are consistent with a combination of S sources and sinks. Sulfur cycling processes include sulfide oxidation, evapoconcentration, mineral precipitation, mineral dissolution, and bacterial reduction and oxidation. Field, petrographic, and molecular analyses of microorganisms in these acid saline environments suggest that some are S-oxidizing and S-reducing bacteria. Sulfur-bearing minerals in the area include both sulfides and sulfates. Sulfides, including pyrite, have been observed in veins and disseminated in surface exposures and shallow cores of Archean igneous and metamorphic host rocks, as well as in overlying sediments. Sedimentary sulfate minerals include gypsum, bassanite, anhydrite, and rozenite that grow directly from lake waters, and gypsum, anhydrite, jarosite, natrojarosite, alunite, basaluminite, hydrobasaluminite, langbeinite, and rozenite that grow diagenetically from shallow groundwaters. These sedimentary minerals are subjected to physical reworking and chemical dissolution and re-precipitation. Sulfur cycling among the lithosphere, hydrosphere, biosphere, and atmosphere in acid saline environments is dynamic. Driving forces are rock weathering, weather, and climate, which ultimately influence the water chemistry. Flooding–evapoconcentration–desiccation cycles accelerate short-term local and regional sulfur cycling. The combination of low pH, highly saline waters, and dynamic changes in environment make sulfur arguably the most important element in these environments.

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1. Introduction

Sulfur is an abundant element on Earth. Although some form of sulfur cycling occurs in most sedimentary environments, ephemeral acid saline systems seem to be an end-member, with great levels of sulfur concentration and cycling that strongly influences the geochemistry of the system. In these extreme environments, sulfur compounds and sulfur minerals exist in great abundance and diversity. Rock weathering, water geochemistry, and weather/climate-driven flooding–evapoconcentration–desiccation stages promote short-term local and regional dynamic sulfur cycling. The combination of low pH, highly saline groundwaters and lake waters and dynamic changes in environment make sulfur one of the most important elements in these environments. Acid brine systems in Western Australia are an example of such a sulfur-rich end-member environment. The goal of this paper is to describe the various processes and products of sulfur cycling in ephemeral acid saline lakes and adjacent environments in southern Western Australia, and to suggest how recent and future global warming enhances sulfur cycling in such an environment.

1.1. Significance of the sulfur cycle

The sulfur cycle is a description of processes that transfer sulfur through the different Earth systems. Sulfur is an important element on Earth because it is a common constituent in the lithosphere, hydrosphere, biosphere, and atmosphere. Due to its multiple valence states, sulfur can make a great number of compounds and be involved

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in a variety of geochemical and biochemical processes (Mandeville, 2010).

Knowledge of sulfur and its reactions is vital for several reasons. Sulfur and its compounds have industrial value, used for "pickling" steel, in gunpowder, in fireworks, in batteries, as a bleaching agent, as a mold- and bacterial-killer, as a fertilizer, as a dehydrator, and as a food preservative. Sulfur is also important in the human body because it aids the transport of oxygen across cell membranes. Atmospheric sulfur emitted in large quantities during huge volcanic eruptions has impacted global climate. The environmental impact of sulfur has been recognized, as well. In addition to volcanic eruptions, burning fossil fuels also releases sulfur into the atmosphere and can result in acid rain. Recently, geoengineering the atmosphere to reduce irradiance of the Earth's surface has been proposed and would involve seeding the stratosphere with sulfate. With the discovery of sulfur on Mars, King and McLennan (2010) argue that the sulfur cycle is the most important geochemical cycle on that planet.

The sulfur cycle has been studied in terms of global cycling, in which the ocean and the seafloor are the most important constituents (i.e., Andreae, 1990; Schlesinger, 1997; Canfield and Raiswell, 1999; Sievert et al., 2007). Contributions from continental sources and sinks for sulfur in global models are typical freshwater rivers, lakes, and soils. Some local sulfur cycling studies have been conducted for continental extreme environments and tend to focus on the details of limited aspects of the system (i.e., Xu et al., 1998). In contrast, this manuscript considers the acid saline lakes of southern Western Australia as a microcosm for: 1) describing the continental geochemical processes that influence sulfur cycling on relatively short (hourly to decadal) time scales; 2) identifying the range of S-bearing sedimentary deposits that record sulfur cycling processes; and 3) exploring how recent and future global change may influence sulfur cycling within these systems.

2. Geologic setting, climatic conditions, and sedimentary processes

The Yilgarn Craton of southern Western Australia is a large (~1.78 million km^2), tectonically stable mass of Archean rocks (Fig. 1). The majority of rocks here are granites, granodiorites, and gneisses, although some mafic and ultramafic rocks, such as metagabbros, mafic schists, and komatiites, are also represented as greenstone belts that trend NNW–SSE through the craton (Myers, 1995). Other Archean rocks represented are anorthosites, ironstones, and quartzites. The Yilgarn Craton comprises more than 10% of the Australian landmass and is a rich source of mineral resources, most notably gold, nickel, and uranium.

