Carbon Dioxide Leakages through Fault Zones: Potential Implications for the Long-term Integrity of Geological Storage Sites

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ABSTRACT

Carbon sequestration has recently become more widely recognized as a potential means of reducing atmospheric carbon dioxide levels. Understanding the tectonic relationship of carbon dioxide discharges and the sealing behavior of faults is conducive for predicting the long-term integrity of geological storage formations. Of primary concern is the influence of crustal deformation on the carbon dioxide leakage through fault zones during large-scale underground injection. This paper examines a record of carbon dioxide leakage from a faulted, natural carbon-dioxide-rich formation, and investigates the crustal tilt in the fault zones. Temporal changes in the crustal tilt reveal pulses of carbon dioxide concentrations ranging from 537.7 up to 1317.1 ppm, and the mean level represents 890.2 ppm. Of particular interest is that each high-frequency pulse coincides with the onset of local solid-earth tide. We show a significant correlation between the crustal tilt magnitude and amount of carbon dioxide leakage. We suggest that carbon dioxide leakage levels increase owing to fracture opening, potentially caused by changes in fault architecture and permeability structure of regions surrounding the faults.

Keywords: Carbon dioxide leakage, Carbon sequestration, Crustal tilt

1 INTRODUCTION

Geological carbon sequestration is the deep injection of carbon dioxide into saline formations, depleted oil and gas reservoirs, and deep coal seams (Benson et al., 2008; Busch, 2008; Cai et al., 2019), as an alternative means either of reducing atmospheric carbon dioxide levels (Kampman et al., 2016; Chatterjee et al., 2019; Yadav et al., 2021; Basal et al., 2019), or of enhancing oil and natural gas recovery (Gunter et al., 2005; Friedmann et al., 2006; Liang et al., 2009; Huang and Tan, 2014; Yang, 2020; Rathnaweera and Ranjith, 2020). In the saline reservoirs there are two mechanisms to the key geochemical reactions. One is dissolution of carbon dioxide into the aqueous fluid, and the other is the dissolution of carbonate minerals. Geochemical reactions in the reservoir can be evaluated by considering dissolution rate and amount of the minerals present in the rock. Four recognized storage mechanisms, such as structural and stratigraphic trapping, residual phase trapping, solubility trapping and mineral trapping (Bickle, 2009; Yu et al., 2012; Soong et al., 2014) go into effect at multiple space and time scales to trap carbon dioxide in the shallow crust. Geological storage site characterization, carbon dioxide leakage detection, and injection-induced hazard assessment and management (Bickle, 2009; White 2009; Armitage et al., 2013; Ding et al., 2018; Ayayi et al., 2019; Zhu et al., 2021) are crucial issues on commercial large-scale underground injection. Of particular concern is the influence of geochemical reactions (Xu et al., 2004; Kampman et al., 2012; Gislason et al., 2014) on the sealing behaviour of faults (Kampman et al., 2016), the impact of stress regime and seismicity on fault reactivation (Talwani and Acree, 1984; Miller et al., 2004; Rutqvist, 2012; Verdon, 2014), and the geomechanical effect...
(Vilarrasa et al., 2010; Rinaldi et al., 2014) on carbon dioxide leakages through fault zones. Carbon dioxide leakages through subsurface geological reservoirs have been attributed to increased permeability (Kampman et al., 2012) resulting from dehydration and dissolution of fracture-filling phyllosilicate minerals in faults and fracture networks (Armitage et al., 2013; Frank et al., 2015), fracturing of solid rocks (Zoback et al., 1997), opening of bedrock fractures (Rojstaczer, 1995), aquifer expansion deformation (Atwater, 1992; Rutqvist, 2012) and pore-pressure diffusion (Muirwood and King, 1993; Yang et al., 2018) after strain occurs in the upper crust. Further to that, interactions between changes in local or regional stress and hydrological processes in critically stressed faults can facilitate the upward migration of carbon dioxide (Uysal et al., 2019; Kampman et al., 2012). Geological repositories far from earthquake-prone fault regions will be preferred on priority (Kampman et al., 2012). As is well known, faults within stable continental regions are always close to failure (Caine et al., 1996; Kampman et al., 2012) and may exhibit periods of seismicity. Understanding the role of faults, as either barriers or flow paths to the leakage of carbon dioxide, is significant for assessing the long-term integrity of geological storage reservoirs (Kampman et al., 2012).

