

# A Case Study of Monitoring Emission from CO<sub>2</sub> Enhanced Oil Recovery by Remote Sensing Data

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## Abstract

Enhanced oil recovery with carbon dioxide (CO<sub>2</sub>-EOR) is considered to be a cost effective way for carbon capture and storage. However, due to the complexity of geological structure in underground reservoirs, long-term leakage is possible. A case study of CO<sub>2</sub>-EOR has been conducted at Citronelle, Alabama in the United State of America. A total of 8,036-ton of CO<sub>2</sub> were injected from November 2009 to September 2010 and some leakages via production were identified by isotopic analysis in May 2010. In this study, remote sensing data of CO<sub>2</sub> and methane (CH<sub>4</sub>) concentrations, and aerosol optical depth (AOD) at a large scale were used to monitor emissions to atmosphere at the study site. Based on the observed monthly CO<sub>2</sub> and CH<sub>4</sub> concentrations in the atmosphere at the study site and surrounding areas, some abnormal values related to possible emission were identified at different time scales by correlation, variance and entropy analysis. The annual average of ratios between CO<sub>2</sub> concentration and CH<sub>4</sub> concentration, which might be due to CO<sub>2</sub> emission, reached the highest value in 2009. In comparison with surrounding areas, the monthly values of AOD at the study site were relatively higher, especially during the time periods of 2008, 2009 and part of 2010. Our results might confirm the isotopic analysis at the ground and may provide more detailed information. Therefore, through this approach remote sensing data could be used to monitor and evaluate emissions from areas involved in CO<sub>2</sub>-EOR at a large scale and provide helpful information for ecological assessment of CO<sub>2</sub>-EOR.

**Keywords:** carbon storage, CO<sub>2</sub> enhanced oil recovery, Citronelle in Alabama, emission, remote sensing

## 1. Introduction

Increasing atmospheric CO<sub>2</sub> and other greenhouse gases is considered to be a critical driving force of global climate change. Dramatic reduction in CO<sub>2</sub> emission is needed to significantly stabilize and decrease atmospheric greenhouse gas concentration while fossil fuels are continuously used. Carbon capture and geological storage, a means to bury CO<sub>2</sub> in geological reservoirs for a long time, is thus important to reduce the current CO<sub>2</sub> emission (Pacala & Socolow, 2004; Schrag, 2007). Carbon capture and storage are well-understood in oil and gas industries (Mertz, Davidson, de Coninck, Loos, & Meyer, 2005; Thomas & Benson, 2005). Based on the possible benefits for carbon storage, enhanced oil recovery with CO<sub>2</sub> (CO<sub>2</sub>-EOR) is considered cost effective. CO<sub>2</sub>-EOR has the potential to recover 30-60% more of the original oil in reservoirs based on Department of Energy Oil Recovery Program, and even without CO<sub>2</sub> credits or taxes, there are revenues from oil or gas production (Zweigel, Arts, Lothe, & Lindeberg, 2004; Thomas & Benson, 2005). CO<sub>2</sub>-EOR practice has been applied for decades by the oil and gas industry in the USA and Canada (van Bergen, Gale, Damen, & Wildenborg, 2004; Thomas & Benson, 2005), such as those projects at Weyburn and Cranfield (Whittakers, White, Law, & Chalaturnyk, 2004; Meckel & Hovorka, 2009).

Although carbon capture and storage is very attractive in potential, this practice is still controversial. The main concern is its potential long-term leakage, which may be a result of fluid migration pathways from carbonate reservoirs in a tectonically complex region or new emerging pathways from reservoirs (e.g., Gilfillan et al., 2009; Jenkins et al., 2012). Roberts, Wood, and Haszeldine (2011) assessed the health risk of natural CO<sub>2</sub> seepage in Italy and indicated that human death is strongly influenced by seepage surface expression, local environmental condition, CO<sub>2</sub> flux and human behavior. Fear of surface leakage and a lack of local benefit are the main factors for negative public opinion of CO<sub>2</sub>-EOR (Roberts et al., 2011).

