



Methanotrophy controls groundwater methane export from a barrier island

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Abstract

Methane concentrations can be high in coastal groundwater, resulting in methane export driven by submarine groundwater discharge. However, the magnitude of this methane flux depends significantly on the rate of methanotrophy, the often overlooked process of microbial methane consumption that occurs within coastal aquifer sediments. Here we describe a zone of methanogenesis within the freshwater lens of a barrier island aquifer and investigate the methane source/sink behavior of the barrier island system as a whole. The median concentration of methane dissolved in fresh groundwater beneath the center of the island was 0.6 mM, supported by high rates of potential methanogenesis ($22 \text{ mmol m}^{-2} \text{ day}^{-1}$). However, rates of microbial methane consumption were also elevated in surrounding sediments ($18 \text{ mmol m}^{-2} \text{ day}^{-1}$). Groundwater flowing from the zone of methanogenesis to the point of discharge into the ocean had a long residence time within methanotrophic sediments (~ 195 days) such that the majority of the methane produced within the barrier island aquifer was likely consumed there.

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1. INTRODUCTION

Methane (CH_4) accounts for 17% of the total radiative forcing of Earth's atmosphere, making it the third most important greenhouse gas behind water vapor and carbon dioxide (Myhre et al., 2013). Since the industrial revolution, the atmospheric concentration of CH_4 has risen by 150% to 1.8 ppm. This increase was driven by human activities such as agriculture, energy production, and waste management

(Ciais et al., 2013). Methane is consumed in the atmosphere by chemical reaction with hydroxyl radicals (Myhre et al., 2013) and in marine and terrestrial waters, sediments, and soils by microbial methanotrophy. Thus, CH_4 plays a critical role in the global heat budget and is active in atmospheric chemistry. Understanding the atmospheric, terrestrial, aquatic, and marine sources and sinks of CH_4 is therefore highly relevant in the context of climate change.

Globally, the dominant CH_4 production mechanism is methanogenesis mediated by members of the archaea. In the terrestrial realm, these microorganisms are active primarily in anoxic, aquatic habitats (Cicerone and Oremland, 1988). Therefore, the most important sites of CH_4 production are organic rich soils saturated with fresh water (e.g., marshes, rice paddies, landfills) (Ciais et al., 2013). Coastal environments are often overlooked as sites of methanogenesis because of the high sulfate content of

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seawater. Under these conditions, sulfate reducing bacteria are expected to outcompete methanogenic archaea for organic carbon substrates since sulfate reduction is a more energetically favorable process. However, the coastal zone contributes substantially to the total marine CH₄ flux to the atmosphere (Bange, 2006), and the groundwater flowing through soils and sediments is a dominant source of CH₄ to the coastal ocean (Bugna et al., 1996; Grunwald et al., 2009).

These interstitial fluids are enriched with dissolved CH₄ derived from methanogenesis in aquifers containing freshwater and sufficient organic carbon substrate (Bugna et al., 1996). Aquifers matching this description are not limited to inland environments. Coastal uplands develop a lens of fresh groundwater floating above a layer of denser, saline groundwater (Collins and Easley, 1999), thereby creating a zone with the potential for CH₄ production. Barrier islands are globally important coastal landforms, occupying 15,100 km (6.5%) of open ocean shorelines (Stutz and Pilkey, 2001) and an additional 7410 km of lower energy coastal environments (Pilkey et al., 2009). Therefore, barrier island methanogenesis could make important contributions to coastal CH₄ budgets.

Methane in coastal surface waters is often associated with submarine groundwater discharge (SGD) and has been used as a tracer for SGD in numerous studies (Cable et al., 1996; Kim and Hwang, 2002). However, before reaching the ocean, groundwater traverses multiple zones of intense biogeochemical cycling within coastal aquifers. These zones, where freshwater mixes with seawater and where rapid porewater advection is driven by wave and tidal action, are referred to as the subterranean estuary (Moore, 1999). Brackish groundwater contains high concentrations of sulfate, and infiltrating seawater carries oxygen into coastal aquifers. Therefore, the subterranean estuary has the potential to host active CH₄ oxidation by both anaerobic and aerobic methanotrophic microorganisms. The balance between CH₄ production, transport, and consumption rates within the subterranean estuary will control the flux of CH₄ to the adjacent coastal ocean.

