

# Methods to quantify diffuse CO<sub>2</sub> emissions from coal fires using unevenly distributed flux data

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### Introduction

- Coal fires occur in all coal-bearing parts of the world, emitting greenhouse gases (CO<sub>2</sub>, CH<sub>4</sub>) and toxic substances (Hg, CO, organics, etc.)
- Coal fires can persist for decades or longer in underground coal mines, coal waste piles, and unmined coal beds.



 The contribution of coal fires to the global pools of atmospheric CO<sub>2</sub> and Hg is poorly constrained.



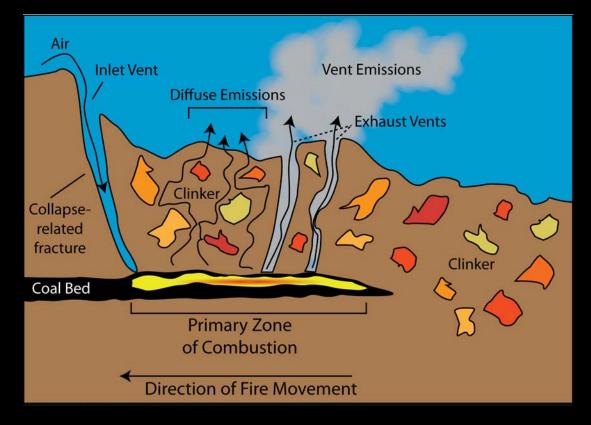
Photo of Glenn and Janet Stracher collecting mineral samples at the Mulga, Alabama, gob pile fire. Taken by James Hower.

### **Types of Emissions**

<u>Vent Emissions-</u> Gas emitted through vents that exhaust from the fire (Hower et al., 2009; O'Keefe et al., 2010).

<u>Diffuse Emissions-</u> Gas diffused through the soil above an underground fire (Carras et al., 2009).

Cartoon showing a conceptual model of a coal fire including depictions of diffuse and vent emissions.





### **Purpose**

 The purpose of this presentation is to demonstrate methods to quantify <u>diffuse</u> CO<sub>2</sub> emissions from unevenly distributed flux data, using the Mulga gob pile fire in northern Alabama, U.S., as an example.

### Focus

- To provide clarity about our methods.
- To explain fundamental principles behind both the field methodology and the data analysis portions of this research.



### The Mulga Gob Fire

Fire started ~1990

Reignited in 2006 as a result of spontaneous combustion (Stracher et al., 2009)

Site being remediated

Data collection = Dec, 2008

Fire Size = 9.1 hectare

Max. Surface

Temperatures >325 ° C



Photo of smoke generated by the Mulga gob pile fire. Taken by James Hower.



### Measuring in situ CO<sub>2</sub> Fluxes

### **Accumulation (Static) Chamber Method**

### Soil flux (F) of CO<sub>2</sub> (g/m<sup>2</sup>/d)

$$F = \left[ \frac{\rho V}{A} \cdot \frac{\partial C}{\partial t} \right]$$

 $\rho$  = gas density

V = flux system volume

A = footprint of the flux chamber

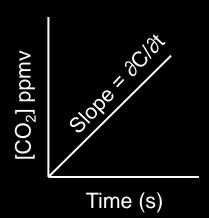
 $\partial C/\partial t$  = rate of accumulation of CO<sub>2</sub>

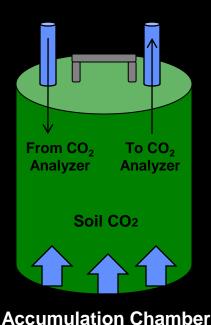
#### Field Instrumentation

- Li-Cor Li-820 non-dispersive infrared (NDIR) gas analyzer
- 3-liter WestSystems accumulation chamber
- Handheld personal digital assistant (PDA)

For more details see Bergfeld et al (2006).







### **Additional Method Details**

### **Ancillary Data**

- Co-collected at every sampling point
- Surface temp.
- Soil temp. at 10 cm depth
- GPS location
- Notes regarding mineralization/alteration and location relative to vents

### **QA/QC** Criteria

- Calibrate NDIR CO<sub>2</sub>
   analyzer daily using
   zero and span
   calibration gases
- Collect at least 10% of the fluxes in triplicate
  - %RSD ranged from 26-37%
- Ensure concentration data fall within range of calibration gases



### In An "Ideal" Situation:

- 1. Select sampling points along a grid around the area of obvious fire activity (green)
- 2. Measure at points of interest such as near vents (orange)
- 3. Add "control" points near areas of high flux (cyan)
- 4. Measure fluxes at background points away from the fire (magenta)



Example of distribution of CO<sub>2</sub> flux sampling points at the Ruth Mullins fire, eastern Kentucky, U.S. Image from USGS.