The likelihood of surface leakage will depend on site specific geological structure and characteristics (Gilfillan et al., 2009; Roberts et al., 2011; Jenkins et al., 2012). Therefore, developing and implementing leakage monitoring and assessing procedures will help to evaluate the accuracy of current concerns (Roberts & Chen, 2012). One possible approach is the use of remote sensing data. The Atmospheric Infrared Sounder (AIRS: Pagano, Aumann, Hagan, & Overoye, 2003) and the Moderate Resolution Image Spectroradiometer (MODIS) data products might be used to monitor CO<sub>2</sub>, CH<sub>4</sub> and aerosol optical depth (AOD) at a large area although there are limited applications for the study of industrial emission. With the emission of CO<sub>2</sub>, then, the regional ratio of CO<sub>2</sub> and CH<sub>4</sub> should be changed. Also, aerosol concentration may increase with gas emission. As a case study, a CO<sub>2</sub>-EOR experimental project has been conducted from 2007 to present at Citronelle, Alabama, USA. In May of 2010 CO<sub>2</sub> seepage relating to CO<sub>2</sub> injection activities was observed above ground and verified by carbon isotopic analysis. This pilot CO<sub>2</sub>-EOR project in Citronelle, AL provides a case where known seepage occurred and thus provides an opportunity to assess the atmospheric changes relating to CO<sub>2</sub>-EOR activities using remote sensing data. The objectives of this study are to (i) provide general information of monthly atmospheric CO<sub>2</sub>, CH<sub>4</sub> and aerosol for evaluation; (ii) identify whether there are sudden increases of atmospheric CO<sub>2</sub>, CH<sub>4</sub> and aerosol; and (ii) compare monthly atmospheric concentrations of CO<sub>2</sub>, CH<sub>4</sub> and aerosol to characterize possible seepage.

## 2. Material and Methods

### 2.1 Study Site

Since 2007 a pilot CO<sub>2</sub>-EOR project has been conducted at the Citronelle field (near 31°05'N, 88° 14'W) of the Mobile County, Alabama in USA. The Citronelle oil field, located on the crest of the dome, has produced millions of barrels of oil from sandstone in the Lower Cretaceous Rodessa Formation (Esposito et al., 2008). The local landscape is mainly composed of forest, agricultural land and town (urban) area. About 31– 34% of the original oil in this area has been collected by primary and secondary methods and CO<sub>2</sub>-EOR was estimated to increase reserves by up to 20% (Esposito, Pashin, & Walsh, 2008). A total of 8,036-ton CO<sub>2</sub> injection was completed during the time period from November 2009 to September 2010. In May 2010 seepages were confirmed at the production well-fed tank battery and verified by carbon isotopic analysis. Higher CO<sub>2</sub> concentrations were also monitored several times at the study site and surrounding areas by routine monitoring work on ground. More information about the project may be found at the National Energy and Technology Laboratory website ([www.netl.doe.gov](http://www.netl.doe.gov)).

### 2.2 Remote Sensing Data

The high-resolution Atmospheric Infrared Sounder (AIRS) was launched into Earth-orbit in May 2002, with the goal to support climate research and improve weather forecasting. AIRS is one of the six instruments on board the Aqua satellite, which is a part of the National Aeronautics and Space Administration (NASA) Earth Observing System. AIRS uses cutting-edge infrared technology and provides information related to air temperature, water vapor, trace gases and cloud property (e.g., Pagano et al., 2003; Chahine, Barnet, Olsen, Chen, & Maddy, 2006). AIRS CO<sub>2</sub> retrievals use an analytical method for the determination of carbon dioxide and other minor gases in the troposphere from AIRS spectra. The AIRS data have been shown to be accurate to within 1.20 ppm of simultaneous measurements by aircraft (Chahine et al., 2005). Moderate Resolution Imaging Spectroradiometer (MODIS) instruments onboard the Terra platform first and then onboard the Aqua platform are uniquely designed to monitor earth change. Terra MODIS and Aqua MODIS are viewing the entire Earth's surface every 1 to 2 days, acquiring data in 36 spectral bands, or groups of wavelengths (e.g., Salomonson, Barnes, Maymon, Montgomery, & Ostrow, 1989; Remer et al., 2005). Aerosol optical depth is negative natural logarithm of the fraction of radiation (e.g., light) that is not scattered or absorbed by aerosol and it is dimensionless. Wang & Christopher (2003) found a linear relationship between MODIS aerosol optical depth and PM<sub>2.5</sub> concentrations measured by tapered-element oscillating microbalances in Jefferson County, Alabama. Based on the knowledge in EOR practices, CO<sub>2</sub> emission may affect CH<sub>4</sub> concentration and AOD. Monthly data of CO<sub>2</sub>, CH<sub>4</sub> from AIRS and AOD of 550 nm from MODIS at 1.0° × 1.0° near the study site (31°-32°N, 88°-89°W) and surrounding areas from January 2003 to September 2011 were used in this study. The extent of the surrounding areas includes the eastern area (31°-32°N, 87°-88°W), western area (31°-32°N, 89°-90°W), southern area (30°-31°N, 88°-89°W) and northern area (32°-33°N, 88°-89°W). Giovanni online data system developed and maintained by the NASA GES DISC were used (Acker & Leptoukh, 2007). The unit for CO<sub>2</sub> and CH<sub>4</sub> is ppm and there is no unit for AOD. More detailed information can be found at <http://airs.nasa.gov> and <http://modis.gsfc.nasa.gov>.

