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Geological CO_2 Monitoring Methods: A Review

Miss.Wagh Sushama M.*, Mr. Devi R. J.
Electronics Department, Karmaveer Bhaurao Patil Collage of Engg. & Poly.Satara, India

Abstract
This review paper discusses various methods of monitoring of atmospheric geological CO_2. The report presents a detailed description of the technologies that can measure CO2 leakage from potential point or diffuse sources. CO2 leakage at the surface may get quickly dispersed into the atmosphere, and therefore may prove difficult to detect using methods used for a wide areal coverage. Therefore, Surface monitoring equipment may preferably be positioned in the region of areas where possible leakage paths have been identified through the reservoir. Long Open Path (IP diode) lasers, Short open path (IR diode) lasers, Short closed path (NDIRs and IR) and Eddy covariance can do monitoring of atmospheric CO2. All these methods with their strengths and weaknesses are discussed here.

Keywords: Long Open Path, Short Open Path,Infra-Red, Non-Dispersive Infra-Red.

Introduction
Due to the nature of a CO2 geological storage site, techniques to detect any potential leakage or likely pathway will be necessary prior to deployment of direct or indirect instrumentation for quantification. So atmospheric methods for monitoring of geological CO_2 will therefore be important to identify any potential leakage. Leaked emissions of CO2 to atmosphere may be quickly dispersed, and then it becomes very difficult to detect using conventional techniques favouring low spatial resolution. Therefore, Surface monitoring is best for such type of leakage. So atmospheric methods are briefly summarised along with their merits and demerits.

Atmospheric monitoring methods
Long open path (IP diode lasers)
Various open-path sensing techniques have been developed that measure the path-integrated concentration of a target gas between two points near the ground surface. These methods have been used to locate gas emission points from various point or non-point sources (e.g. landfills, coal mines, wastewater treatment plants) and estimate their leakage rates to the atmosphere. Recently these methods have also been applied to the monitoring of CO2 geological storage sites.

At present there are a number of systems have been proposed for this type of monitoring (EPA 2006), including: Open-Path Fourier Transform Infrared (FTIR) Spectroscopy; Ultra-Violet Differential Optical Absorption Spectroscopy (UV-DOAS); Open-Path Tuneable Diode Laser (TDL) Absorption Spectroscopy; Differential Absorption LIDAR (DIAL). Of these technologies, the FTIR and UV-DOAS technologies have the advantage that they can simultaneously detect a large number of different gas species. To date, the TDL and the FTIR have been applied more in the field of CO2 monitoring. Only the workings of the TDL will be discussed here due to its more common usage. Open-Path Tuneable Diode Lasers, which consist of an integrated transmitter/receiver unit and a reflector, measure the distance-averaged concentration of a specific gas in the air. A signal emitted from the transmitter propagates through the air to the reflector (positioned metres to hundreds of metres away) and returns to the instrument where it is focused onto a detector. Because of absorption of the signal by the gas of interest, the decrease in the signal returned to the detector is directly proportional to the total amount of that gas over the entire path length.

Strengths and Weaknesses
As can be seen by the review above, the long open path technique is used due to a number of advantages that it has with respect to monitoring and leakage quantification. First, the large atmospheric volume that it can cross gives this method the potential to intersect a temporally and spatially variable plume. Second, because the lasers can be

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mounted on automated rotating platforms controlled by aligning software, and most units have an internal reference cell for self-calibration, the unit is well adapted for long-term, unattended monitoring of a site. Combined, these two advantages mean that it may be possible to monitor a large area with a single laser unit and a number of fixed reflectors (i.e. optical pathways).

Despite these advantages there are certain limitations that will affect the eventual deployment of this technique. To begin with, although it may be physically possible to measure across great distances with the laser, the fact that the method yields a distance-averaged concentration means that with increasing path length there is a corresponding reduction in resolution and sensitivity due to dilution of the plume signature by background atmospheric air. The distance between the leakage point and the optical path is also critical, as long distances will decrease plume concentrations and reduce the potential wind directions that would result in plume intersection.

Short open path (IR diode lasers)

Along with long open path instruments, various analytical instruments have been applied to the class defined as short open path sensors, including tuneable diode lasers (TDL) and Fourier Transform Infrared (FTIR) detectors.

The basic principle of the short open path technique is that transmitter and receiver are separated by a fixed distance (typically the order of 1-2 m), between which there is a free exchange of atmospheric air. Compared to long open path instruments, which provide distance-averaged concentrations, short open path equipment is mounted on ground or airborne vehicles for mapping of point concentrations. Because the sensor measures concentrations directly in the air response times are very rapid with very little memory effects, and thus surveys can be conducted at high speeds. Again, the TDL is the most commonly deployed method for CO2 monitoring applications, and thus described in more detail below.

The system consists of two main units. A control box (housing a laser diode, drive electronics, and microcomputer controller) is linked via a fibre optic cable to the external probe (consisting of reflectors and a photo-detector). As with the open path TDLs, laser light of a given wavelength is beamed across a distance and the amount of light absorbed by the gas of interest is proportional to its concentration. In this case the laser makes four passes across the fixed-length probe prior to quantification by the photo-detector. As stated, the laser pathway is open to the atmosphere and thus measured values are essentially instantaneous.