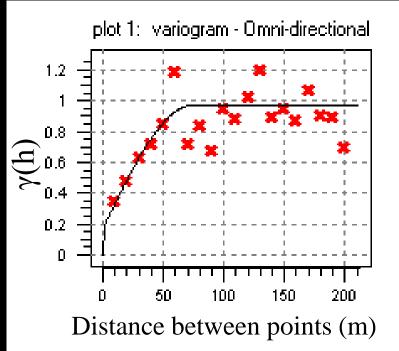
### Basic Concepts Of Geostatistical Interpolation Methods

 Geostatistical methods focus on spatial

correlation within the data

- i.e., are points closer together better correlated than those further apart?
- Quantify spatial correlation using a semivariogram





In this semivariogram plot, when points are closer together, the semivariance ( $\gamma$ (h)) is smaller (i.e., they are more similar) than when they are further apart. The black line shows the fitted model used to describe this spatial continuity in the data.

## For the Geostatistically Savvy - Interpolation For Ideal Data: Sequential Gaussian Simulation (SGS)

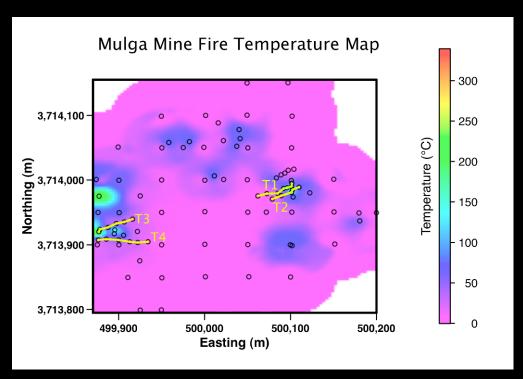
- 1. Convert CO<sub>2</sub> flux data to normal scores (mean=0, variance=1)
- Develop semivariogram model to describe the spatial correlation among the data
- 3. Break the study area into cells, or nodes, of equal size
- 4. Define number of realizations (n>100)
- 5. For each realization, define a random path through the nodes
- 6. For each node, use the original hard data plus any previously simulated values to estimate, via kriging, the mean and variance for that node

- 7. Use the mean and the variance to describe a Gaussian distribution for that node
- 8. Draw a value from that Gaussian distribution and apply the simulated value to that node
- 9. Complete steps 6-8 until all nodes in the realization have been assigned values
- 10. Repeat steps 5-9 until all realizations have been simulated
- 11. Back transform the normal scores to raw values for each realization

### Mulga Sampling Scheme

- Due to site access constraints, CO<sub>2</sub> flux was only measured at 24 points along 4 transects (T1-T4)
- Surface and soil temperatures were measured at an additional 69 points
- So, soil temperature was measured at a total of 93 points





Interpolated soil temperature map for the Mulga fire showing location of soil temperature points (circles) and transects along which CO<sub>2</sub> flux was measured (yellow lines). The gradual trends in these interpolated values suggest spatial continuity within the data.

### **Application of ideal methods?**

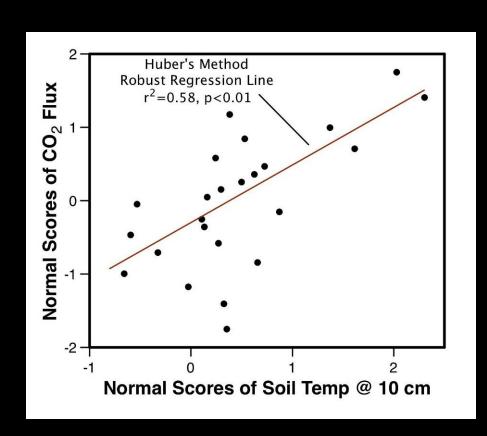
- To interpolate the CO<sub>2</sub> flux data, we need many (>40) data points distributed evenly over the study area
- Instead we have few data and they are concentrated in two areas



Application of the CO<sub>2</sub> accumulation chamber at the Mulga Gob Fire. Photo by Glenn Stracher.



### Robust regression results



But, we have a statistically significant (p<0.01)relationship between CO<sub>2</sub> flux and soil temperature (10 cm depth)



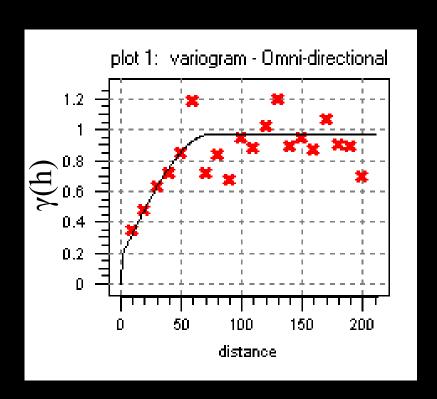
### Sequential Gaussian Co-simulation (SGC)