### 2.3 Data Analysis and Statistics

ANOVA was used to analyze monthly concentrations of atmospheric CO<sub>2</sub>, CH<sub>4</sub> and values of AOD. Significant level is at  $p < 0.05$ . The variances of CO<sub>2</sub>, CH<sub>4</sub> and AOD monthly values were calculated using the following:

$$V_i = \sum_{i=1}^n (x_i - \bar{x})^2 \quad (1)$$

$V_i$  is the variance at time scale  $j$ ,  $x_i$  is the monthly concentration for atmospheric CO<sub>2</sub>, CH<sub>4</sub> and AOD from remote sensing,  $\bar{x}$  is the average value.

$$STD = \sqrt{V_i / n} \quad (2)$$

Where  $STD$  is the annual standard deviation.

$$T_j = \sum_{j=1}^m V_i \quad (3)$$

Where  $T_j$  is the sum of the variances at the scale  $j$ .

$$P_j = \frac{V_j}{T_j} \% \quad (4)$$

Where  $P_j$  is the percentage of variance in the total variance of this time scale.

$$S_j = P_j \log(P_j) \quad (5)$$

Where  $S_j$  is the entropy (similar as Shannon-Wiener index, no unit) at the time scale. The higher the values in entropy, the less variance there is or the more even for the concentration data. The high fluctuation of concentration data or local value of entropy might be related to seepages. In this study, the time scales for entropy calculation include 1, 3, 5, 7, 15 and 21 months. Similar method was used for studying plant spatial distribution (Chen, Li, & Scott, 2005).

## 3. Results and Discussion

### 3.1 Dynamics of CO<sub>2</sub> Concentration

The monthly atmospheric CO<sub>2</sub> concentration increased from 376 ppm to 394 ppm from 2003 to 2011 (Figure 1). The annual CO<sub>2</sub> concentration at the study site increased each year. There exists annual fluctuation of atmospheric CO<sub>2</sub> concentration at the study site. Usually the CO<sub>2</sub> concentrations reach the highest values near the start of summer (such as May or June) and then decline to the lowest values until fall time due to vegetation growth. If the monthly change of atmospheric CO<sub>2</sub> concentration should be smooth, then, the CO<sub>2</sub> concentrations during the following time periods in Figure 1 might be considered as abnormal: March – May 2005, January – April 2006, March – May 2008, January – March 2009, April – July 2010 and September – November 2010. The variances between the monthly CO<sub>2</sub> concentration at 2003-2006 and 2007-2010 were not significant ( $p = 0.5218 > 0.05$ ). If the monthly CO<sub>2</sub> concentrations are substituted by the CO<sub>2</sub> concentration in January at each year and then these data are analyzed by ANOVA, the monthly CO<sub>2</sub> concentrations in 2010 and 2004 have more significant changes (Table 1). This also indicates that time scale is important for monitoring CO<sub>2</sub> emission.

