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Origin of CO_2 in Upper Devonian Duperow Formation and the Bakken Petroleum System at Kevin Dome, Northwest Montana

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ABSTRACT

Carbon capture and storage (CCS) is a key mitigation strategy in achieving global net-zero emissions. It is therefore essential to identify and characterize potential subsurface storage repositories. Natural CO_2 accumulations provide an opportunity to understand the behavior of CO_2 in the subsurface. Here, we investigate the source(s), migration, and storage of CO_2 in the Upper Devonian Duperow Formation at Kevin Dome, northwest Montana, USA within the Bakken Petroleum System. We report, major gas, stable and noble gas isotopic compositions in bulk gas samples ($n = 19$) produced from nearby hydrocarbon-bearing reservoirs at Kevin Dome and compare with CO_2 bearing fluid inclusions ($n = 24$) from the Duperow Formation. Using the same methods, bulk gas samples ($n = 9$) from the adjacent Ferguson Field, (Exshaw/Bakken Petroleum System) in southern Alberta, Canada, and fluid inclusions from the Sweetgrass Hills igneous complex ($n = 2$) were analyzed to understand CO_2 generation and the subsequent processes affecting CO_2 regionally. We find that CO_2 in the Upper Devonian Duperow Formation is magmatic in origin, likely related to the nearby Sweetgrass Hills igneous complex intrusion (~52 Ma), whereas CH_4 and N_2 gases are generated predominantly by thermogenic processes associated with hydrocarbon generation during burial. Since emplacement, most CO_2 in the hydrocarbon-bearing reservoirs of the Bakken petroleum system at Kevin Dome (~98%) and the Ferguson Field (~82%) has subsequently been dissolved into the groundwater. We employ a solubility model to calculate minimum gas/water ratios in Kevin Dome and the Ferguson Field, which are consistent with more groundwater interaction and more dissolution at Kevin Dome. Understanding the subsurface processes affecting CO_2 is critical for future CO_2 storage site selection.

1. Introduction

The potential for geologic structures to preserve (97–99% pure) CO_2 over geologic time demonstrates the viability of geological carbon sequestration, a key component of many national net-zero strategies (Smith et al., 2022; Hale et al., 2021; Allis et al., 2001). Previous studies demonstrated that CO_2 bearing geologic structures such as McElmo Dome, Bravo Dome, Sheep Mountain, Escalante Anticline, and Kevin Dome serve as nature's laboratory where scientists can evaluate the feasibility of a long-term subsurface carbon sequestration repository (Allis et al., 2001; Baines and Worden, 2004; Gilfillan et al., 2009; Bowen et al., 2009; Spangler, 2011; Spangler et al., 2020; Dubacq et al., 2012). Natural CO_2 reservoirs also provide an opportunity to study the origin of CO_2 and other gases as well as to characterize reservoir rocks and trapping systems, which will help determine if the geologic conditions are conducive to long-term carbon storage.

Kevin Dome is an historic oil and gas producing geologic structure in the Northwest United States (Collier, 1929). Hydrocarbons are trapped in Devonian, Mississippian, Jurassic, and Cretaceous reservoirs (Collier, 1929; Nordquist and Leskela, 1968; Wennekers, 1985). Importantly, Kevin Dome's Upper Devonian Duperow Formation hosts one of the largest known natural accumulations of ~96% pure CO_2 estimated at $\sim 283 \times 10^9$ m^3 (Bowen et al., 2012; Atlantic Analytical Laboratory, 2015; Onishi et al., 2019). Additionally, the saline aquifer associated with the Duperow Formation has the potential to sequester large volumes of CO_2 (~1GT) (Bowen et al., 2012). However, the source, timing

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of emplacement and subsequent processes affecting CO_2 in the Duperow Formation as well as CO_2 associated with hydrocarbon-bearing reservoirs in the Bakken petroleum and adjacent areas are not well constrained.

Several previous studies have been conducted to address the origin and migration history of natural accumulations of CO_2 across the United States. High purity (95–99%) CO_2 gas producing fields (McCallum Dome, Sheep Mountain and McElmo Dome, in Colorado; Bravo Dome, in New Mexico, St Johns Dome, in Arizona and New Mexico and Jackson Dome, in Mississippi) were shown to be magmatic in origin (Gilfillan et al., 2008, 2009; Zhou et al., 2012). Crustal CO_2 sources appear to be dominant in lower purity CO_2-rich fields, e.g., Mississippian Madison Limestone at the LaBarge Platform (65% CO_2) in Wyoming (Stilwell, 1989; Becker and Lynds, 2012; Merrill et al., 2015) and on the Colorado Plateau (> ~37% CO_2) (Craddock et al., 2017). In this study, we investigate gas sources, migration history and storage of CO_2 in the Upper Devonian Duperow Formation at Kevin Dome. We use CO_2-bearing fluid inclusions from the Duperow CO_2 reservoir as a proxy for the main gas accumulation within Kevin Dome. The proximity of the Ferguson Field (within the Western Canadian Sedimentary Basin (WCSB), southern Alberta, Canada) to Kevin Dome and its elevated CO_2 concentrations (~26%) suggests it may be genetically related. Fluid inclusions from the nearby Sweetgrass Hills (SGH) igneous complex were also examined to potentially define a CO_2 source endmember. This study aims to provide an understanding of the natural CO_2 generation migration and storage in the northernmost part of the Rocky Mountains.

2. Geologic setting of Kevin Dome, Northwest Montana

Kevin Dome is a 320 km-long, north-plunging structural culmination on the Sweetgrass Arch in northwest Montana (Fig. 1). The dome is bound to the west by the Cordilleran fold-and-thrust belt and to the east by the Williston Basin (Shepard and Bartow, 1986). Kevin DOme extends over 2590 km^2 and is composed of Cambrian, Devonian, and Mississippian, Jurassic, and Cretaceous strata with an average thickness of 1.7 km (Collier, 1929; Michener, 1934; Shepard and Bartow, 1986). More than ~3 km of sedimentary strata is estimated to have been eroded from the Sweetgrass Arch region, including Kevin Dome based on apatite and zircon (U—Th)/He thermochronology, carbonate clumped isotope thermometry and geothermobarometry proxies (Adeniyi, 2022). The Sweetgrass Arch, upon which Kevin Dome developed is a north to north-northwest plunging anticline that extends approximately 266 km from the Little Belt Mountains in Central Montana to the northeast trending Bow Island Arch of southern Alberta, Canada (Dowling, 1917; Michener,

