Reply to Comment of Smith and Pallasser on “Factors controlling the origin of gas in Australian Bowen Basin coals”

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Firstly, we wish to acknowledge the leadership and contribution that Smith and his various coworkers have made over the last three decades to the understanding of the origin of natural gas and coal seam gas, particularly through the use of stable isotopes. Our intention was in no way to criticise their views. Rather, as expressed in Boreham et al. (1998) our view was and remains “that the origin of coal seam gases especially methane, is not as straightforward as previously thought”. Indeed, Smith (1999) has recently offered an alternative abiogenic origin for methane and CO₂ via decomposition of acetic acid (Smith et al., 1998). The essence of our paper was that laboratory studies, using open system pyrolysis methods with extrapolation to geological heating rates, demonstrate that a carbon isotope composition heavier than −50‰ for methane is consistent with a thermo-genic origin.

Secondly, we acknowledge the incorrectness of a statement in Boreham et al. (1998) which credited Smith and Pallasser (1996) as reporting that they have “argued for a predominantly microbiological origin for methane and CO₂ (sourced from a magmatic origin) by methanogenic bacteria”. We inadvertently included the phrase “sourced from magmatic origin”, which has lead to this rebuff. However, this observation is also consistent with microbial reduction of magmatic CO₂ where the kinetic isotope effect is ~55‰ and the CO₂ availability is non-limiting; i.e. an open system. Additionally, coal seam gases in the Sydney Basin (Fig. 1) with CO₂ contents < 10 mol% show
both isotopically heavy (enriched in $^{13}$C) and light (depleted in $^{13}$C) CO$_2$ compared to CO$_2$ derived from a magmatic source (where CO$_2 > 10$ mol%). If the heavy CO$_2$ is indeed a marker for methanogenesis and represents the remaining CO$_2$, then the 'parent' CO$_2$ cannot solely be the isotopically light CO$_2$ since both have similarly low mol% (Fig. 1). The parent CO$_2$ must have had an originally higher mol%, possibly within the range identified as having a magmatic origin. Hence, the magmatic CO$_2$ could play an active role in the bacterial alteration process.

In their Comment, Smith and Pallasser make particular reference to our lack of recognition of their large dataset of over 300 seam gas analyses and the restriction of our experience to experimental studies. With respect to the former, the purpose of our paper was not to review the available isotope datasets for coal seam gas compositions in the Bowen Basin, but rather to present new experimental results bearing on the origin of these gases. With respect to the latter our recent investigation used a range of methodologies to establish the thermal history of a set of Bowen Basin coals as well as their composition, gas chemistry and isotope composition (Glikson et al., 1995, 1999; Golding et al., 1999, 2000; Uysal et al., 2000a,b).

We present here for completeness a re-analysis of the data from Smith et al. (1984), the majority of which appears in Smith and Pallasser (1996). The frequency distribution of the $\delta^{13}$C for methane from coals of the Bowen and Sydney Basins (Fig. 2) shows a similar average isotope value for methane. However, in the Bowen Basin there is a much higher proportion of samples with $\delta^{13}$C CH$_4$ heavier than $-50\%$, suggesting a higher thermogenic component in this region. In the Sydney Basin, samples with low CO$_2$ contents show a wide range in carbon isotopes (Fig. 1). The isotopically light CO$_2$ is most likely sourced from the associated organic matter (Smith and Pallasser, 1996). Those samples with positive carbon isotope values are, we assert, residuals of methanogenesis, necessitating a higher initial CO$_2$ (see discussion above). In the Bowen Basin, no seam gases with low CO$_2$ contents have yet been analysed for their isotopic composition. However, these two processes are obviously also active in the Bowen Basin if the light and heavy isotopes for CO$_2$ in the natural gases are used as a guide. The situation is slightly different in other sedimentary basins. In the Carnarvon and Gippsland basins, Australia’s two main natural gas producing provinces, no methanogenic utilisation of CO$_2$ has been observed (although recent evidence does suggest methanogenic utilisation of CO$_2$ in the Carnarvon Basin; Crostella and Boreham, 2000). The molecular and isotopic compositions are solely governed by the degree of mixing of organic-derived (thermogenic) and inorganic-derived (mantle and/or igneous) CO$_2$ (Fig. 3).

A further complication for the Bowen Basin coals is that vitrinite reflectance and clay mineral diagenesis studies indicate thermal maturation occurred largely as a result of a short-lived hydrothermal event in the Late Triassic rather than during maximum burial in the
The exchange between CO₂ and CH₄ has not been demonstrated in laboratory pyrolysis experiments (Sackett and Chung, 1979), the extent of isotopic exchange in geothermal systems is a function of cooling rate and the final temperature of the rising geothermal fluid (Giggenbach, 1982). Carbon isotopic equilibrium between CO₂ and CH₄ in geothermal systems is unlikely at temperatures less than 250°C although equilibrium compositions from higher temperature parts of the system may be preserved with rapid temperature decline (Giggenbach, 1982; Ohmoto, 1986). The latter possibility is supported by the carbon and oxygen isotope systematics of carbonate mineralisation in Bowen Basin coals, which indicate that much of the carbonate was deposited from a mixed CO₂/CH₄ bearing fluid of relatively constant carbon isotopic composition (Boreham et al., 1998; Golding et al., 2000).

We contend that the majority of the large coal seam gas resources-in-place, estimated using an economic cutoff depth of 1200 m (Miyazaki and Korsch, 1993), are of thermogenic origin. Coal seam gases at shallow depths (< 500 m) may be more biased towards a bacterial input. However, a major contribution from a thermogenic origin for these shallow seams as proposed in Boreham et al. (1998) and, as originally suggested by Rigby and Smith (1982) and Smith et al. (1982), may have been subsequently underrated in Smith and Pallasser (1996). Other factors not yet considered that may contribute to, though not solely explain, the isotopic depletion of the methane and the leaness in wet gas content are secondary processes such as (i) diffusion of methane through and within the coal matrix, which could lead to large isotopic fractionation (Krooss et al., 1999; Prinzhofer et al., 1999), (ii) preferential ‘flushing’ of wet gas components through invasion of the magmatic CO₂ (analogous to dry gas stripping of condensable C₂⁺ hydrocarbons; Price, 1995), and (iii) selective loss of wet gas components due to uplift to near-surface conditions; notwithstanding a possible abiogenic origin (Smith, 1999). Obviously, more detailed work is required to identify the origin of coal seam gases in Australian coal-bearing basins.

References


