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Stable isotopic and molecular composition of desorbed coal seam gases from the Walloon Subgroup, eastern Surat Basin, Australia

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article info abstract

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This study used compositional and stable isotopic analysis to test hypotheses on the distribution and origins of Walloon Subgroup coal seam gas (CSG) in the eastern Surat Basin, Queensland, Australia. The Middle Jurassic Walloon Subgroup play differs from many other low-rank CSG plays—particularly in methane carbon isotopic signature, i.e., the CSG is not as 'microbial' as could be expected. The carbon isotope compositions of desorbed methane from three cored appraisal wells fall within the generally accepted range for thermogenic or mixed gas ($\delta^{13}C - 58.5\%$ to -45.3%). The $\delta^{13}C$ –CH₄ values from stratigraphically placed coal core samples increased (became more 'thermogenic') from the top of the upper (Juandah) coal measures to the base of the Tangalooma Sandstone. Below the Tangalooma Sandstone, in the lower (Taroom) coal measures, the $\delta^{13}C$ –CH₄ values decreased with increasing depth. These positively parabolic $\delta^{13}C$ profiles tracked total measured gas content in two out of the three wells studied. The third well displayed lower variance of $\delta^{13}C$ –CH₄ and gas content increased uniformly with depth.

A genetic classification based on methane stable carbon isotopes alone might interpret this pattern as a transition from microbially- to thermogenically-sourced methane in the central coal seams. However, a $\delta^{13}C$ –CO₂ profile for one well tracks total gas content and δ^{13} C–CH₄, and exhibits an inverse relationship with δ D–CH₄. These results, together with the mostly dry nature of the gas samples $[(C_1/(C_2 + C_3))$ ratios up to ~10,000] and relatively uniform δ D–CH₄ values (δ D −238‰ to −202‰), suggest that microbial CO₂ reduction is the primary source of Walloon Subgroup methane. As such, stratigraphic variations in gas content mainly reflect the extent of microbial methanogenesis. We suggest that peak microbial utilisation of H_2 -CO₂ occurred at the Tangalooma Sandstone level, enriching the residual CO₂ pool and derived methane in ¹³C. Carbon $\lbrack \Delta^{13}C(CO_2-CH_4) \rbrack$ and deuterium isotopic differences [ΔD(H₂O–CH₄)], and cross-plots of δ D–H₂O and δ ¹⁸O–H₂O are also consistent with kinetic isotope fractionation during microbial-mediated carbonate reduction. The results are relevant for applying microbially enhanced coal bed methane (MECoM) in the Surat Basin.

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1. Introduction

Microbial methane is an important contributor to fossil coal seam gas (CSG) resources, and extant methanogens have the potential to generate future resources if subsurface ground conditions optimum to their cultivation can be determined (microbially enhanced coal bed methane; MECoM; [Scott, 1999\)](#page--1-0). The eastern Australian CSG basins, including the Surat Basin, host considerable microbial methane resources and methanogenesis is thought to be ongoing (e.g. [Draper and Boreham,](#page--1-0) [2006; Faiz and Hendry, 2006; Golding et al., 2011, 2013a, 2013b;](#page--1-0) [Kinnon et al., 2010; Li et al., 2008; Papendick et al., 2011; Smith and](#page--1-0) [Pallasser, 1996\)](#page--1-0).

