International Journal of Coal Geology 112 (2013) 164–172

Contents lists available at SciVerse ScienceDirect

International Journal of Coal Geology

journal homepage: www.elsevier.com/locate/ijcoalgeo

Direct estimation of diffuse gaseous emissions from coal fires: Current methods and future directions

Mark A. Engle [a,b,*], Ricardo A. Olea [a], Jennifer M.K. O'Keefe [c], James C. Hower [d], Nicholas J. Geboy [a]

[a] U.S. Geological Survey, Reston, VA, USA
[b] University of Texas at El Paso, Department of Geological Sciences, El Paso, TX, USA
[c] Morehead State University, Department of Earth and Space Sciences, Morehead, KY, USA
[d] University of Kentucky, Center for Applied Energy Research, Lexington, KY, USA

ARTICLE INFO

Article history:
Received 1 May 2012
Received in revised form 12 October 2012
Accepted 15 October 2012
Available online 23 October 2012

Keywords:
Coal fires
Emissions
Greenhouse gases
Geostatistics
Sequential Gaussian simulation

ABSTRACT

Coal fires occur in nature spontaneously, contribute to increases in greenhouse gases, and emit atmospheric toxicants. Increasing interest in quantifying coal fire emissions has resulted in the adaptation and development of specialized approaches and adoption of numerical modeling techniques. Overview of these methods for direct estimation of diffuse gas emissions from coal fires is presented in this paper. Here we take advantage of stochastic Gaussian simulation to interpolate CO_2 fluxes measured using a dynamic closed chamber at the Ruth Mullins coal fire in Perry County, Kentucky. This approach allows for preparing a map of diffuse gas emissions, one of the two primary ways that gases emanate from coal fires, and establishing the reliability of the study both locally and for the entire fire. Future research directions include continuous and automated sampling to improve quantification of gaseous coal fire emissions.

Published by Elsevier B.V.

1. Introduction

In situ combustion of coalbeds, coal waste piles, and exposed coal in mines is a known source of greenhouse gases and a multitude of organic and inorganic contaminants (Finkelman, 2004; Hower et al., 2009; Stracher and Taylor, 2004). In attempts to quantify air pollution impacts, recent studies assessed gaseous emissions from coal fires (Carras et al., 2009; Engle et al., 2011, 2012a,b; Ide and Orr, 2011; Litschke, 2005; O'Keefe et al., 2010, 2011; van Dijk et al., 2011), most typically for carbon dioxide (CO_2) and methane (CH_4), using both direct and indirect techniques. Indirect techniques include estimating coal consumption for a given fire through: 1) thermal heat flux derived from airborne thermal infrared (TIR) imagery (Engle et al., 2011, 2012b) and satellite imagery (Tetzlaff, 2004); 2) coal loss estimates reported by coal mine engineers (van Dijk et al., 2011); 3) rate of coal fire advance (Engle et al., 2012b); and 4) growth rates of areas which have been magnetically reset due to heating above the Curie temperature (Ide and Orr, 2011). Coal consumption rates estimated from indirect methods, scaled by the coal heat-content values and concentration of the element of interest in the coal, provide an estimate of the total coal fire emission for that element. Current direct emission estimation methods (i.e., measuring in situ gas emissions or fluxes) require separate measurement of the two major emission pathways: advective vent transport and diffuse soil flux (Engle et al., 2011). Currently, all known direct diffuse soil emission estimates from coal fires rely on measurements from flux chambers. However, the type and application of flux chambers vary from study to study and the geostatistical methods employed to scale-up point-flux measurements are poorly described.

This paper presents and discusses differences between current direct methods used to estimate diffuse soil emissions from coal fires. A detailed case study for estimating diffuse CO_2 emissions from a coal fire, showing field and geostatistical methods, is presented using data collected in November 2009 for the Ruth Mullins coal fire, Perry County, Kentucky, USA (O'Keefe et al., 2010). Lastly, future research directions for estimating diffuse gas emissions and unresolved problems are discussed. The purpose of this research is to provide a snapshot of current techniques and to present ideas for how more robust estimates of coal fire gas emissions can be generated in the future.

2. Field Survey Design and Flux Chamber Measurements

There are several popular approaches for designing field-sampling surveys (Fletcher et al., 1986). Probably the best known is mapping a regularly spaced grid (i.e., square, rectangular, or triangular) across a study area and taking measurements or collecting samples at the grid nodes. This approach has two major problems in terms of application to coal fire surveys: 1) potential for under sampling or missing data for small, highly active emission zones (i.e., in proximity to vents and other thermal features) and 2) lack of data point pairs in close

* Corresponding author at: University of Texas at El Paso, Department of Geological Sciences, El Paso, TX, USA.
E-mail address: engle@usgs.gov (M.A. Engle).

0166-5162/$ – see front matter. Published by Elsevier B.V.
http://dx.doi.org/10.1016/j.coal.2012.10.005

proximity, which makes for difficulty in examining autocorrelation and, thus, developing geostatistical relationships needed to scale-up the flux data to the entire fire area (Section 3). In our own research (Engle et al., 2011, 2012a,b), we have typically started with a grid design where the first set of flux measurements is taken at the grid nodes (see grid points arranged on a triangular geometry in Fig. 1). Another set of measurements is taken in close proximity to vents and thermally active features to ensure that data from high emission areas are represented (vent points in Fig. 1). The final set of measurements within the active portion of the fire is taken slightly further away from the thermally active areas than the vent points, to provide information about how emissions decrease away from point sources (control points in Fig. 1). Another strategy which should also work well for field surveys of diffuse flux measurements is that of Generalized Random Tessellation Stratified (GRTS) spatially-balanced designs (Stevens and Olsen, 2004). The GRTS method produces a spatially-balanced framework while also allowing for more intense sampling in active coal fire regions (i.e., stratified sampling) but requires specialized software and a field survey of the study area prior to sampling. Regardless of the approach, pre-flux measurement trips to the study area to define the extent of the coal fire area combined with remote sensing imagery, such as TIR data, allow for better field survey design. One weakness in all of these designs is a lack of techniques to deal with sampling points that are inaccessible, due to unsafe conditions in the field. This issue is a serious one and highlights one of the potential problems with direct measurements. The GRTS method does allow for on-the-fly modification of the sampling strategy. We have previously dealt with inaccessibility by measuring fluxes at several locations in the nearest safe vicinity around the planned sampling point but acknowledge that this may introduce bias into the estimates. Once a survey design has been selected, measurement of fluxes, using chamber techniques, at the specific sampling points can be initiated.