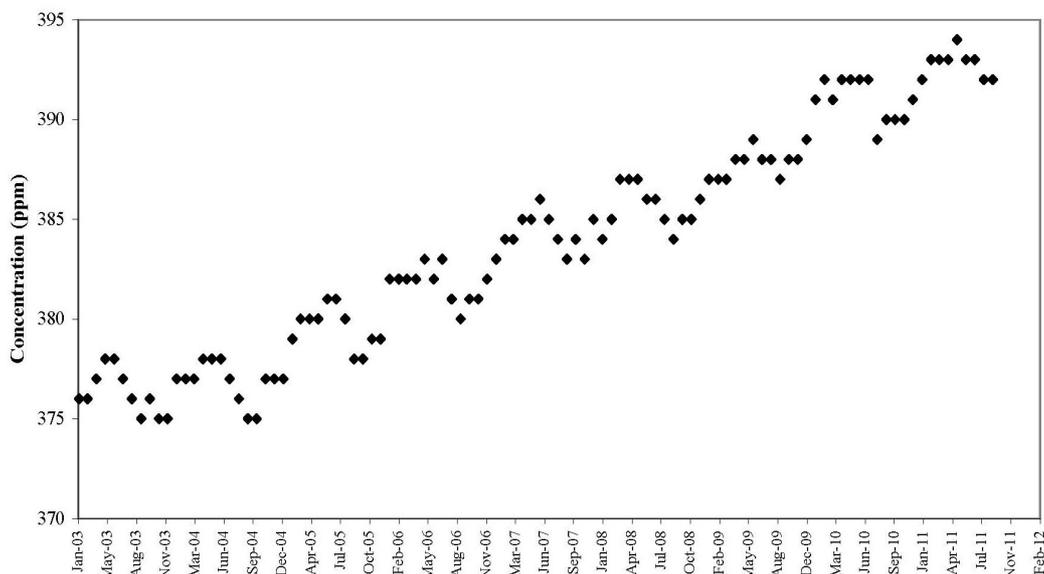


Figure 1. Dynamics of monthly CO<sub>2</sub> concentration in atmosphere at the study site

Table 1. The *p* values of ANOVA for monthly CO<sub>2</sub> concentration from 2003 to 2011 after the January CO<sub>2</sub> concentration in each year is substituted. *p* < 0.05 is statistically significant here

	Y04	Y05	Y06	Y07	Y08	Y09	Y10	Y11
Y03 (1.167 ±0.322)	0.006*	0.024*	0.002*	0.847	0.363	0.393	0.015*	0.333
Y04 (-0.167 ±0.297)		0.000*	0.832	0.002*	0.001*	0.012*	0.698	0.020*
Y05 (2.331 ±0.355)			0.000*	0.026*	0.128	0.002*	0.000*	0.002*
Y06 (-0.250 ±0.250)				0.001*	0.000*	0.003*	0.530	0.006*
Y07 (1.250 ±0.279)					0.435	0.244	0.006*	0.201
Y08 (1.583 ±0.313)						0.060	0.001*	0.049*
Y09 (0.833 ±0.207)							0.034*	0.857
Y10 (0.000 ±0.302)								0.052
Y11 (0.778 ±0.222)								

Note. Y03 represents year 2003 and others follow the same way. The numbers in ( ) of the first column at the left side are averages and standard errors of the above year.

The standard deviations of annual atmospheric CO<sub>2</sub> concentration declined from 2003 to September 2010, but there were fluctuations (Figure 2). There is a high correlation between the monthly atmospheric CO<sub>2</sub> concentrations in the same months among different years (Figure 3) ( $R^2 = 0.9314$ ,  $p < 0.05$ ). The following points were deviated from the regression line visually: (375, 375) September 2003 and 2004, (376, 375) October 2003 and 2004, (377, 382) January 2005 and 2006; (377, 381) July 2004 and 2005, (382, 386) June 2006 and

2007, (386, 386) June 2007 and 2008, (387, 387) March 2008 and 2009, (387, 392) February 2009 and 2010. The confidence interval is not given here since the  $X$  and  $Y$  values are not independent.