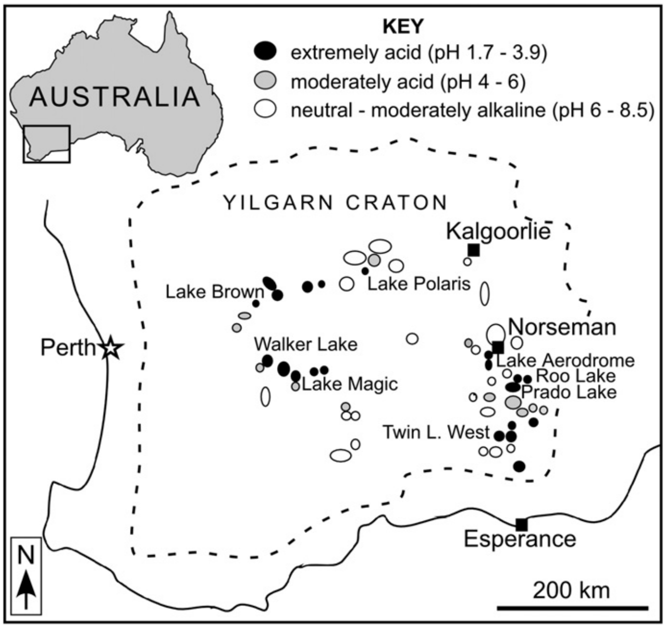

Fig. 1. Map of southern Western Australia showing the outline of the Yilgarn Craton and lakes studied. Lake names labeled represent those named specifically in the text.

There is no Paleozoic or Mesozoic record of deposition in the Yilgarn Craton. However, the Precambrian rocks have undergone several episodes of intense weathering (Anand and Paine, 2002). Fluvial systems active in the Jurassic through Eocene likely eroded a drainage system, resulting in paleochannels now filled with Cenozoic sediments (Clarke, 1994a,b; Clarke et al., 1996; Lawrance, 2001; de Broekert and Sandiford, 2005).

The topography of the Yilgarn Craton is relatively flat with elevations ranging from ~250 to ~365 m above sea level. The landscape includes eucalypt forests, vegetated sand dunes, and ephemeral saline lakes amid the many low relief and some domed outcrops of weathered Archean rocks.

Modern climate of southern Western Australia is classified as semiarid steppe (Australian Bureau of Meteorology). Air temperatures range from −5 °C to 50 °C. Average rainfall is ~26 cm/year in the northeastern Yilgarn Craton and ~34 cm/year in the southwest. Average annual evaporation is ~180–280 cm/year. Winds can be strong and multidirectional, with an average year round velocity of ~30 km/h (Australian Bureau of Meteorology). Dust storms and wildfires are relatively common, especially during the austral summers (McTainsh and Pitblado, 1987).

Natural surface waters of southern Western Australia are restricted to saline lakes, with the exception of short-lived sheetfloods caused by infrequent local rains. Hundreds of shallow and ephemeral saline lakes exist through the Yilgarn Craton. Fig. 1 shows approximate locations of 54 lakes that have been studied (Benison et al., 2007) and Fig. 2 shows typical appearances of lakes from the air and from the ground. Some of these lakes are situated directly on Archean rocks. Other lakes are hosted by recent sediments over paleochannels (Benison et al., 2007; Story et al., 2010b). All lakes of the Yilgarn Craton are shallow (less than ~1 m) and saline. Many of the lakes are also acid. Lake water pH ranges from 1.5 to 8.5 throughout the Yilgarn, with a temporal variation of ~2 pH units at individual lakes. Groundwater, focused under the lakes, has a more consistent pH of ~3.5 and is also saline (Bowen and Benison, 2009). Neutral lakes are separated from the underlying acid groundwater by clay beds, making them essentially perched aquifer lakes (Benison et al., 2007).

Lakes undergo flooding, evapoconcentration, and desiccation at irregular time periods of hours to decades (Fig. 2D–F). Flooding stages are prompted by rainstorms, which cause sheet floods to carry meteoric water, sediment, and decayed plant matter to the lakes. Flooding also dissolves some evaporite minerals, such as halite, bassanite, and, to a lesser extent, gypsum. Flooding events are typically short, on the order of an hour to 1–2 days. However, lake waters may remain in a flooded state, characterized by increased depth (~0.5–2 m deep) and higher pH, for weeks to months. Salinity of water during a flooded state depends on the amount of halite which was dissolved during the flooding event; abundant halite prior to flooding leads to high salinity in flood waters as halite dissolves, yet little halite prior to flooding results in dilute lake waters during flooding stage (Benison et al., 2007).

Evaporation of lake waters causes lake diameters and water depths to decrease (to cm-scale depths), water pH to decrease, and water salinity to increase. In addition, minerals precipitate from lake waters during the evapoconcentration stage. Halite and gypsum are the most common minerals to precipitate directly from the lakes in Western Australia, but bassanite, rozenite, iron oxides, and kaolinite also precipitate directly from lake waters. The most extreme geochemistry, characterized by extremely low pH and extremely high salinity lake waters, exists during evapoconcentration.

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