



Isotope reversals and universal stages and trends of gas maturation in sealed, self-contained petroleum systems



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ABSTRACT

Isotope geochemistry is now a tool for shale gas exploration, largely due to the association of isotope reversals with mature, highly productive shale gas. Its utility, however, depends on an understanding of the isotope systematics for the particular region of interest, as well as for shale gas maturation in general. This paper reviews and re-examines isotope data from four published papers that include shale gas from the Barnett and Fayetteville Shales (Rodríguez and Philp, 2010; Zumberge et al., 2012), and gas from fractured reservoirs in the Appalachians (Burruss and Laughrey, 2010) and the Foothills of the Western Canada Sedimentary Basin (WCSB) (Tilley et al., 2011). New shale and tight sandstone gas data are also presented for the WCSB. Comparisons of these data show that the progression through three stages of gas maturation (pre-rollover zone, rollover zone and post-rollover zone) is universal in sealed, self-contained petroleum systems and that each zone has characteristic isotopic relationships and trends that are seen in all areas examined.

Gases in the pre-rollover zone are isotopically normal ($\delta^{13}\text{C}_{\text{methane}} < \delta^{13}\text{C}_{\text{ethane}} < \delta^{13}\text{C}_{\text{propane}}$) unless mixing of gases from different sources has occurred. In the rollover zone, $\delta^{13}\text{C}_{\text{ethane}}$ and $\delta^{13}\text{C}_{\text{propane}}$ become progressively more negative as $\delta^{13}\text{C}_{\text{methane}}$ becomes less negative, and ethane and methane are reversed ($\delta^{13}\text{C}_{\text{ethane}} < \delta^{13}\text{C}_{\text{methane}}$) only towards the most mature portion of the rollover zone. At the beginning of the post-rollover zone, $\delta^{13}\text{C}_{\text{ethane}} < \delta^{13}\text{C}_{\text{methane}}$, but as $\delta^{13}\text{C}_{\text{ethane}}$ and $\delta^{13}\text{C}_{\text{propane}}$ become increasingly less negative at varying rates, ethane and propane may or may not be reversed with respect to each other at the highest maturities. $\delta^2\text{H}_{\text{methane}}$ in gases of the post-rollover zone approaches isotopic equilibrium with local formation water, generally becoming more negative with increasing maturity. Correct assignment of maturity stage could be of importance because the rollover zone may represent the peak of high productivity shale gas whereas the post-rollover zone may represent a decline in productivity (Burruss and Laughrey, 2010).

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1. Introduction

Until recently, reports of isotopically reversed thermogenic gases ($\delta^{13}\text{C}_{\text{propane}} < \delta^{13}\text{C}_{\text{ethane}} < \delta^{13}\text{C}_{\text{methane}}$) were rare in the literature (Jenden et al., 1993; Dai et al., 2004; Seewald and Whelan, 2005). Their origin was poorly understood and was generally attributed to mixing of different gases. However, in the past three years, four papers that each present significant sets of data featuring isotopically reversed gases from North American sedimentary basins (Fig. 1) have been published. These include isotope data for shale gas from the Barnett and Fayetteville Shales (Rodriguez and Philp, 2010; Zumberge et al., 2012), and gas from fractured reservoirs in the Appalachians (Burruss and Laughrey, 2010) and the Foothills of the Western Canada Sedimentary Basin (WCSB) (Tilley et al., 2011). Though no longer oddities, the mechanism that created the reversed gases is still not well understood and is controversial. Burruss and Laughrey (2010) postulate mixing of gases combined with Rayleigh fractionation during redox reactions at late stages of diagenesis (250–300 °C) to explain the isotopic reversals that they observe in the Appalachians. Zumberge et al. (2012) support Tang and Xia's (2011) 'water reforming of residual organic matter in shale' mechanism whereby isotopically light ethane (and presumably propane) is created by the reaction of the resulting H_2 and CO_2 . Tilley et al. (2011) emphasize that (1) isotopically reversed gases (except where created by mixing, Jenden et al., 1993) are restricted to sealed, self-contained hydrocarbon systems ('shale gas') where gas is a mixture resulting from conventional maturation of kerogen and cracking of residual oil; and (2) isotopically reversed gases in fractured reservoirs may be migrated mature shale gas, in which case, these gases can be interpreted to be related to shale gas.

Isotope geochemistry is now a tool for shale gas exploration. Its utility however depends on an understanding of the isotope systematics for the particular region of interest as well as for shale gas maturation in general. The purpose of this paper is to review and compare data from the aforementioned papers, along with new tight gas and shale gas data from the WCSB, to derive a general model for the isotopic evolution of 'shale gas' from low to high maturity. The graphical presentation of the data given here, which is different from that in the