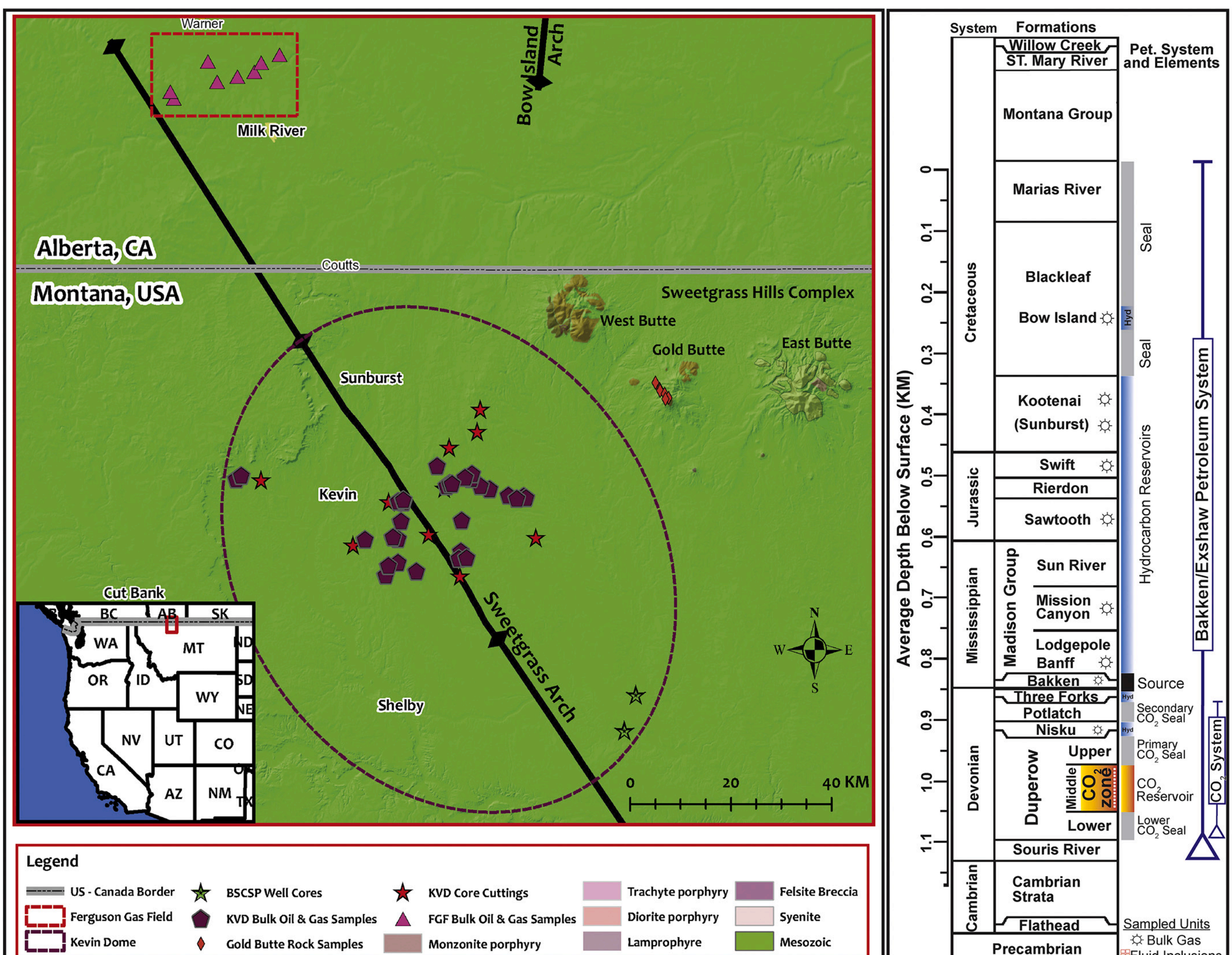

Fig. 1. Sample location at Kevin Dome, northwest Montana and the Ferguson Field, southern Alberta, Canada with a simplified stratigraphic column showing Bakken/Exshaw Petroleum System, the CO_2 System, sampled hydrocarbon-bearing reservoirs and the Middle Duperow CO_2 zone, where the fluid inclusions were collected (Peterson, 1987; Downey, 1986; Adeniyi and Steuer, 2019; Bowen et al., 2012).

1934; Tovell, 1958; Herbaly, 1974; Shepard and Bartow, 1986; Pană et al., 2021). The arch is separated into a north and south arch along the hinge of the anticline by the northeast-striking, right-lateral Pendroy Fault zone. The Pendroy fault movement causes ~48 km offset and is thought to be associated with the development of Kevin Dome as a bulge on top of the Sweetgrass Arch crest (Shepard and Bartow, 1986). Kevin Dome has repeatedly experienced episodes of uplift and subsidence, with deposition punctuated by erosional events at different stages during its evolution (Sloss, 1950; Peterson, 1966; Lochman-Balk and Wilson, 1967; Nordquist, 1953; Peterson, 1986; Shepard and Bartow, 1986; Jorgensen, 2004). Recent work has shown exhumation at Kevin Dome during the Oligocene-Miocene, which has been tied to a regional unroofing event across Northwestern Montana and the southern part of the WCSB (Adeniyi, 2022). The SGH complex, located approximately 40 km east of Kevin Dome, consists of an igneous complex intruded 54–49 Ma into Phanerozoic siliciclastic and carbonate strata in Northern Montana (Rukhlov et al., 2013; Lopez, 1995; Marvin et al., 1980). These intrusions are exposed as four main buttes: West Butte, Gold Butte, Middle Butte, and East Butte (Fig. 1)

Supplementary information (SI) contains a regional geologic map (Fig. A.1) of Kevin Dome with various structural elements associated with its development.

2.1. Stratigraphy

The stratigraphy at Kevin Dome is controlled by basement collisions, faults, episodes of uplift, subsidence, and sedimentation, punctuated by several erosional events (Jorgensen, 2004; Sloss, 1950; Peterson, 1966). Basement drilling encountered granite beneath Kevin Dome (Sloss, 1950). Proterozoic strata (e.g., Belt Group) are completely absent on Kevin Dome due to erosion and uplift of the Sweetgrass Arch prior to the Mid-Cambrian (Sloss, 1950) (Fig. 1). Ordovician and Silurian carbonate strata, including lower Devonian rocks, are also absent on the dome due to Mid-Devonian uplift and erosion (Peterson, 1986; Sloss, 1950). The Upper Devonian strata preserved on Kevin Dome include the Souris River, Duperow, Nisku, Three Forks, and Potlatch Formations (Sanberg, 1965; Macqueen and Sandberg, 1970). The Early Mississippian Bakken Formation is present across the dome. The Madison Group (Upper Mississippian) lies unconformably above the underlying Bakken Formation and is unconformably overlain by Jurassic strata (Sloss, 1950; Nordquist, 1953). The overlying Jurassic and Cretaceous layers are predominantly siliciclastic rocks. Jurassic strata on Kevin Dome include the Sawtooth, Rierdon, Swift, and Morrison Formations while Cretaceous strata include the Kootenai Formation, the Colorado Group, and the Montana Group with their respective formations (Fig. 1). A detailed depositional history and facies analysis study of the CO_2 zone within the Duperow Formation at Kevin Dome and the Banff Formation at the Ferguson field are presented in SI B.

2.2. Bakken petroleum system

The organic-rich Bakken Formation is the primary regional source rock found in Northwestern Montana, therefore the petroleum system at Kevin Dome is referred to as the Bakken petroleum system. In southern Alberta, the Bakken petroleum system is recognized as the Exshaw petroleum system, after the Exshaw Formation, which is equivalent to the Bakken Formation at Kevin Dome and the Williston Basin (Schenk et al., 2018). Based on similar depositional environments, Bakken Petroleum System reservoirs at Kevin Dome can be broadly grouped into Devonian-Mississippian and Jurassic-Lower Cretaceous units. The Devonian-Mississippian reservoirs are mainly carbonate units, consisting of the (Upper Devonian) Duperow and Nisku Formations, and the (Mississippian) Madison Group; while the Jurassic-Lower Cretaceous reservoirs are sandstones consisting of the Swift, Kootenai (Sunburst and Cutbank members) and Bow Island Formations. Though not all the reservoirs that are producing in the Bakken petroleum system elsewhere are present or producing at Kevin Dome, Madison Group is the major conventional oil reservoir on the dome with production coming from the Lodgepole and Mission Canyon Formations, while the Nisku Formation is the major oil-bearing reservoir to the east of Kevin Dome (Romine, 1929; Tonnsen and Montana Oil and Gas Fields Symposium, 1985; Bowen et al., 2009; Spangler, 2011). Seals within the Bakken petroleum system are created mainly by carbonate to evaporite facies changes, shales, paleosol and karst systems (Dyman, 1995). Evaporite and clastic shales serve as seals in the Devonian-Mississippian strata, while only shales seal the Jurassic-Lower Cretaceous reservoirs. The Potlach Formation (anhydrite) serves as the main tight regional seal above the Devonian-Mississippian reservoirs while the Rierdon and Blackleaf shales provided regional seals for the Jurassic-Lower Cretaceous reservoirs (Fig. 1).

3. Methods

Bulk oil and gas samples and/or fluid inclusion isotopic geochemistry proxies are useful for determining the origin(s) of natural gases (Aulstead et al., 1988; Goldstein, 2001; Zhou et al., 2003; Tseng and Pottorf, 2003; Gilfillan et al., 2008) and providing a geochemical snapshot of a given gas reserve. In contrast, fluid inclusions preserve the geochemical history of subsurface fluid sources, movement, migration, and accumulation (Parnell et al., 2001). The CO_2 at Kevin Dome was characterized using molecular compositional analysis, stable and noble gas isotope analyses of the CO_2-bearing fluid inclusions extracted from the Duperow Formation and igneous fluid inclusions extracted from Gold Butte (SGH) rocks. Bulk gas samples were also analyzed from hydrocarbon-bearing formations at Kevin Dome and the Ferguson Field to gain a broader understanding of processes affecting the CO_2.