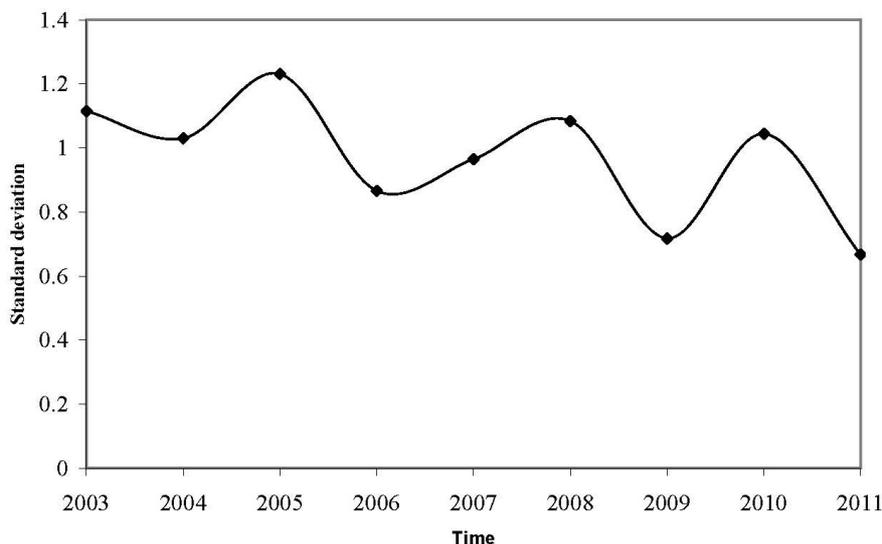


Figure 2. Change of annual standard deviations of monthly CO<sub>2</sub> concentration at the study site

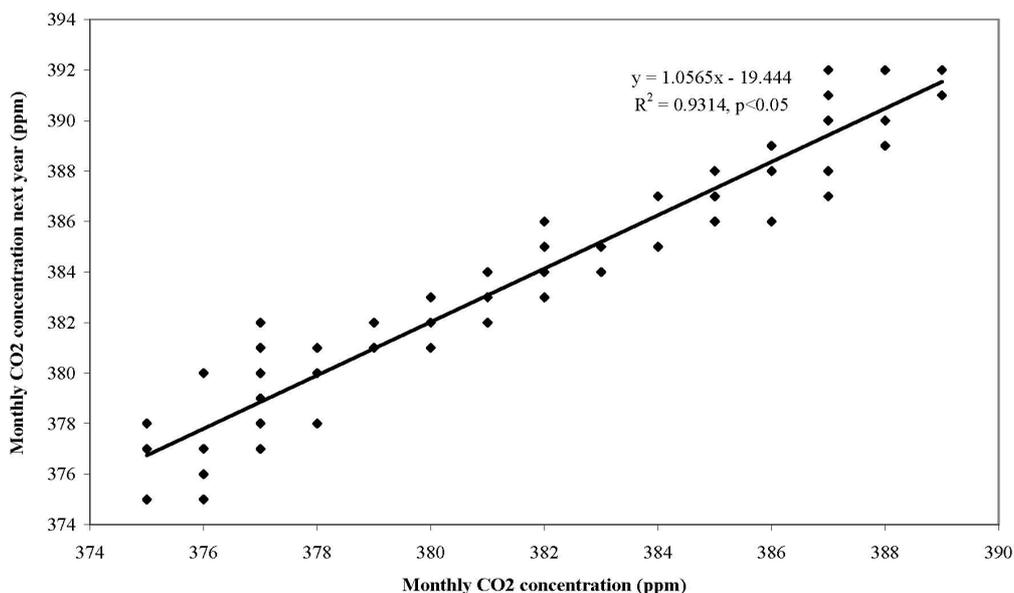


Figure 3. Correlation between monthly CO<sub>2</sub> concentrations at the same months but in different years

Sudden changes of atmospheric CO<sub>2</sub> concentration were obvious in the monthly data, but the linkage with CO<sub>2</sub> emission is not straightforward. The main problems are possibly related to (i) limited CO<sub>2</sub> emission into the atmosphere as CO<sub>2</sub> emission occurred on ground and the covered area is 1° by 1° in latitude and longitude and also the CO<sub>2</sub> concentration is measured from the mid-tropospheric layer; (ii) a total 8,036-ton of injected CO<sub>2</sub> might be too limited for CO<sub>2</sub>-EOR activities to cause obvious change in atmospheric monthly CO<sub>2</sub> concentration. Any slight change in atmospheric CO<sub>2</sub> concentration might be related to a big change on ground; (iii) Citronelle is oil field with hundreds of oil wells. Emission from some wells was not monitored. Unidentified or missing emissions are possible. Remote sensing might provide more records of abnormal CO<sub>2</sub> concentrations than the discrete ground monitoring; and (iv) the rise of global atmospheric CO<sub>2</sub> concentration may hide local noises of CO<sub>2</sub> emission. Global CO<sub>2</sub> concentration increases about 2 ppm each year based on the NOAA observation data from Mauna Loa at Hawaii. All these factors might possibly work together to some extent, the original