Naturally occurring carbon dioxide reservoirs have stored carbon-dioxide-rich gas in underground formations over geological timeframes (Kampman et al., 2016), and provide a unique chance to examine the geological factors dominating carbon dioxide leakages (Kampman et al., 2012). Seismically active fault zones are frequently consistent with sites of anomalous crustal carbon dioxide flux (Irwin and Barnes, 1980; Kerrick, 2002; Tamburello et al., 2018). Increased carbon dioxide fluxes and increased fault activity have been found to be causally correlated (Hunt et al., 2017) by lots of scientific works. The fault activity is crustal deformation-associated (Liu et al., 2009), and can act to enhance fault-related carbon dioxide-degassing (Uysal et al., 2019; Yang et al., 2019).

Here, we examine fault leakage of carbon dioxide and crustal deformation of faults, which lie within the Bayanhot seismically active zone located at the Alxa plateau (Fig. 1). The faults are considered to be northeast-southwest dipping extensional. The carbon dioxide source at the Bayanhot fault zone is postulated to be crustal in origin. The relationship is investigated between variable carbon dioxide-leakage levels and local crustal tilt changes from Jan. 1, 2019 to Aug. 31, 2019. Measurements of carbon dioxide-leakage response to crustal deformation of faults have quantified changes in carbon dioxide-leakage levels. As a result of these observations, we suggest that carbon dioxide-leakage levels increase owing to fracture opening, potentially caused by changes in fault architecture and permeability structure of regions surrounding the faults.

2 METHODS OF OBSERVATIONS

Alxa Seismological Bureau established the crustal tilt and carbon dioxide observation station in the Bayanhot fault zone, as part of an integrated geodetic network. Fig. 1 shows the locations of the crustal tilt observation point and the 220 meter-deep borehole for carbon dioxide measurement. The solid-line depicts the faults, and the contours show the elevation of the Bayanhot fault zone. A quartz horizontal pendulum tiltmeter is installed for the crustal tilt observation in cavern. A Red-Infrared data acquisition instrument is adopted for recording carbon dioxide-leakage levels from the borehole and for monitoring atmospheric temperatures. All recorded data have been corrected for sensor recalibration, and erroneous data due to telemetry malfunctions, sensor failure, and other reasons have been removed. The data acquisition period is one minute, and observations exhibit characteristics of good-quality borehole gas level and crustal tilt data with high reproducibility and continuity. Detailed descriptions of the Red-Infrared data acquisition instrument (Yang et al., 2019; Xiang et al., 2019) and the crustal tilt observation in cavern (Yamamoto et al., 2004; Chen et al., 2015) can be found in references. Eventually, we have examined measurements of data from Jan. 1, 2019 to Aug. 31, 2019, during which period both crustal tilt and carbon dioxide levels are simultaneously recorded.

3 RESULTS AND DISCUSSION

3.1 Patterns of Carbon Dioxide Leakage

Here, we examine a record of carbon dioxide leakages from the fault zone. The preliminary
relationship between variable carbon dioxide-leakage levels and atmospheric temperatures is examined (Fig. 2). The carbon dioxide source is considered to be crustal in origin, and deeper aquifer formations are carbon dioxide charged at present. Systematic patterns of carbon dioxide levels show short-term fluctuations on a long-term increasing trend. Carbon dioxide levels and atmospheric temperatures vary systematically through time and are highly correlated. These phenomena may be indicative of the degassing of the carbon-dioxide charged fluids (Xu et al., 2004; Kampman et al., 2016; Uysal et al., 2019) as they migrate upwards and eventually reach shallower depths at lower pressure influenced by atmospheric pressure changes as a result of changes of atmospheric temperature. The geochemical reaction of carbon-dioxide degassing and carbonate precipitation (Kampman et al., 2012; Armitage et al., 2013; Frank et al., 2015; Jean et al., 2016) can control carbon dioxide-leakage levels from the fault zones (Ayayi et al., 2019). The temporal trend in carbon dioxide levels comprises a repeating tide-like pattern, and potentially indicates pulsing of carbon dioxide into the shallow groundwater system from deeper crustal formations. Replenishment of the shallow carbon dioxide reservoir (Kampman et al., 2012) apparently induces a sharp increase in carbon dioxide-leakage levels from the fault zone.
Fig. 2. (a) Carbon dioxide leakage levels, (b) atmospheric temperature records and (c) a histogram representing the relative frequency of the amount of observation data for Jan. 1, 2019 to Aug. 31, 2019. The temporal profiles are characterized by a short-term fluctuation along a long-term increasing trend. The data acquisition period is one minute. The top blue area and in right hand side, the orange area denote the frequency distribution of the carbon dioxide leakages and the temperatures, respectively.