3.1. Sampling and analyses

3.1.1. Fluid Inclusions

The geochemical composition of fluid inclusions can be used to evaluate the origin and evolution of subsurface fluids in different geologic settings (e.g., Roedder, 1984; Hilton et al., 1993, 2011; Merrill et al., 2015; Barry et al., 2015; Horton et al., 2021). CO_2-bearing fluid inclusions ($n = 24$) were extracted from the Duperow Formation and igneous fluid inclusions (n = 2) from the Gold Butte (SGH) igneous complex, northwest Montana (Fig. 1).

Duperow Formation and Gold Butte (SGH) fluid inclusions were analyzed at the WHEEL Noble Gas Lab, Ohio State University, and in the Barry Lab, Woods Hole Oceanographic Institution, respectively. Thin sections from Duperow drilled core and Gold Butte (SGH) outcrop samples were examined to determine the nature and overall distribution of fluid inclusions prior to isotopic analyses (SI C). Olivines were targeted in the Gold Butte (SGH) samples, as they best preserve the chemistry of the fluids present during emplacement of the intrusion (e.g., Barry et al., 2015; Horton et al., 2021). Selected samples were crushed under vacuum to release any trapped volatiles in fluid inclusions (e.g., Norman and Moore, 1999). The released gases were then purified and measured using mass spectrometry. Sampling and sample preparation, including the full analytical procedures employed at each lab, are summarized in SI C.

3.1.2. Bulk gas

Samples were collected directly from well heads, at Kevin Dome and the Ferguson Field, using (150 or 300 cm^3) double-ended stainless-steel cylinders, following standard ISOTECH Laboratories, Inc., procedures. Produced gases ($n = 19$) were collected from the Bow Island, Sunburst, Kootenai, Swift, Bakken, Madison, and Nisku hydrocarbon-bearing reservoirs at Kevin Dome, while additional samples ($n = 9$) were analyzed from the Banff Formation in the adjacent Ferguson Field, southern Alberta, Canada (Fig. 1).

Major gas abundances were determined by gas chromatography (GC), while the isotopic ratios of the main gases were measured by gas chromatography combustion isotope ratio mass spectrometry (GC/C/IRMS) techniques at ISOTECH Laboratories, Inc. Noble gas elemental compositions and isotopic ratios were analyzed at the WHEEL Noble Gas Lab, Ohio State University, following procedures outlined in Darrah et al. (2014 and 2015). Well details and full analytical procedures employed at each lab are summarized in SI C.

4. Results

Gas compositions and isotope ratios for fluid inclusion and bulk gas samples are provided in SI D table D.1 to D.9.

4.1. Duperow CO_2-bearing fluid inclusions

CO_2 is the most abundant gas (Fig. 2) found in the Duperow fluid inclusions (54.55–99.33%) and analytical limitations prevented other gases from being quantitatively measured. Noble gas isotopic ratios were determined using static mass spectrometry. Air normalized $^3He/^4He$ (R/R_A) values, where R_A is the ratio in air, ranged from 0.187 ± 0.002 to 0.460 ± 0.006 R_A, while $^{20}Ne/^{22}Ne$ and $^{21}Ne/^{22}Ne$ were mostly within error of air, ranging from 9.774 ± 0.055 to 9.949 ± 0.064 and 0.028 ± 0.0003 to 0.030 ± 0.0003 respectively, the non-air-like samples plot on the air-MORB mixing line (Fig. 4b). $CO_2/^3He$ ranged from 3.40 ± 0.23 × 10^8 to 1.35 ± 0.08 × 10^{10}.

4.2. Gold butte (SGH) igneous fluid inclusions

Within the Gold Butte (SGH) olivine mineral fluid inclusion, helium isotope ratios ranged from 0.03 ± 0.01 to 0.68 ± 0.03 R_A, while $^{20}Ne/^{22}Ne$ and $^{21}Ne/^{22}Ne$ ranged from 9.29 ± 0.08 to 9.85 ± 0.09 and 0.04 ± 0.001 to 0.10 ± 0.002, respectively.

4.3. Hydrocarbon-bearing reservoirs

Kevin Dome hydrocarbon-bearing reservoirs contain higher CH_4 (61.48–93.06%) than those observed in the Ferguson Field (35.35–56.07%). The concentration of CO_2 and N_2 is higher in the Devonian-Mississippian reservoirs at Kevin Dome and Ferguson Field compared to the Jurassic-Lower Cretaceous reservoirs at Kevin Dome (Fig. 2). CO_2 is found in trace concentrations (0.10–3.39%) in the Jurassic-Lower Cretaceous reservoirs at Kevin Dome, whereas CO_2 in the Ferguson Field's Early Mississippian Banff Formation ranged to higher values between 17.24 and 40.34%. Kevin Dome's Devonian-Mississippian reservoirs (e.g., Madison and Bakken Formations) show elevated CO_2 concentrations of 33% and 11% respectively (Fig. 2 and Table D.4). Also, N_2 concentrations in the Ferguson Field are higher, ranging from 5.88 to 12.24%, while N_2 concentrations within Kevin Dome's reservoirs are slightly lower (1.28 to 8.59%). The concentration of ethane and propane at Kevin Dome were lower to those in the Ferguson Field (Table D.4). No hydrogen was detected at the Ferguson Field, however trace amounts (0.01–2.54%) were measured in some Kevin Dome samples. Helium (0.03–0.27%) and argon (0.01–0.06%) concentrations were within similar ranges in both fields. $^3He/^4He$ ratios ranged from 0.151 ± 0.01 to 0.276 ± 0.01 R_A and 0.152 ± 0.03 to 0.171 ± 0.03 R_A, $^{20}Ne/^{22}Ne$ ranged closely from 9.708 ± 0.05 to 9.806 ± 0.05 and 9.70 ± 0.07 to 9.75 ± 0.07, while $^{21}Ne/^{22}Ne$ also ranged closely from 0.029 ± 0.002 to 0.033 ± 0.002 and 0.030 ± 0.003 to 0.032 ± 0.003 at Kevin Dome and the Ferguson Field, respectively, suggesting mixing between air and a nucleogenic ^{21}Ne enriched crustal component. $CO_2/^3He$ ranged from 2.16 ± 7.80 × 10^5 to 5.46 ± 0.21 × 10^8 at Kevin Dome and 1.54 ± 0.06 × 10^8 to 1.27 ± 0.05 × 10^9 at the Ferguson Field.

5. Discussion

5.1. Origin of Gasses

High CO_2 (54.55–99.33%) concentrations in Duperow fluid inclusions (Fig. 2) are consistent with the high CO_2 composition (~96%) previously measured in free gas phase samples from the Duperow at Kevin Dome (Bowen et al., 2012; Atlantic Analytical Laboratory, 2015). The Duperow Formation is the primary reservoir where free phase CO_2 is observed in the region (Lee, 1962; Lane, 1987; Lake and Whittaker, 2006; Bowen et al., 2012; Ryerson et al., 2013) and our CO_2-rich fluid inclusions data support this interpretation. Elevated CO_2 concentrations within the Devonian – Mississippian reservoirs at Kevin Dome and Ferguson Field, compared to Jurassic and Lower Cretaceous reservoirs at Kevin Dome (Fig. 2), are likely a result of stratigraphic proximity to CO_2 source or to the Duperow Formation. The relationship between the three main gas phases (CH_4, N_2 and CO_2) at Kevin Dome and the Ferguson Field can be used to determine the sources of gas in the region (SI Fig. E.1). For example, CO_2 is negatively correlated with CH_4 and N_2 in both study areas (Fig. E.1a-b), while CH_4 and N_2 are positively correlated, except for the Lower Jurassic – Cretaceous reservoirs, potentially due to gas mixing from shallower reservoirs (SI Fig. E.1c). This suggests that CH_4 and N_2 likely have the same source, whilst CO_2 has a different source.