assumption about sudden increase of atmospheric CO<sub>2</sub> concentration related to emissions from CO<sub>2</sub>-EOR became indirectly. However, from the decreasing annual standard deviations from the monthly CO<sub>2</sub> concentration, abnormal points in correlation map of CO<sub>2</sub> concentration at some months but in different years, and also the comparison of CO<sub>2</sub> concentrations at different time scales with CO<sub>2</sub> concentrations at surrounding areas, it still could be inferred that possible CO<sub>2</sub> emission occurred at the study site if there were no other sources (such as biomass burning).

The entropy of variance of CO<sub>2</sub> concentration at different time scales was similar with its entropy values in the eastern, western and southern areas, but lower than the value in the northern area (Table 2). This means the variances of atmospheric CO<sub>2</sub> concentrations at the study area were higher than the CO<sub>2</sub> concentrations in its northern area across all time scales.

Table 2. Comparison in the entropy (no unit) of atmospheric CO<sub>2</sub> concentration at the study site and surrounding areas across different time scales

Time scale (month)	Study site	East	West	South	North
1	17.77	17.69	17.76	17.76	19.6
3	64.29	64.32	64.31	65.23	65.97
5	85.36	85.18	85.36	85.36	86.88
7	99.35	99.05	99.35	99.35	100.84
15	129.96	129.88	129.96	129.96	131.39
21	144.68	144.54	144.68	144.68	146.51

The CO<sub>2</sub>-EOR project, just like other geological carbon storage, was considered to operate under zero or very limited predictable leakage (Holloway, Pearce, Hards, Ohsumi, & Gale, 2007). Currently there is no standardized value for tolerable seepage. But the minimum retention for geological carbon storage is at least 99% stored CO<sub>2</sub> for 1,000 years. To ensure effective climate abatement, leakage rates of less than 0.1% y<sup>-1</sup> are needed (Haugan & Joos, 2004). Roberts et al. (2011) estimated from Italian seepage that 0.1 — 1% from storage of 3.6 Mt per year would be reasonable. The numerous models based on knowledge of fluid flows, usually turned out several orders lower in magnitude than the Italian gas seeps. In this study, it is still not clear how much atmospheric CO<sub>2</sub> concentration would increase if there is 1% emission of 8,036 tons injected CO<sub>2</sub>.

### 3.2 Dynamics of CH<sub>4</sub> Concentration

In comparison to monthly CO<sub>2</sub> concentrations at the study site, monthly CH<sub>4</sub> concentrations were relatively stable from 2003 to 2011 and these values were close to 1.0 ppm (Figure 4). The correlation between monthly CH<sub>4</sub> concentrations at the same time among different years was not significant ( $p > 0.05$ ).