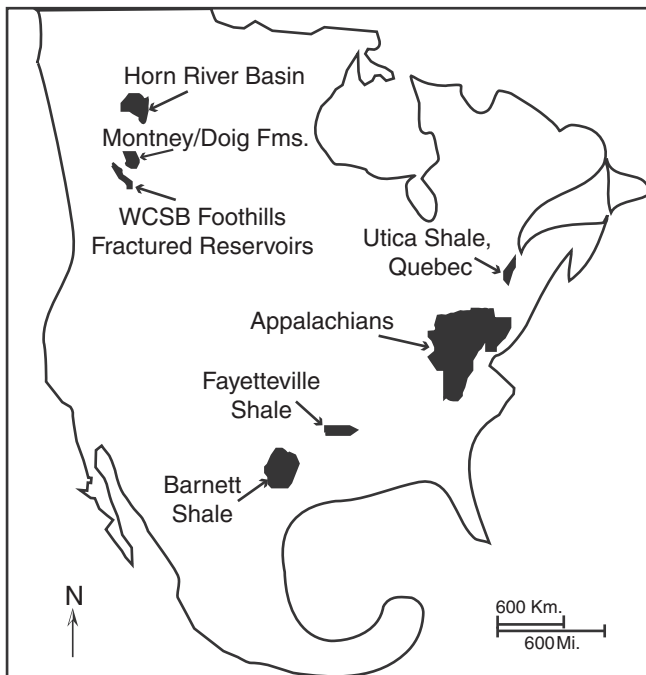


Fig. 1. Generalized map of North America showing relative locations of the sedimentary basins with isotopically reversed gases that are examined in this study. WCSB = Western Canada Sedimentary Basin.

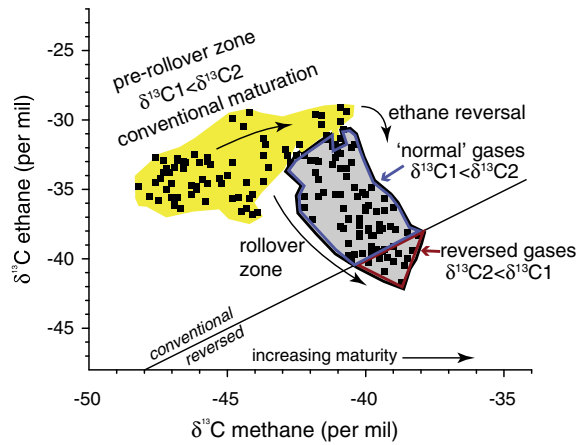


Fig. 2. $\delta^{13}\text{C}_{\text{methane}}$ (C1) versus $\delta^{13}\text{C}_{\text{ethane}}$ (C2) cross plot illustrating the distinction between the ethane reversal and the rollover zone (gray shaded area outlined in black) in which gases can be normal (area outlined in blue) or reversed (triangular area outlined in red). Curved black arrows indicate increasing maturity. Data are from the Barnett Shale (Zumberge et al., 2012).

previous papers, will clearly illustrate that there are three distinct stages of shale gas maturation that each have distinct isotope relationships and trends.

2. Terminology

The terms rollover, isotope reversal, and isotopically reversed gas have been used interchangeably when discussing shale gas. However, these terms are not synonymous and a clarification may be useful.

Isotope rollover refers to the change in the direction of the maturity trend of isotope ratios of one gas component (e.g. ethane) from becoming less negative with increasing maturity (or decreasing wetness) as occurs in conventional gas systems to becoming more negative with increasing maturity (or decreasing wetness). In other words, a reversal in the direction of the maturation trend of the isotope ratios for ethane occurs at the rollover. In this paper, we will refer to the rollover as a zone in which the isotope ratios of ethane become more negative with increasing maturity. Fig. 2 illustrates the rollover zone on a cross plot of methane and ethane isotope ratios (reported in per mil with respect to the VPDB zero point on the conventional carbon isotopic scale), where increasing maturity is represented by increasingly less negative

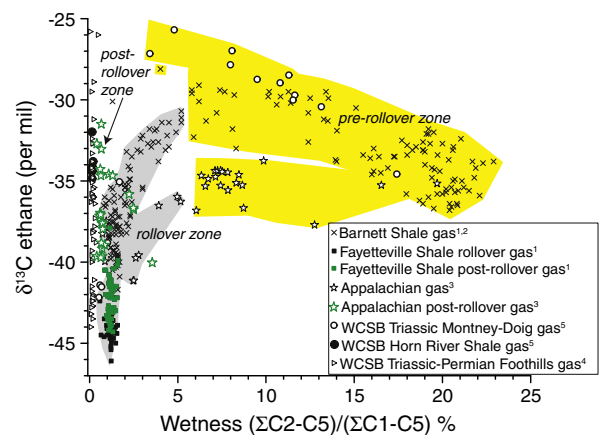


Fig. 3. Wetness versus $\delta^{13}\text{C}_{\text{ethane}}$ plot with data from areas described in this paper. Yellow shading denotes the pre-rollover zone, gray – the rollover zone, and green squares and stars are post-rollover zone gases in the Fayetteville Shale and the Appalachians, respectively. Triangles and solid black circles at $\delta^{13}\text{C}_{\text{ethane}} > -40\text{‰}$ also represent post-rollover zone gases. Data sources: ¹Zumberge et al. (2012), ²Rodriguez and Philp (2010), ³Burruss and Laughrey (2010), and ⁴Tilley et al. (2011), ⁵Table 1.

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