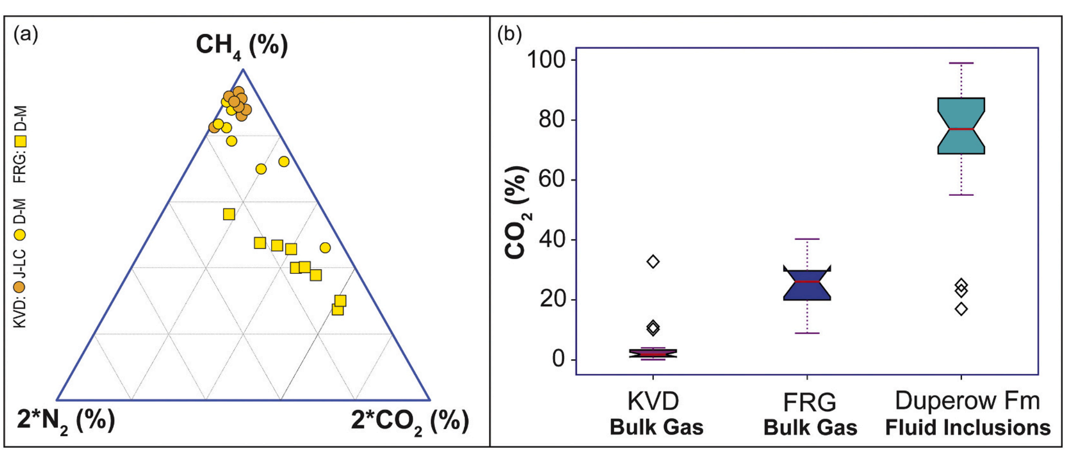

Fig. 2. Gas composition at Kevin Dome, northwest MT USA and Ferguson Field, southern AB Canada: (a) shows bulk gas samples major gas composition; where CH_4 is the main gas component with elevated CO_2 and N_2 in the D-M hydrocarbon reservoirs at both study sites (b) shows bulk gas samples CO_2 proportions at both study sites compared with those in the Duperow Formation CO_2 reservoir fluid inclusions; where CO_2 exits from trace (in the J-LC hydrocarbon reservoirs) to elevated proportions (in the D-M hydrocarbon reservoirs) and CO_2 is the main gas component in the Duperow fluid inclusions. CO_2 and N_2 compositions in (a) were multiplied by a factor of 2 to better visualize the variations. **KVD** – Kevin Dome; **FRG** – Ferguson Field; **J-LC** – Jurassic-Lower Cretaceous and **D-M** – Devonian-Mississippian.

5.2. Origin of CH_4 and N_2

Stable isotope signatures of natural gases are indicative of their origin, and when combined with total gas chemistry, can help delineate gas sources and subsequent post-genetic alterations (e.g., Bernard et al., 1976, 1977, 1978; Whiticar et al., 1986; Whiticar, 1999; Norman and Moore, 1999; Liu et al., 1997; Milkov, 2011; Milkov and Etiope, 2018). Using a Whiticar plot (Fig. 3a, Whiticar et al., 1986), CH_4 at Kevin Dome appears to be a mix of oil-associated thermogenic gas and secondary microbial gas, while those in the Ferguson Field appears to be a mix of primary microbial (methyl-type fermentation), abiotic and early mature thermogenic gas. However, using a Bernard plot (Fig. 3b), the CH_4 in both study areas plots as early mature thermogenic and oil-associated thermogenic gas. The "oil associated thermogenic" CH_4 at Kevin Dome and Ferguson Field is consistent with oil being present within the reservoirs. N_2 in all the bulk gas samples suggests an organic and/or deep crustal (thermogenic) origin (Fig. 3f and SI Fig. E.2). Stable isotopic systematics from all the schemes suggest mostly thermogenic gas sources for CH_4 (e.g., Fig. 3d) and N_2, with some microbial source mixing within the Bakken petroleum system at Kevin Dome and Ferguson Field. Methane could be from the thermal evolution of organic matter, and/or rocks during burial, with additional inputs from the biodegradation of generated hydrocarbon, while N_2 could be from the thermogenic evolution of organic matter during petroleum generation of clay minerals in mudrocks within the Bakken petroleum system (Norman and Moore, 1999; Zhu et al., 2000; Faramawy et al., 2016).

5.3. Origin of CO_2

The CO_2-rich fluid inclusions within the Duperow Formation are located within calcite grain-filled fractures and rock matrix, indicating they formed after crystal growth and are secondary in nature (SI C). This means they contain information about subsurface fluid processes such as the passage of different fluid pulses or those related to later stages of deposition (Polito et al., 2001; Fairmaid et al., 2011; Kendrick and Burnard, 2013).

The generation of CO_2 at Kevin Dome could be a result of microbial, thermogenic, or magmatic processes. Microbial CO_2 could be generated from the bacterial oxidation of existing hydrocarbons. Thermogenic CO_2 could be sourced from either the decomposition of organic-rich Bakken Shale sequences beneath the Disturbed Thrust Belt, west of the dome (Stein, 2008; Snowdon, 1999), or as a byproduct of thermal degassing of Paleozoic carbonate rocks during the SGH igneous complex intrusion, east of the dome (Bowen et al., 2012; Snowdon, 1999). Mantle derived CO_2 could also have accumulated in the Duperow during the SGH igneous complex intrusion or from the mantle along basement faults, since Kevin Dome sedimentary strata sit directly on Archean basement (Xianbin et al., 1998; Stevens et al., 2005; Zhou et al., 2003).

Stable isotope-based CO_2 classification schemes of bulk gas samples suggest a thermogenic CO_2 source with some secondary microbial gas contributions, especially in the CO_2-rich Devonian-Mississippian reservoirs (Fig. 3c-d, section 5.2). In contrast, the $\delta^{13}C_{CO2}$ and $\delta^{18}O_{CO2}$ suggest a mix of both magmatic and microbial CO_2 sources in both study areas (Fig. 3e).

The mantle has a distinct $CO_2/^3He$ range from 1 to 3 × 10^9 (Marty and Jambon, 1987), in contrast, crustal CO_2 has a higher $CO_2/^3He$, up to an order of 10^{13}, due to the addition of CO_2 with no associated mantle ^3He (e.g., O'Nions and Oxburgh, 1988; Sherwood Lollar et al., 1997; Gilfillan et al., 2008; Ballentine et al., 2001; Barry et al., 2013, 2022). Therefore, helium and CO_2, when combined, are powerful indicators of gas origin. In contrast, $CO_2/^3He$ below mantle values are suggestive of CO_2 loss from the system (e.g., Sherwood Lollar et al., 1997; Gilfillan et al., 2008; Tyne et al., 2021). Here, we find that Duperow fluid inclusions have $CO_2/^3He$ values (3.34 ± 0.187 × 10^9 avg.) overlap with the magmatic range, whereas Kevin Dome (0.09 ± 0.003 × 10^9 avg.) and Ferguson Field (0.59 ± 0.029 × 10^9 avg.) bulk gases have $CO_2/^3He$ below (Fig. 4a) the mantle range. This suggests the CO_2 is magmatic in origin and subsequent gas phase loss has likely occurred. A portion of the fluid inclusion neon isotope data (SI Table D.1) plot on a trend that is consistent with magmatic contributions into the Duperow fluid inclusions. In contrast, all bulk gas samples are enriched in nucleogenic ^{21}Ne and consistent with crustal radiogenic overprinting (Fig. 4b). Notably, helium isotopes in all the hydrocarbon-bearing gas samples and fluid inclusions are also consistent with significant radiogenic production, which has overprinted any potential magmatic signature (Albarède, 2001; Barry et al., 2016). Using the He isotope endmember values in the mantle, and crust, we estimate the helium is approximately 98% crustal and 2% mantle derived within the petroleum systems at Kevin Dome (Ballentine and O'Nions, 1992, 1994; Ballentine and Burnard, 2002).