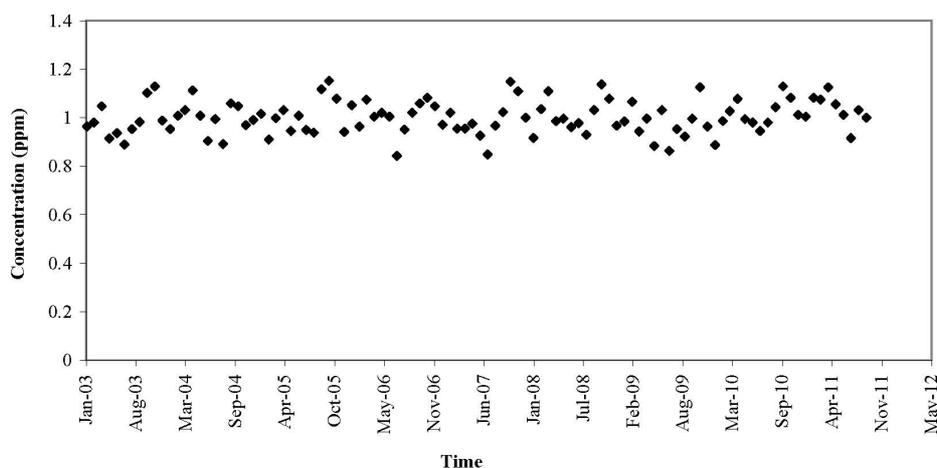


Figure 4. Monthly CH<sub>4</sub> concentration in atmosphere at study site

The entropy of CH<sub>4</sub> concentrations at the study area was lower than the values from the surrounding areas (Table 3), especially at the southern area, although the values were larger at some time scales. The annual average of ratios between CO<sub>2</sub> concentration and CH<sub>4</sub> concentration was relatively stable from 2003 to 2006, but it reached the highest value in 2009 (Figure 5).

Table 3. Comparison in the entropy (no unit) of atmospheric CH<sub>4</sub> concentration at the study site and surrounding areas across different time scales

Time scale (month)	Study site	East	West	South	North
1	28.26	31.71	27.86	28.41	28.38
3	55.51	58.01	55.17	57.71	55.36
5	73.01	74.07	72.21	75.33	74.32
7	85.17	85.45	84.11	86.79	86.57
15	116.14	117.31	116.51	116.67	116.80
21	130.80	130.78	130.31	130.89	131.46

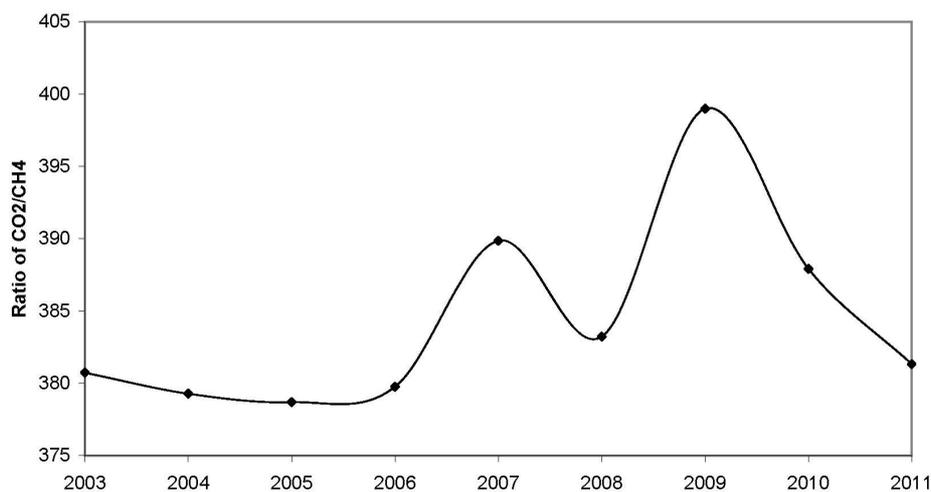


Figure 5. Annual average of ratios for atmospheric CO<sub>2</sub>/CH<sub>4</sub> at study site

Although there were lots of small fluctuations in the monthly CH<sub>4</sub> concentration at the study site, the atmospheric CH<sub>4</sub> concentration at study site was within the range of 0.8 — 1.2 ppm, which is considered as relatively stable. However, there were large increases in the annual averages of the ratio of CO<sub>2</sub>/CH<sub>4</sub> concentration in 2007 and 2009, which might be a helpful indicator for atmospheric CO<sub>2</sub> change, as this ratio might be a good indicator for some CO<sub>2</sub> sources.

### 3.3 AOD Change

The monthly AOD at the study site declined after 2007 (Figure 6). Usually AOD increases during spring and early summer time and reaches the highest value in July or August and then declines in the fall and winter time. The highest AOD in August 2009 was the lowest one of all these higher values. The annual average of monthly AOD is the lowest in 2009 for the study site and the surrounding areas (Figure 7). For the study site the annual average of monthly AOD was relatively lower than the AOD at the surrounding areas, but it was relatively higher after 2007.