In this work, we tested the hypotheses that: (1) barrier island aquifers support zones of microbial CH₄ production, (2) a fraction of this CH₄ is exported to the atmosphere or ocean, and (3) the dominant fate for CH₄ is microbial consumption within the barrier island system. The ratio of CH₄ export from the barrier island aquifer to total CH₄ production within the aquifer is controlled by the CH₄ consumption rate in sediments along the groundwater flow path and the residence time of CH₄-bearing groundwater within those methanotrophic sediments. We investigated these processes on a barrier island in Georgia, USA by surveying groundwater CH₄ concentrations, CH₄ production and consumption rates, and CH₄ efflux to the atmosphere across the island, and by modeling groundwater flow paths and rates.

2. METHODS

2.1. Site description

Sapelo Island is located on the coast of Georgia, USA (Fig. 1a). The study site, Cabretta Island, is a small

(~250 m wide), Holocene barrier island on the seaward side of Sapelo Island (Fig. 1b). The mean annual rainfall on Cabretta Island is 130 cm and the mean tidal range is 2.5 m (Fig. 1c). Trees and shrubs grow on the upland of the island, which is flanked by a small tidal creek and salt marsh on one side and a dune and beach system on the other. A transect of groundwater monitoring wells spanning this range in environment type was installed across the island in 2008 (Fig. 1b, c). Wells were placed in 7 clusters with 1 to 3 wells per cluster. Within each cluster, wells with 30 cm screened intervals were installed from 1 to 5 m below the sediment surface. Each well was capped and contained a tightly fitting internal casing to minimize the internal storage volume of the well and prevent contamination of the aquifer with materials from the surface.

Cabretta Island is a transgressive barrier island composed of alternating layers of mud and sand. A simplified version of the island stratigraphy described by Wilson et al. (2011) is provided in Fig. 1c. Surficial salt marsh sediments were composed of mud and silt on the leeward side of the island. This stratum was connected with older marsh sediment buried beneath the island upland and beach, outcrops of which could be seen near the low tide line on some portions of the beach (Fig. 1d). An unconfined sandy aquifer overlaid the buried marsh platform on the island upland and beach, and a confined sandy aquifer was located beneath the marsh mud. The particulate organic carbon content of the mud found in the marsh platform buried beneath the island center was $0.64\% \pm 0.17\%$ ($n = 3$) and $0.05\% \pm 0.01\%$ ($n = 6$) in the unconfined sandy aquifer. Rain falling on the island upland percolated through the sediment creating a freshwater lens within the sediment of the unconfined sandy and buried marsh platform aquifers (Fig. 1c). This freshwater mixed with infiltrating seawater, creating a salinity gradient that is not represented in the figure.

2.2. Groundwater and surface water geochemistry

An initial survey of Cabretta Island groundwater and surface water geochemistry was carried out with samples collected every other month from August 2008 to August 2010. Salinity, temperature, and dissolved oxygen (DO) concentrations were measured in the field, while samples were preserved and returned to the laboratory to measure the concentrations of sulfide (H₂S), CH₄, dissolved organic carbon (DOC), chloride, and sulfate. Prior to sample collection, the stagnant groundwater was purged from each monitoring well using a peristaltic pump. The salinity and temperature of each sample was determined using a handheld conductivity meter (YSI Model 30) with an accuracy of $\pm 2\%$, and the DO concentration was measured using a polarographic DO probe connected to a handheld meter (Thermo Fisher Orion 5-Star portable DO meter). When the scent of H₂S was present in a sample, the DO concentration was assumed to be zero and was not measured to prevent poisoning the DO probe. Freshly recharged groundwater was pulled into a disposable syringe using tygon tubing. Surface water samples were collected directly from the adjacent tidal creek and ocean surf zone using the

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