5.3.1. Quantifying CO_2 loss from Kevin Dome and Ferguson Field hydrocarbon-bearing reservoirs

Dissolution has previously been shown to be a CO_2 sink in large gas fields (Gilfillan et al., 2008, 2009; Zhou et al., 2012; Craddock et al., 2017). The extent of CO_2 dissolution can be estimated using $CO_2/^3He$, whereby a decrease in this ratio in the residual gas phase could indicate CO_2 loss for example due to dissolution, precipitation, or microbial consumption (e.g., Gilfillan et al., 2008; Barry et al., 2019; Tyne et al., 2021). CO_2 is typically undersaturated in groundwater, and thus could serve as a sink for CO_2 if it encountered a CO_2-rich gas system. Assuming the Duperow fluid inclusions record the theoretical initial $CO_2/^3He$ composition for Kevin Dome, we use a Rayleigh Fractionation approach to model $CO_2/^3He$ and $\delta^{13}C$-CO_2 signatures (Gilfillan et al., 2009) with dissolution.

$$\boldsymbol{\delta^{13}C - CO_2 = \delta^{13}C - CO_{2i} + \varepsilon_{DIS}}\ ln\ \mathbf{f} \qquad (1)$$

where $\delta^{13}C$-CO_2 and $\delta^{13}C$-CO_{2i} are the measured and assumed initial $\delta^{13}C$-CO_2 values of the gas, respectively. f is the fraction of CO_2 remaining in the reservoir. ε_{DIS} is the carbon isotope fractionation for dissolution, which is a function of reservoir temperature, salinity, and pH conditions. We estimate a pH range for Kevin Dome and the Ferguson Field hydrocarbon reservoirs ranging from 5.8 to 6.3 and 6.5 to 8.3, respectively. Table 1 shows the average estimated pH and fraction of CO_2 dissolved within Kevin Dome and Ferguson Field hydrocarbon reservoirs (see SI F for the method summary, parameters and assumptions used). Using the average pH or Kevin Dome (6.12) and the Ferguson Field (7.25) hydrocarbon reservoirs, our model (Fig. 6) predicts that 98 ± 3% and 82 ± 13% of CO_2 gas, respectively, has been dissolved into surrounding fluids. This suggests that the shallower (<1 km) Kevin Dome gas reservoir has had a greater extent of groundwater interaction, leading to more CO_2 dissolution, whereas the deeper (>1 km) Ferguson Field has had less groundwater flow and thus has less CO_2 dissolution. In contrast, Duperow fluid inclusions do not interact with groundwater interaction and preserve the isotopic composition of the initial CO_2 charge with no evidence for CO_2 dissolution. Therefore, differing groundwater interaction controls the various isotopic signatures recorded in the different sample types.

5.3.2. Constraining Kevin Dome and Ferguson Field hydrocarbon-bearing reservoirs gas-water ratios

Radiogenic ^4He and groundwater-derived ^{20}Ne in the gas phase increase as a result of them being stripped from the groundwater (Gilfillan et al., 2009). Therefore, the higher ^4He and ^{20}Ne concentrations observed in the Kevin Dome are also consistent with greater groundwater interaction in these samples compared to the Ferguson Field (Fig. 5). To quantify the amount of groundwater interaction in both fields we use a zero-order (ZO) total gas-stripping model (Eq. (2)) to estimate gas-water (G/W) ratios within the reservoirs after Barry et al. (2016). The ZO model quantifies the minimum volume of water required to produce the measured concentrations of noble gas *(i)* assuming

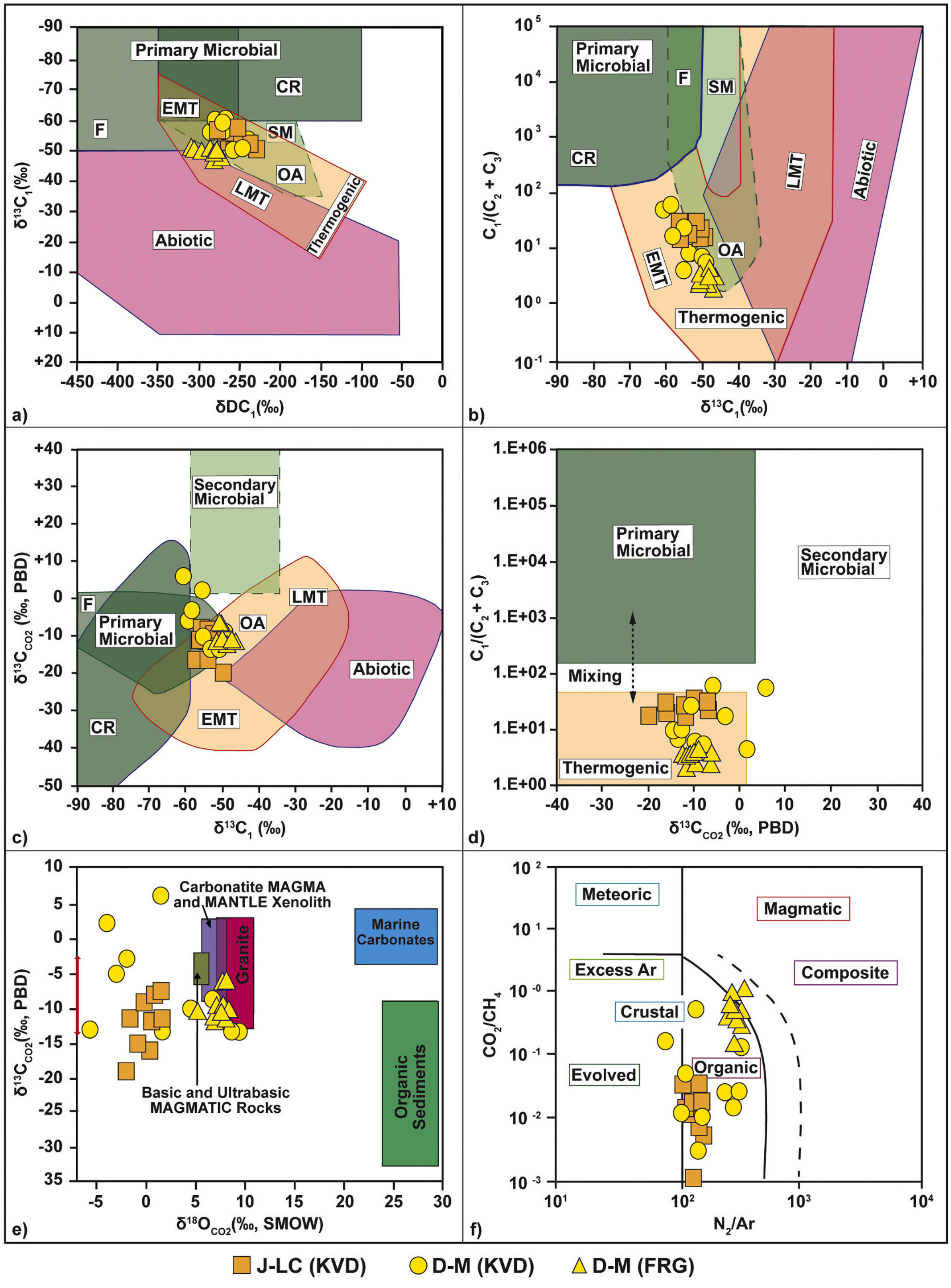

Fig. 3. CH_4, CO_2 and N_2 gas origins at Kevin Dome, northwest MT USA and Ferguson Field, southern AB Canada using bulk gas stable isotopes signature: (a) Whiticar and (b) Bernard schemes show CH_4 gas generated mainly by thermogenic processes with inputs from secondary microbial generation from associated oil (Whiticar et al., 1986; Whiticar, 1999; Bernard et al., 1976, 1977; Milkov and Etiope, 2018) (c) Modified CO_2 gas origin classification scheme (Gutsalo and Plotnikov, 1981; Milkov and Etiope, 2018) (d) Modified Bernard scheme type plot for CO_2 origin based on gas dryness ($C_1/C_2 + C_3$) vs $\delta^{13}C_{CO2}$ (Bernard et al., 1978; Milkov, 2011) (e) $\delta^{13}C_{CO2}$ vs $\delta^{18}O_{CO2}$ shows CO_2 gas of mixed origin (primary microbial and magmatic sources) in both study areas (Liu et al., 1997) (red arrow highlights where magmatic and microbial $\delta^{13}C_{CO2}$ signatures in this study overlap – *see section 5.3*) and (f) CO_2/CH_4 vs N_2/Ar ratio shows N_2 gas generated from organic and/or deep crustal (thermogenic) sources (Norman and Moore, 1999). **KVD** – Kevin Dome; **FRG** – Ferguson Field; **J-LC** – Jurassic-Lower Cretaceous and **D-M** – Devonian-Mississippian hydrocarbon reservoirs. Gas origin domains: **CR** – CO_2 reduction; **F** – methyl-type fermentation; **SM** – secondary microbial; **EMT** – early mature thermogenic; **OA** – oil-associated thermogenic and **LMT** – late mature thermogenic. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