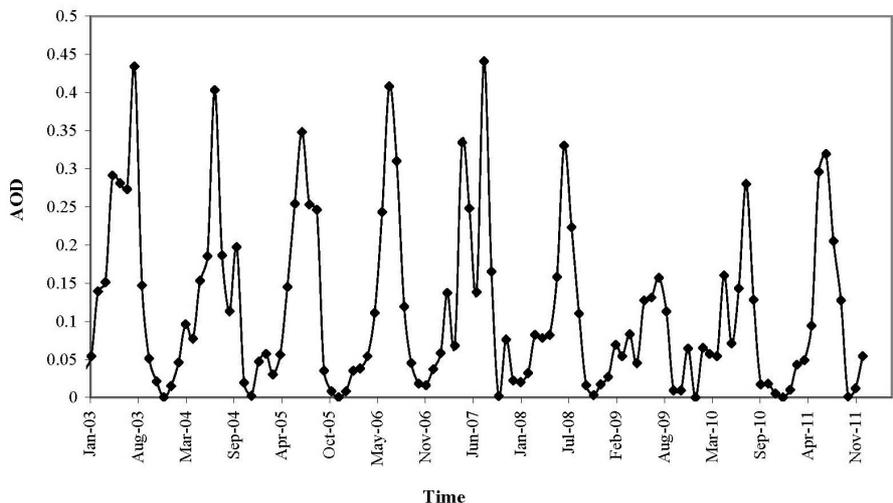


Figure 6. Monthly AOD (dimensionless) dynamics at study site

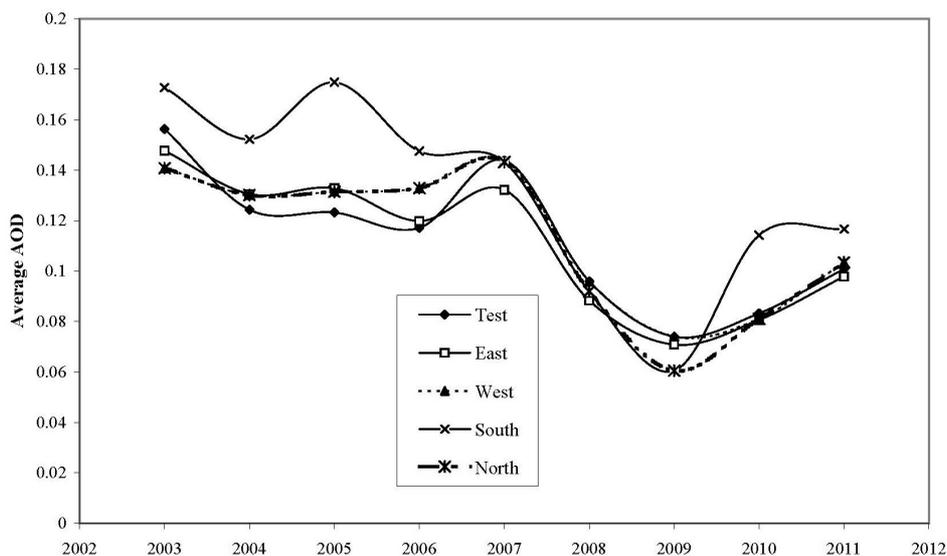


Figure 7. Comparison of annual averages of AOD (dimensionless) at the study site and surrounding areas

The annual averages of monthly fine AOD data at study site and surrounding areas decreased after 2008, which means there were less fine aerosols in the air. However, in comparison with surrounding areas, the AOD values at the study site were relative higher, especially during the time periods of 2008, 2009 and part of 2010. These relatively higher values of AOD might be related to emission from CO<sub>2</sub>-EOR.

**4. Conclusion**

There was limited environmental monitoring for previous CO<sub>2</sub>-EOR projects and this project is the first to implement some monitoring practices. However, due to the limitations of private land ownership at the study site, it is impossible to install monitoring instruments within the study site or have frequent visits. Thus, remote sensing is the best choice for environmental monitoring at the study site and surrounding areas at a large scale.

Through multiple-scale analysis of remote sensing data at the study site and surrounding areas, such as atmospheric CO<sub>2</sub> concentration, CH<sub>4</sub> concentration, the ratio of CO<sub>2</sub>/CH<sub>4</sub> and AOD changes, it could be inferred that there were CO<sub>2</sub> emissions from the study site although the magnitude might be low. CO<sub>2</sub>-EOR may not be operated with zero risk of CO<sub>2</sub> emission. However, it is necessary to conduct environmental monitoring and risk management procedures for CO<sub>2</sub>-EOR projects. This method can be developed for monitoring emission from CO<sub>2</sub>-EOR and other industrial operations at other locations.

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