There are two basic designs for chambers used to measure soil gas fluxes: accumulation and dynamic open chambers. Accumulation designs, in which the chamber is placed firmly against the soil surface and exchange with the surrounding atmosphere is limited, rely on determining the rate of gas accumulation inside the chamber as a result of exchange across the soil–air boundary. Eventually, the gas concentration inside the chamber achieves a steady-state with the underlying soil gas, but it is the period of accumulation which is of most interest. Dynamic closed chambers, a type of accumulation design in which air is circulated from chamber to an analyzer and then returned to the chamber, have been employed to measure CO_2 fluxes in most coal fire studies (Engle et al., 2011, 2012a,b; Litschke, 2005). Circulation of air within the closed-loop system is repeated multiple times until a stable measurement of the linear gas accumulation rate ($\partial C/\partial t$) can be made. Gas flux (F) is then calculated via:

$$F = \left[\frac{\rho V}{A}\frac{\partial C}{\partial t}\right] \quad (1)$$

where ρ is gas density, V is the total volume of the closed-loop flux measurement system, and A is the area of the chamber footprint (Bergfeld et al., 2001). Dynamic closed chambers are commonly used for CO_2 flux measurement (Rochette et al., 1992), owing to the ability to measure CO_2 concentrations at a relatively high frequency (>1 Hz) with the use of an infrared gas analyzer (IRGA).

By comparison, dynamic open chambers allow ambient air to enter through holes on the side of the chamber, the inlet, follow a pathway across of the top of the soil, and exit through a chamber outlet. In this case, concentrations of the gas being investigated are being measured both at the chamber inlet and at the outlet, and the difference in concentration between the two measurement locations (ΔC) is ascribed to contribution from the soil underneath the chamber. In this case, if the rate of airflow through the chamber is steady, soil gas flux (F) is determined via:

$$F = \frac{Q\Delta C}{A} \quad (2)$$

where Q is the flow rate of ambient air through the chamber (Gao and Yates, 1998). Dynamic open chambers are more commonly used in environmental applications for flux measurement of compounds that require long (several minutes) sampling or measurement intervals, such as volatile organic compounds and mercury (Eckley et al., 2010; Reichman and Rolston, 2002). Carras et al. (2009) used a dynamic open chamber to measure fluxes of CO_2 and CH_4 at coal fires in Australia.

Both types of chamber designs have distinct advantages and disadvantages, which should be considered in coal fire studies. A general overview of potential issues and artifacts with flux chamber sampling is provided by Welles et al. (2001). Issues stemming from both chamber designs include soil disturbance from placement or insertion of chamber into the soil and change in air temperature, soil temperature, barometric pressure and wind speed at the soil–air interface inside the flux chamber, relative to ambient conditions (Rochette et al., 1992; Welles et al., 2001). Dynamic closed chamber measurements

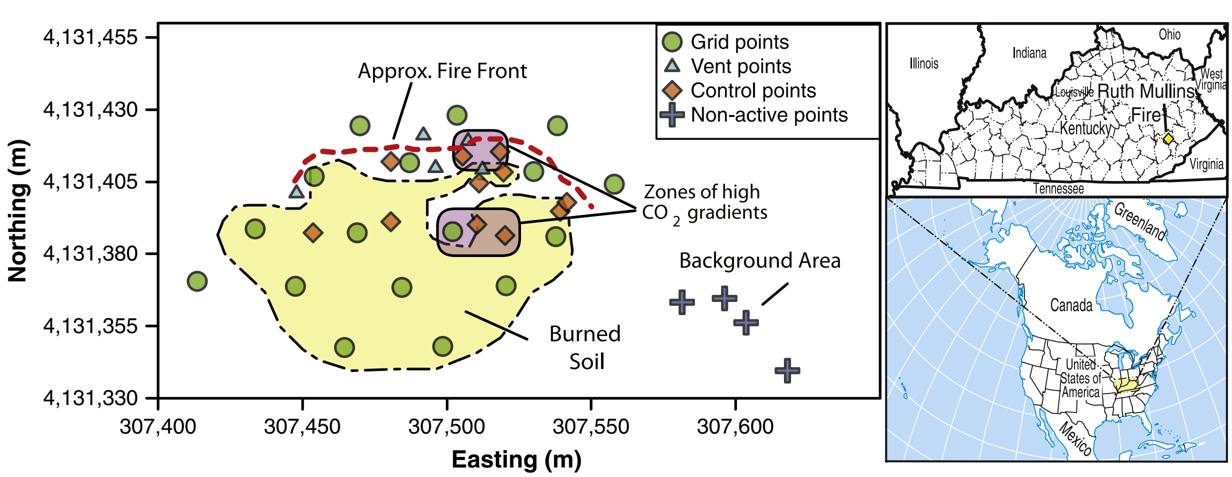

Fig. 1. Map showing locations of CO_2 flux measurement points, extent of area burned by fire the previous summer, and approximate boundary of the fire front for the Ruth Mullins coal fire as of November 2009.