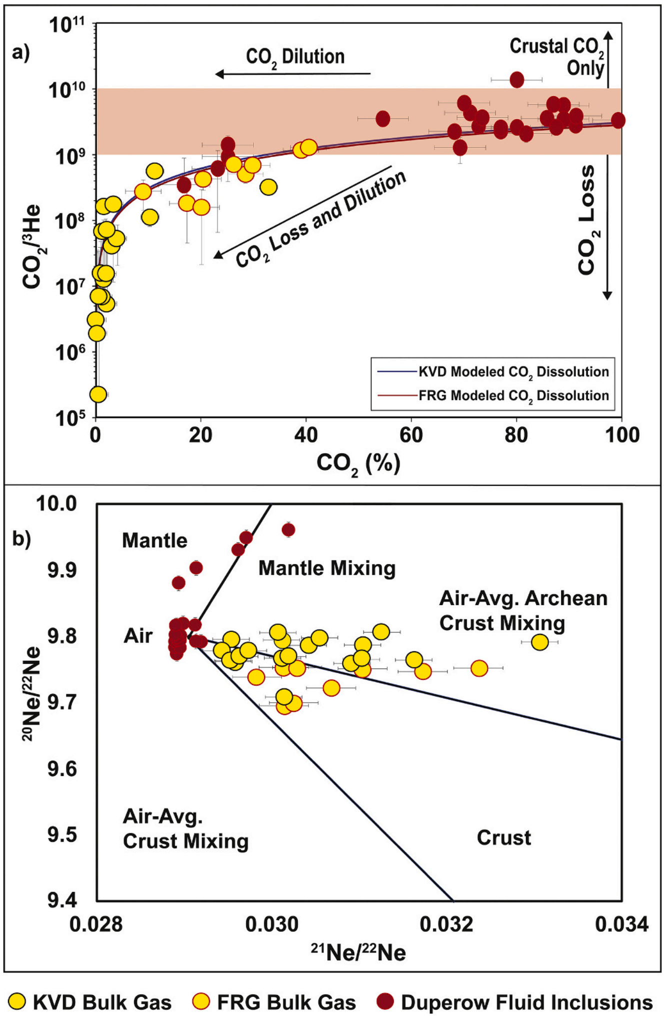

Fig. 4. CO_2 origin at Kevin Dome, northwest MT USA and Ferguson Field, southern AB Canada using bulk gas and fluid inclusions noble gases signatures: (a) $CO_2/^3He$ vs CO_2 (%) shows Duperow fluid inclusions within the magmatic range, while Kevin Dome and Ferguson Field bulk gas samples plot below the magmatic range (b) Neon isotopic ratios ($^{20}Ne/^{22}Ne$ vs $^{21}Ne/^{22}Ne$) shows slight magmatic influence in the Duperow fluid inclusions while Kevin Dome and Ferguson Field bulk gas samples plots within air-crust mixing domains. The red-shaded region in (a) is the magmatic range as defined by Marty and Jambon (1987) while the modeled CO_2 dissolution evolution pathways are represented by the blue and red lines for Kevin Dome and Ferguson Field respectively (see SI F for the CO_2 dissolution model estimation summary). Solid lines in (b) represent air, Precambrian crust, and Archean crust endmembers (Ballentine and O'Nions, 1992, 1994; Ballentine and Burnard, 2002) KVD – Kevin Dome; FRG – Ferguson Field. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

complete removal from the water into the gas phase (Barry et al., 2016). It is expressed as:

$$G/W(ZO) = \left[C_{i(ASW)}\right]/Z^{*}\left[C_{i(meas.)}\right] \quad (2)$$

where $[C_{i(ASW)}]$ and $[C_{i(meas.)}]$ are the initial concentrations of noble gas (i), in air-saturated water (ASW) and the measured gas phase, respectively. Z is the compression factor for converting all parameters measured in STP (273.15 K at 1 bar) to reservoir temperature and pressure (RTP) and is defined as $Z = 273.15{*}P/T$ (Barry et al., 2016). Using the measured ^{20}Ne and an assumed ^{20}Ne concentration in ASW of 1.51×10^{-7} cm^3STP/g (Kipfer et al., 2002), we estimate a G/W of 0.00024 ± 0.0001 and 0.00033 ± 0.0001 at Kevin Dome and Ferguson Field, respectively (Table 2). A lower G/W indicates that relatively more groundwater has interacted with the gas in the system and thus there is likely greater groundwater flow (Barry et al., 2016; Tyne et al., 2022). The lower G/W at Kevin Dome is consistent with greater groundwater interaction and thus more dissolution compared to the Ferguson Field (Fig. 7a). In addition, we note a slight trend with depth where the G/W decreases, likely due to groundwater flow being restricted by decreased porosity or different hydrological systems. Again, this is consistent with less groundwater movement and dissolution in Ferguson Field than the shallower Kevin Dome fields (Fig. 7b).

5.4. *Gas origin and Kevin Dome's evolution*

In this section we aim to bring together information about gas origins

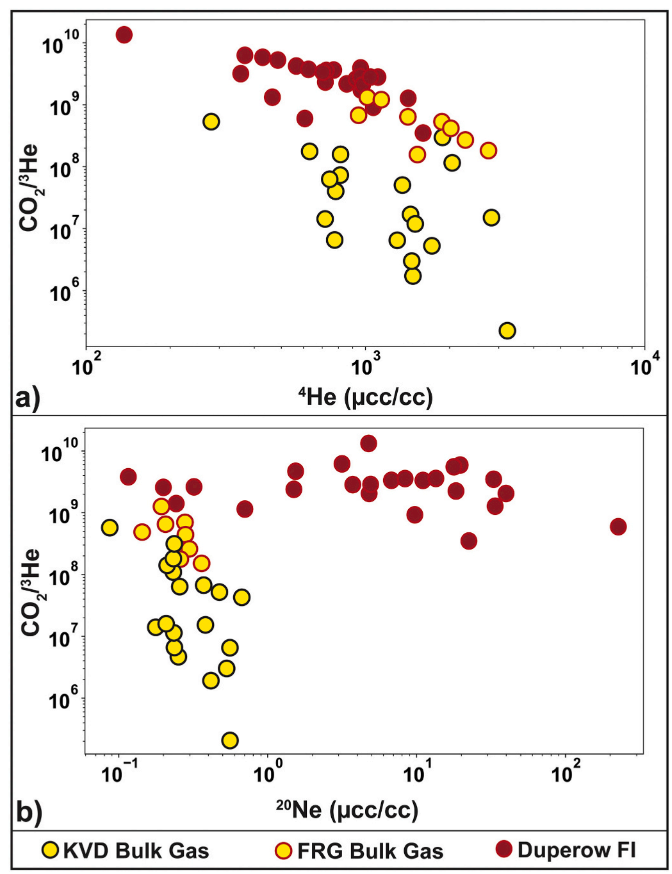

Fig. 5. Kevin Dome, northwest MT USA and Ferguson Field, southern AB Canada bulk gas data with Duperow Formation Fluid Inclusions showing decreasing $CO_2/^3He$ to increasing radiogenic (a) ^4He and (b) ^{20}Ne concentrations. Similar trends have been observed in other CO_2-rich reservoirs across the USA, China, and Europe (Gilfillan et al., 2009). FI – Fluid Inclusions.

Table 1
Average pH and fraction of CO_2 dissolved within Kevin Dome and Ferguson Field hydrocarbon-bearing reservoirs. BHT = Borehole Temperature, S = Salinity.