3.2 Characteristics of Crustal Deformation

Here, we examine a record of crustal tilt data for characterization of crustal deformation in the Bayanhot fault zone. The crustal tilt data is taken at a one-minute sample rate. Crustal tilt records in the north-south and east-west directions from Jan.1, 2019 to Aug. 31, 2019 are shown in Fig. 3.
Fig. 3. Crustal tilt records in the (a) north–south and (b) east–west directions from Jan. 1, 2019 to Aug. 31, 2019. It indicates the relative frequency of the crustal tilt data. The tilt data is taken at a one minute sample rate. Here tidal components are clearly observed from the original tilt records. Anomalous tilt change in the north–south direction commenced in the middle of May, subsequently changing its trend in the beginning of July. The tilt in the east–west direction exhibited a remarkable change in the beginning of July.

The upward tilt represents the crustal tilt in the north–south direction, while the downward tilt indicates the crustal tilt in the east–west direction. Here tidal components (Chen et al., 2015; Yamamoto et al., 2004) are clearly observed from the original tilt records. Crustal tilt data exhibit characteristics of good-quality cavern tilt data with response to seismic waves and clear solid-earth tidal signals (Fig. 5). Systematic characteristics of crustal tilt data show short-term coherent tidal fluctuations on a long-term increasing trend. Anomalous tilt changes in the north–south direction commenced in the middle of May, subsequently changing its trend at the beginning of July. The tilt in the east–west direction exhibited a remarkable change at the beginning of July. We see anomalous tilt changes in association with distant earthquakes, indicating that the earthquake-induced crustal deformation and ground shaking can alter crustal tilt. Seismicity and stress regime can have significant impacts on fault stability (Rutqvist, 2012). Crustal tilt responses to earthquakes experience negative tilt changes with some subsequent recovery. Changes in local or regional stress in critically stressed faults can stimulate fault deformation (Verdon, 2014). The temporal trend of the crustal tilt can be connected with a subsurface source and, given the tectonic setting, the main mechanism is the fault slip and creep with increasing time (Liu et al., 2009).

3.3 Effects of Crustal Deformation on Carbon Dioxide Leakages

In this section, we examine the relationship between variable carbon dioxide-leakage levels and local crustal tilt changes. From a survey of the crustal tilt data, we divide the tilt record into three stages as follows (Fig. 3). In the first phase, from the beginning of January, the tilt direction was east-northeast upward and the tilting rate increased. In the second phase, from the middle of May, the tilt became northeast upward and the tilting rate sharply decreased as a result of a distant earthquake, and this trend lasted until the end of June. In the final phase, from the middle
of July, the tilting rate again increased owing to a distant earthquake while preserving the east-northeast upward direction. Meanwhile, we identify three phases of carbon dioxide-leakage levels during the same period (Fig. 2). We show that carbon dioxide-leakage levels and crustal tilt changes vary systematically through time and are highly correlated. The arrival of each tilt pulse triggers a sharp increase in carbon-dioxide-leakage levels as shown in Figs. 4 and 5, indicating increased degrees of carbon dioxide degassing from the fault zone (Kampman et al., 2012).

Fig. 4. An expanded section of (a, b) tilt data and (c) carbon dioxide levels for 744 hours in May. The temporal variations of carbon dioxide emissions generally coincide with the occurrences of the crustal tilt change, which described the fault deformation and the solid-earth tide.

From a survey of a record of carbon dioxide leakage from a faulted, natural carbon-dioxide-rich formation, and of the crustal tilt in the fault zone, we show that temporal changes in the crustal tilt reveal pulses of carbon dioxide leakage levels, and especially that each high-frequency pulse coincides with the onset of local solid-earth tide. We show a significant correlation between the crustal tilt magnitude and amount of carbon dioxide leakage in a long-term timescale. Carbon dioxide leakages through the fault zone vary with a periodicity controlled by the crustal deformation.

Previous research (Yang et al., 2019) found deep-buried limestone and sandstone formations with fault-parallel well-developed fractures. There is evidence for the dilation and propagation...
Fig. 5. An expanded section of (a, b) tilt data and (c) carbon dioxide leakage levels for Feb. 10 to Mar. 10. Arrow represents the occurrence of earthquakes.

of these fractures during critically stressed fault deformation (Zoback et al., 1997; Liu et al., 2009; Yang et al., 2019). Compaction and dilation of fracture well-developed bedrocks are prone to local or regional stress changes (Vilarrasa et al., 2010; Rinaldi et al., 2014), resulting in high strain rates (Yang et al., 2019; Zhang et al., 2020). Crustal deformation can lead to changes in the hydraulic conductivity of the fault because of fracture opening (Rojstaczer, 1995; Zoback et al., 1997).