Strengths and Weaknesses

Advantages of the airborne and ground-based short open path TDL systems include temperature changes, ground, and cloud conditions do not affect the performance of the TDL. Detectors are very sensitive for CH4 (< 1 ppm) and moderately sensitive for CO2 (5-10 ppm). Internal calibration and lack of moving parts results in good instrument stability and low maintenance costs and the relatively small size and weight facilitates the mounting of this instrument on ground or airborne vehicles. Disadvantages of the system is that a series of point measurements in the atmosphere, changes in wind speed and direction will greatly affect absolute and relative measured values during the period of the survey. The influence of background variations in CO2 concentrations on leak detection capability has not been quantified. For quantification purposes, it appears difficult to use airborne instruments due to flight elevation restrictions and the dense nature of a CO2 leak. Although these techniques may develop to be highly useful monitoring tools for CO2 storage sites, the basic physical restrictions listed above probably mean that the method is not particularly well adapted for leakage quantification.

Short closed path (NDIRs and DIR)

Short closed path detectors involve the introduction of a gas sample into a closed chamber via a pump or by diffusion, and the quantification of a specific gas component by passing light across the chamber (i.e. short path) and through the sample. Thus these methods are similar to long open path and short open path due to the use of optical sources and detectors, but differ due to the use of the measurement chamber. This chamber allows for greater portability, reduces interferences like dust, and reduces costs. However, it can have a lower sensitivity due to the shorter path length and has a slower response time/greater sample memory due to mixing and dilution in the flow-through cell volume. Systems that use infrared light are an example of this type of detector.

In its simplest form, an infrared analyser consists of an infrared source and an infrared detector separated by a measurement cell. Recent advances have involved the addition of internal reference cells for internal calibration or drift correction. An air sample

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is passed through the measurement cell (or diffused into it), and the gas species of interest absorbs some of the radiation coming from the source. As the amount of absorption is proportional to concentration, according to the Lambert-Beer law, the decrease in signal arriving at the detector is converted into a concentration value. Each gas species absorbs light at a series of different wavelengths as a function of its interatomic bond types and strengths, resulting in a spectrum of absorption peaks that is unique for each gas (Chou 1999).

There are two types of infrared detectors, non-dispersive (NDIR) and dispersive (DIR). In a non-dispersive infrared detector all the light from the source passes through the sample, after which it is filtered to the desired wavelength just prior to impinging on the detector. In a dispersive system, a grating or prism is used prior to the sample to select a specific wavelength, and thus only this wavelength passes through the sample on its way to the detector. Dispersive IR detectors are typically used in laboratory instruments because they can scan a wide range of wavelengths, however they tend to be more costly and less suitable for portable instruments. Instead NDIR units are the most commonly used short closed path detectors for field applications, due to their robust nature, portability, low cost, stability and selectivity. In addition, a well-designed NDIR unit have a life expectancy of more than 10 years.

Strengths and Weaknesses

Advantages of the NDIR instruments are their low cost, flexibility in terms of concentration range and individual deployment configurations, and the limited interferences from other gases. In particular, the low cost per NDIR sensor means that a relatively large number could be deployed, while their small size and point measurement nature can allow for deployment also close to the ground (i.e. no line-of-sight or vegetation problems). Disadvantages are like most of the atmospheric methods influenced by wind dilution and dispersion, on sensitivity. To date all reported experiments have been conducted at very short distances from the actual emission point, ranging from 0 to a maximum of 30 m. In contrast the estimates conducted using the CH4 detector were much more accurate, although again errors were larger for the greater source to measurement point distances.

Eddy covariance

One of the most useful methods to measure and determine gas fluxes in the atmospheric boundary layer (surface layer or constant flux layer) is the Eddy Covariance Method (ECM). The ECM uses statistics to compute turbulent fluxes of heat, water and gas exchange (e.g. CO2, CH4 and trace gases). It has the ability to average the integral flux of gases over larger areas (fetch, m2 -km2 areas) and different temporal scales). Although the method is very complex in terms of hardware design and processing the large amounts of data, it has been proposed as a potential methodology for monitoring geologic carbon dioxide storage sites.

The ECM relies on the assumption that transport between the surface and the atmosphere is occurring by turbulent movement. These small confined turbulences are called eddies and the air flow can be imagined as a horizontal flow of numerous rotating three dimensional eddies with different sizes distributed over the measurement height.

In simple terms, gas flux can be described as the number of molecules crossing a unit area per unit time. By means of EC measurements the numbers of molecules (or any other entity like temperature or humidity) is determined and the gas flux is based on the covariance between concentration and vertical air movement/speed.

strengths and weaknesses

Though more expensive and technically more complex, the eddy covariance method provides a powerful tool that allows for spatial integration and near-continuous, long-term monitoring of the soil-atmosphere flux. This holds for CO2 but also for other greenhouse gases, but the method is restricted to onshore sites. Flux rates measured, i.e. the conventional application range, usually lie within the typical range of CO2 emissions from soils and different land covers (10th's of $g/m^2/d$). Higher emission rates can easily be determined. However, whether the ECM can detect a release of carbon dioxide from a storage site strictly depends on the ratio between the integral CO2 flux from the footprint area and the seepage rate from the point source.

Conclusion

In terms of atmospheric monitoring of CO_2, it can be concluded that NDIR sensors are more useful if deployed around areas of higher potential for leakage. Also can be noted that elevated concentrations were not observed at greater heights except directly above the leakage point when deploying NDIR. Short closed path tuneable diode

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lasers (TDL) can have better sensitivities and faster response times but tend to be more expensive.

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Author Biblography

	**Miss. Wagh Sushama Mohan** M.E. Electronics, Karmaveer Bhaurao Patil College of Engg. & Poly., Satara.
	Prof. Mr. Devi R. J. Electronics Dept., Karmaveer Bhaurao Patil College of Engg. & Poly., Satara.