Reservoirs	BHT (°C)	S (M)	pH	ε_{DIS}	$\delta^{13}C - CO_2$	$\delta^{13}C - CO_{2i}$	CO_2 (%)	Depth (km)	CO_2 Dissolved (%)
Kevin Dome	26	0.17	6.12	2.29	−11.24	0	4.15	0.53	98.04
Ferguson Field	48	1.97	7.25	5.27	−9.55	0	25.55	1.280	82.02

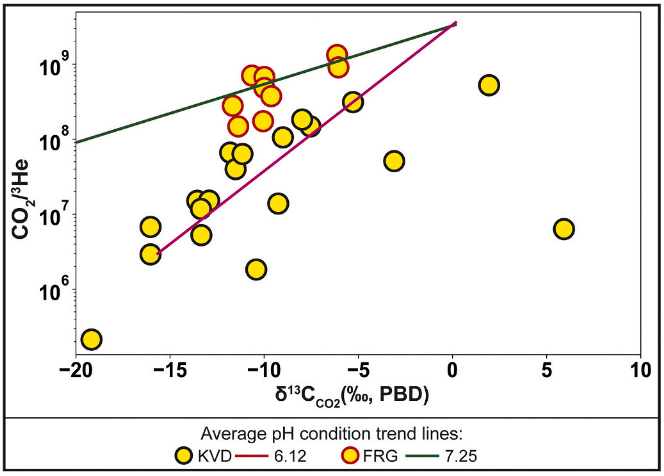

Fig. 6. CO_2 dissolution modeled with changing $CO_2/^3He$ and $\delta^{13}C$-CO_2 with the average pH conditions (colored lines) at Kevin Dome (KVD) pH = 6.12, northwest MT USA and Ferguson Field (FRG) pH = 7.25, S Alberta, Canada.

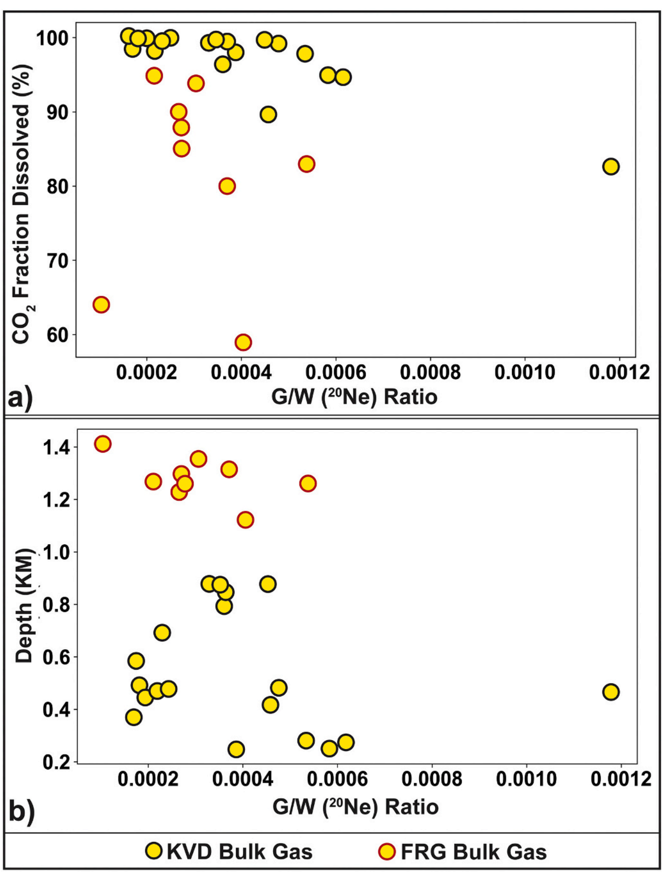

Fig. 7. ^{20}Ne G/W estimates from our zero-order (ZO) total gas-stripping model at Kevin Dome, northwest MT USA and Ferguson Field, southern AB, Canada vs (a) CO_2 fraction dissolved within the hydrocarbon reservoirs and (b) depth. G/W trends indicate increased groundwater interaction with the gas in the system that are controlled with depth, and consistent with less groundwater movement and dissolution in Ferguson Field than the shallower Kevin Dome fields.

with the geologic setting, stratigraphic framework, and overall evolutionary context at Kevin Dome. We also summarized the hydrocarbon generation events associated with the Bakken/Exshaw petroleum system as well as the CO_2 emplacement events that are presumed to have occurred at Kevin Dome. Fig. 8 illustrated the orogenic and erosional events that controlled the development and stratigraphy at northwest Montana, USA and southern Alberta, Canada.

5.4.1. *Generation, source and timing of CH_4 and N_2 at Kevin Dome*

Compositional data and stable isotope systematics of Bakken petroleum system gases, show that CH_4 and N_2 gases were generated from the thermal evolution of organic matter, and/or rocks during burial and the biodegradation of the generated hydrocarbon. This is consistent with the burial and exhumation history both at Kevin Dome and in the Ferguson Field, which has been inferred from thermochronological and thermal maturity data. These show that rocks in this region have experienced burial temperatures to the oil and gas generation windows consistent with their mixed thermogenic, biogenic origin (section 5.2; Bustin, 1991; Dolson et al., 1993; Dyman, 1995; Schietinger, 2013; Aviles and Cheadle, 2015; Dembicki Jr., 2017; Adeniyi, 2022). Oil and gas generation had begun in the Bakken petroleum system by Early Cretaceous (Fig. 8) but was shut off by an Oligocene-Miocene exhumation as a result of a regional unroofing event (Adeniyi, 2022).

5.4.2. *Generation, source and timing of CO_2 emplacement at Kevin Dome*

High purity (95–99%) magmatic CO_2 reservoirs are typically found in close proximity to magmatic centers, igneous intrusions, and/or volcanic regions where basement sutures, shear zones, or faults acted as conduits for CO_2 migration from magmatic intrusions (e.g., Gilfillan et al., 2008, 2009; Zhou et al., 2012; Karlstrom and Humphreys, 1998; Broadhead, 2005; Craddock et al., 2017). Existing seismic data suggest there is no major basement fault below Kevin Dome (Bowen et al., 2012). Therefore, the nearly pure (~ 96%) bulk gas CO_2 (Bowen et al., 2012; Atlantic Analytical Laboratory, 2015) and CO_2 in the Duperow secondary fluid inclusions is most likely magmatic in origin (based on $CO_2/^3He$), due to its proximity to the SGH igneous intrusion event (~52 Ma, (Lopez, 1995)) (Fig. 1 and 8). Elevated CO_2 concentrations in Devonian-Mississippian hydrocarbon reservoirs around Kevin Dome, northwest Montana, and adjacent regions in Canada (Lake and Whittaker, 2006; Ryerson et al., 2013), also suggest that the emplaced CO_2 has migrated into the Bakken petroleum system. The Duperow has two main porosity zones, one located in the Middle Duperow, the other located in the Lower Duperow, which are separated by a thick anhydrite (Bowen et al., 2009, 2012; Adeniyi and Steuer, 2019). The stratigraphically higher Middle Duperow is the more porous and serves as the main CO_2 zone. The overlying Potlatch Formation also provided a reliable secondary seal for the Duperow CO_2 trap (Fig. 1 and 8). The Upper Duperow has low porosity and permeability and likely serves as a baffle or seal. The emplacement of the CO_2occurred after the Early Cretaceous hydrocarbon generation and trapping at Kevin Dome (Fig. 8).