The present-day state of maximum principal horizontal stress across the eastern margin of the Alxa plateau is characterized by a near northwest-southeast compression (Zoback, 1992), and there exist near northeast-southwest trending thrust faults, which are critically stressed. Changes in regional or local stress may deteriorate the stability of critically stressed faults (Zoback et al., 1997; Verdon, 2014), triggering changes in fault hydraulic conductivity and pore pressure (Vilarrasa et al., 2010; Rinaldi et al., 2014; Kampman et al., 2012; Yang et al., 2018), especially at depth, leading to migration of carbon dioxide-rich fluids from deeper reservoirs to the near surface (Miller et al., 2004; Kampman et al., 2016; Uysal et al., 2019; Yang et al., 2019). As the shallow reservoir is charged with low-density carbon dioxide, the pore pressure at the fault-reservoir interface would
increase (Kampman et al., 2012; Gasda et al., 2009; Lei et al., 2017), given that the hydrostatic gradient is considerably more than the pore-pressure gradient in the gas (Kampman et al., 2012).

Due to an ongoing increase in the height of the gas column, the pore pressure would finally overcome the minimum confining stress and tensile strength of bedrocks (Wiprut and Zoback, 2000; Kampman et al., 2012; Soong et al., 2014), inducing hydraulic fracturing, and subsequently generating conduits for carbon dioxide leakage to the surface (Rutqvist, 2012; Kampman et al., 2012; Lei et al., 2017). Moreover, unloading of solid-earth tide and lower atmospheric pressure on the land surface both decrease the normal force across the fault and increase the shear stress on a reverse fault (Liu et al., 2009), resulting in changes in the hydraulic conductivity of the fault damage zone (Min et al., 2004). At depth, deformation due to a buried shear dislocation (Verdon, 2014; Frank et al., 2015; Yang et al., 2019) may contribute to changes of hydraulic behaviour of the fault zone, potentially contributing to flow paths for carbon dioxide escape to the surface. Globally, amounts of large-scale fault zones are regions of high crustal carbon dioxide flux (Irwin and Barnes, 1980; Kerrick, 2002; Tamburello et al., 2018). The fault activity is crustal deformation-associated (Violay et al., 2015; Uysal et al., 2019), and may act to enhance fault-related carbon dioxide-degassing. Carbon dioxide leakage levels fluctuate as a result of partial blockage or reopen of the bedrock fractures (Rojstaczer et al., 1995; Min et al., 2004) in the faults. Whether faults act as barriers or flow paths to leakage of solid-Earth carbon dioxide will be reliant on in situ stress states and hydrological properties and processes (Kampman et al., 2012; Frank et al., 2015) at scales ranging from pores to faults (Montgomery and Manga, 2003). Responses of carbon-dioxide leakage to crustal deformation in fault damage zones would offer the potential for new insights into predicting the long-term integrity of carbon dioxide geological storage sites (Kampman et al., 2012).

4 CONCLUSIONS

We examine the relationship between variable carbon dioxide-leakage levels and local crustal tilt changes. Temporal patterns of carbon dioxide levels show short-term pulses on a long-term increasing trend, reflecting replenishment of shallow carbon dioxide reservoirs. The carbon dioxide levels vary from 537.7 up to 1317.1 ppm, and the mean level is 890.2 ppm. Temporal profiles of crustal tilt data show short-term coherent solid-earth tidal fluctuations on a long-term increasing trend. Anomalous tilt changes are associated with distant earthquake-induced crustal deformation and ground shaking. Carbon dioxide-leakage levels and crustal tilt changes vary systematically through time and are highly correlated, indicating increased degrees of carbon dioxide degassing from the fault zone. Carbon dioxide leakages vary with a periodicity controlled by the crustal deformation.

Stability degradation of critically stressed faults can alter fault hydraulic conductivity and pore pressure, especially at depth, triggering upward migration of carbon dioxide from deeper reservoirs. Whether faults act as barriers or conduits to leakage of solid-Earth carbon dioxide will be dependent of in situ stress states and hydrological properties and processes at scales ranging from pores to faults. Responses of carbon-dioxide leakage to crustal deformation in fault damage zones would offer the potential for new insights into assessing the long-term integrity of carbon dioxide geological storage sites.

DECLARATION OF COMPETING INTEREST

The authors declare no conflict of interest including financial interests and personal relationships.

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