In this context, recent research suggests the Surat Basin may have significant in situ bioreactor potential ([Golding et al., 2011;](#page--1-0) [Papendick et al., 2011\)](#page--1-0). Surat Basin methane is produced from the Middle Jurassic Walloon Subgroup, which accounts for over 64% of Australia's '2P' (proven and probable) CSG reserves ([Geoscience](#page--1-0) [Australia, 2010](#page--1-0)). Ramping development and the potential for future microbial regeneration of this resource call for detailed geochemical studies across the core production region in the eastern Surat Basin, Queensland. Despite the proliferation of Walloon CSG, few studies have used isotopic fingerprinting to test specific hypotheses relating to source inputs. [Draper and Boreham \(2006\)](#page--1-0) interpreted Walloon Subgroup CSG as mainly secondary microbial in origin, generated by the reduction of $CO₂$, on the basis of analyses of the initial production gases. The carbon isotope compositions of produced methane plot in the 'mixed origins' field of [Whiticar \(1999\),](#page--1-0) despite the low rank of the coals (average vitrinite reflectance $R_0 \sim 0.5\%$), suggesting that they may contain a component of migrated thermogenic gas [\(Draper and](#page--1-0) [Boreham, 2006\)](#page--1-0). However,Walloon Subgroup CSG is typically produced from multiple coal seams over $a \geq 300$ metre-thick depth range, potentially obscuring any stratigraphic differences in isotopic composition. This is problematic, as previous studies of gas content distribution have suggested that a strong stratigraphic control exists ([Hamilton](#page--1-0) [et al., 2012; Ryan et al., 2012; Scott et al., 2007\)](#page--1-0).

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This study tests the degree of compartmentalisation in the reservoir by comparing δ^{13} C and δ D for desorbed coal core gases that were sampled in stratigraphic sequence relative to trends in gas distribution. Specifically, this study aims to: (1) determine whether there are significant geochemical differences up-sequence and across the eastern Surat Basin, which could explain the variation in gas contents and relative saturation; and (2) elucidate evidence for controls on the spatial variability of methanogenesis. Desorbed gases were sampled across the entire stratigraphy for 3 wells (Fig. 1), resulting in extensive compositional and isotopic data through the core zone of CSG production in the eastern Surat Basin. Gas geochemistry is compared with coal bed gas content, water stable isotope and coal property data, and previous work on production gases ([Draper and Boreham, 2006\)](#page--1-0), to better understand gas origins and microbial recharge processes.

2. Geological and hydrogeological setting

The intracratonic Surat Basin contains a latest Triassic–Jurassic fluvio-lacustrine succession, overlain by a Cretaceous shallow marine and coastal plain interval ([Exon, 1976\)](#page--1-0). The study wells are located in the eastern Surat Basin in southeast Queensland. Here, the Walloon Subgroup [\(Jones and Patrick, 1981; Scott et al., 2004\)](#page--1-0) crops out in an arcuate belt and dips gently south and west toward the axis of the Mimosa Syncline (Figs. 1, 2). The eastern Surat Basin unconformably overlies Palaeozoic basement (New England Orogen) and the southern Taroom Trough of the Permian–Triassic Bowen Basin. The major structural elements, the meridional Moonie–Goondiwindi and Burunga– Leichhardt thrust fault systems, initiated as active scarps in the Triassic [\(Elliott, 1993; Korsch et al., 2009](#page--1-0)) and later reactivated, producing brittle faults [\(Sliwa and Esterle, 2008](#page--1-0)) and low-amplitude anticlinal structures ([Hodgkinson and Grigorescu, 2012\)](#page--1-0) (Figs. 1, 2).

Walloon Subgroup gas content and saturation values vary significantly, and reservoirs are commonly undersaturated [\(Ryan et al., 2012; Scott](#page--1-0) [et al., 2007\)](#page--1-0). The coals are subbituminous to high-volatile bituminous C in rank, with a narrow vitrinite reflectance range $(R_0 0.35 - 0.70%)$ that generally follows basin structure [\(Beeston et al., 1997; Salehy, 1986;](#page--1-0) [Scott et al., 2007; SRK Consulting, 2008\)](#page--1-0) [\(Fig. 2](#page--1-0)). Gas contents in the eastern Surat Basin range from <1 to >15 m³/t (dry-ash-free; d.a.f.) and average 5.36 ± 2.5 m³/t d.a.f. [\(Hamilton et al., 2012](#page--1-0)), which is high for low-rank coals (cf. [Mares and Moore, 2008; Sosrowidjojo and](#page--1-0) Saghafi[, 2009; Warwick et al., 2008](#page--1-0)).

The Walloon Subgroup comprises, from oldest to youngest: the Durabilla Formation, Taroom Coal Measures, Tangalooma Sandstone, Lower Juandah Coal Measures, Juandah Sandstone and Upper Juandah

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