5.5. *CO_2 Dissolution and implication on carbon capture and storage*

In assessing the viability of carbon capture and storage (CCS), several studies have focused on the risks of CO_2 leakage and subsequent contamination of potable aquifers, reactivity of injected CO_2 with the host reservoir, including the various subsurface CO_2 trapping mechanisms and seal rock integrity (Moore et al., 2003; Benson and Cole, 2008; Gilfillan et al., 2009; Trautz et al., 2013; Celia, 2017; Alcalde et al., 2018). Among the various CCS options, deep saline aquifers have the largest storage capacity and multiple secondary CO_2 trapping mechanisms can occur (Bachu and Adams, 2003; Celia et al., 2015). Aside from suitable stratigraphic trapping mechanism, studies have shown that CO_2 is also effectively trapped by dissolution into brine or precipitation as carbonate minerals within deep saline aquifers such as those at Kevin Dome over geologic times (Weir et al., 1996; Allis et al., 2001; Baines and Worden, 2004; Gilfillan et al., 2009; Bowen et al., 2009; Dubacq et al., 2012; Ryerson et al., 2013; Sathaye et al., 2014). CO_2 dissolution has been suggested to be an important post-genetic solution trapping mechanism (Gilfillan et al., 2009; Sathaye et al., 2014; Zhou et al., 2012).

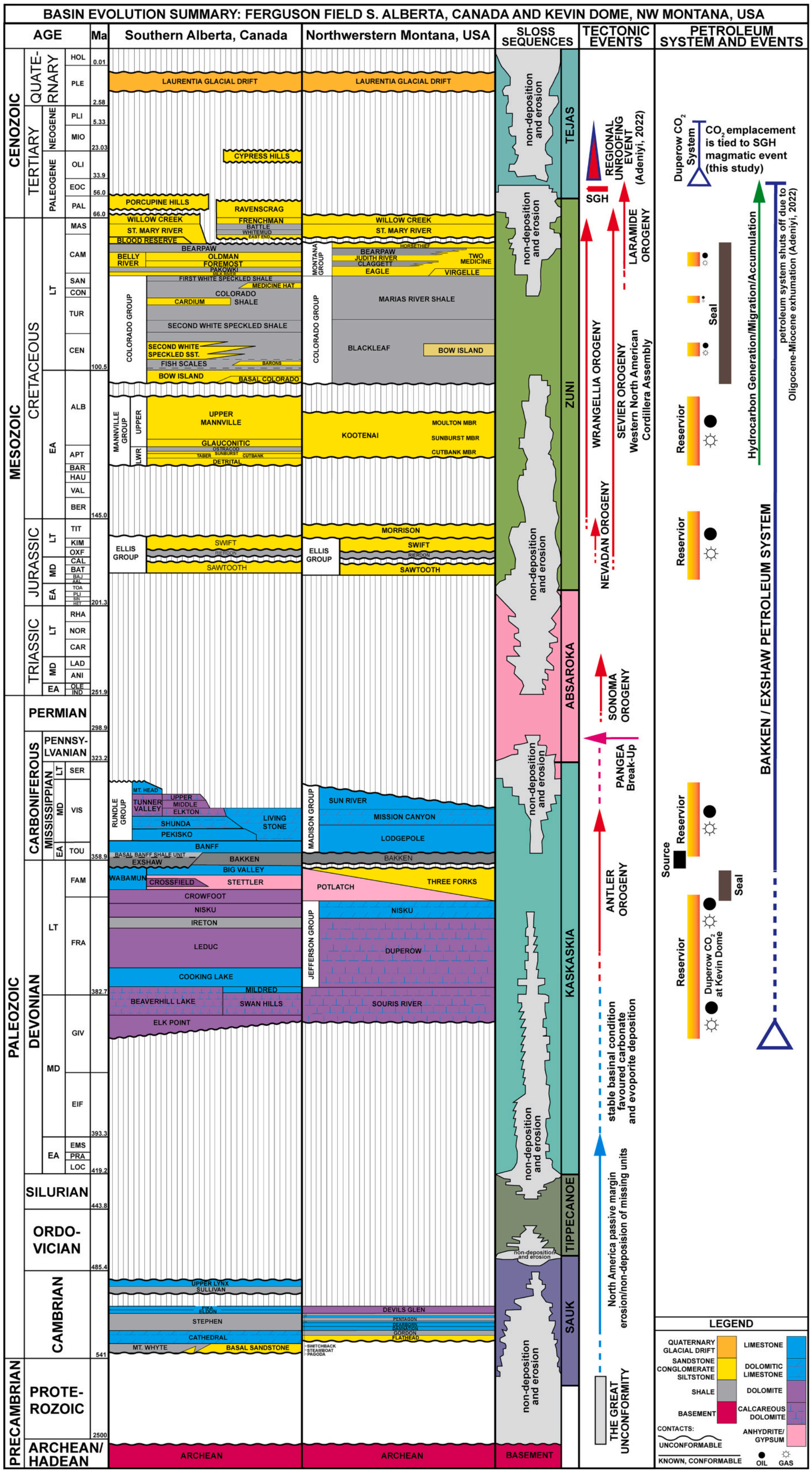

Fig. 8. Basin evolution summary showing tectonic events that controlled stratigraphy, development of the Bakken/Exshaw Petroleum System and the Duperow CO_2 system at Kevin Dome, NW Montana, USA and Ferguson Field, S Alberta Canada (Deiss, 1936; Peterson, 1987; Downey, 1986; Dyman, 1995; Smith et al., 1995; Dolson et al., 1993; Vuke-Foster et al., 2007; Dembicki Jr., 2017; Adeniyi, 2022; Chart, 2019). SGH – Sweetgrass Hills Igneous complex.

Table 2
Average ^{20}Ne G/W ratio within Kevin Dome and Ferguson Field hydrocarbon-bearing reservoirs.

Reservoirs	BHT (°C)	P (bar)	Depth (km)	^{20}Ne	^{20}Ne ASW	Z	G/W
Kevin Dome	26	149	0.530	3.5E-07	1.51E-07	1484	0.00024
Ferguson Field	48	347	1.280	2.5E-07	1.51E-07	1976	0.00033

This study shows that CO_2 that migrated into the Bakken petroleum system at Kevin Dome, and proximal fields in Canada, was subsequently dissolved into the formation waters. Here, we estimate ~98% and ~ 82% CO_2 dissolution at Kevin Dome and the Ferguson Field since emplacement ~52 Ma ago, respectively. A separate study estimated only 22% dissolution of emplaced CO_2 at the Bravo Dome over 1.2 Ma (Sathaye et al., 2014). This suggests that the extent of dissolution may vary from site to site, which is important for CCS because CO_2 dissolution has previously been shown to be one of the safest CO_2 trapping mechanisms (IPCC, 2005). This work shows the value of characterizing subsurface geochemical conditions of these systems both prior to and following injection, which is required to ensure safe CO_2 storage.

6. Conclusions

Using a multi-faceted geochemical approach, we have constrained the genetic origin and timing of CO_2 in the Upper Devonian Duperow Formation, Kevin Dome, and the Bakken petroleum system in northwest Montana, USA and southern Alberta, Canada. We find:

1. CO_2 is the main gas within the Upper Devonian Duperow Formation at Kevin Dome, whilst CH_4 is the dominant gas within the Bakken petroleum system and is associated with varying proportions of N_2 and CO_2.
2. CO_2 within the Duperow Formation at Kevin Dome is magmatic in origin (as indicated by $CO_2/^3He$) while the CH_4 and N_2 gases accumulated in the hydrocarbon-bearing reservoirs at the dome were generated mainly by thermogenic processes, with inputs from second-stage microbial generation from existing oil.
3. CO_2 emplacement at Kevin Dome is tied to the adjacent SGH igneous complex intrusion event (~52 Ma) after the Early Cretaceous hydrocarbon generation at the dome. The emplaced CO_2 primarily reservoired in the Duperow, has subsequently migrated into the other hydrocarbon reservoirs within the regional Bakken petroleum system.
4. Groundwater interaction and subsequent dissolution is consistent with CO_2 migration from a magmatic source into hydrocarbon-bearing reservoirs. Using a dissolution model, we estimate CO_2 dissolution of ~98% and ~ 82% at Kevin Dome and the Ferguson Field, respectively. We suggest that the different fractions of CO_2 dissolved into groundwater, as indicated by G/W estimates, are controlled by stratigraphic depth and different hydrological systems.

Identifying gas migration pathways and directionality relative to source as well as evaluating subsurface geochemical conditions that are favorable for CO_2 post-injection solubility and mineral trapping are important for geologic characterization of carbon sequestration target sites.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

All the data used in this study can be obtained from the figures and tables presented in the main text and supplementary information.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemgeo.2023.121733.

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