- A data interpolation method similar to SGS
- Based on:
  - Distribution and spatial correlation of each variable
  - Utilizing high soil temp measurement density
  - Using relationship between the two variables to create robust estimates of CO<sub>2</sub> emission

- Software
  - SGeMS 2.1 (Remy et al., 2009)
    - Open-source
    - Actively updated
    - GUI-based

### SGC Steps – Part 1

- Transform both variables to normal scores (NS)
- Model variogram for NS of each variable
  - In this case there are too few pairs to do this for CO<sub>2</sub> flux
- Calculate correlation between NS of each variable

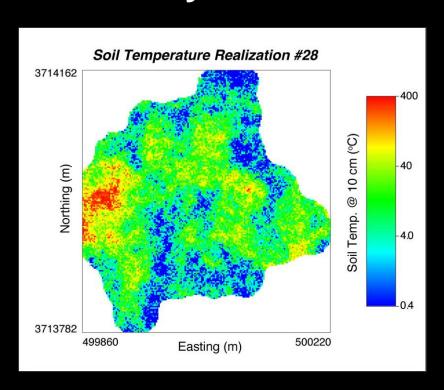


Omni-directional semivariogram model of the soil temperature data



### SGC Steps – Part 2

 Use SGS to generate 100 realizations of soil temperature for the study area

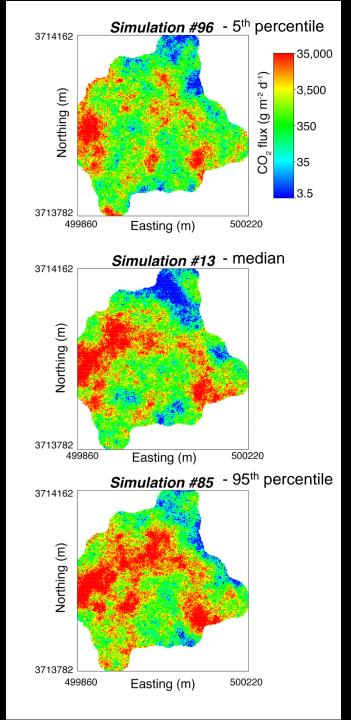


- Perform SGC on the NS of the CO<sub>2</sub> flux data using:
  - SGS maps of soil temp
  - Variogram model of the soil temp data as a surrogate for CO<sub>2</sub> flux
  - Correlation results of the NS scores
    - r=0.66-0.76 depending on method
  - MM1 model for crossvariance (Almeida and Journel, 1996)

### **SGC** Results

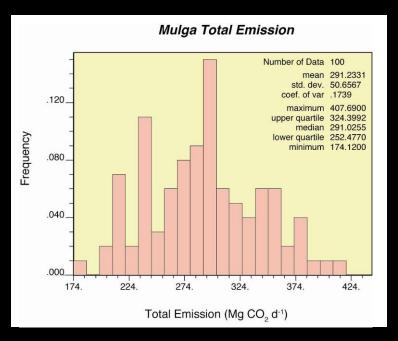
- SGC generates 100 realizations of CO<sub>2</sub> flux and soil temp for the study area
  - We are most interested in the former
  - Because it's a stochastic method, each realization is different

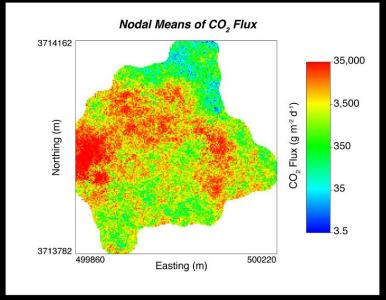




## Estimating Site CO<sub>2</sub> Emissions

- Summing the CO<sub>2</sub>
   flux data for each
   realization provides
   100 estimates of total
   emissions
  - median = 291 Mg/day
  - 5<sup>th</sup> percentile = 210 Mg/day
  - 95<sup>th</sup> percentile = 378Mg/day







### **Caveats**

- Data were collected on a single day in December, 2008
  - May not be representative of "typical" conditions
- These procedures require expertise in flux measurement techniques and geostatistical methods

- Analysis improved by more data and evenly spaced locations
- No measurement technique is without errors
- Unquantified vent emissions

### Conclusions

- Methods to estimate diffuse CO<sub>2</sub> emissions from ideal and non-ideal data were presented
  - sequential Gaussian simulation (ideal)
  - sequential Gaussian cosimulation (non-ideal)
    - need a second variable that correlates with CO<sub>2</sub> flux
- For the 8.7 ha area modeled, CO<sub>2</sub> emissions estimated at 210-378 Mg/day
  - Mean Flux =  $2,400-4,400 \text{ g m}^{-2} \text{ d}^{-1}$
  - For comparison, a 500 MW coal fired power plant emits ~10,000 Mg d⁻¹ (O'Keefe et al., 2010).



### References - I

- Almeida, A., and Journel, A., 1996, Joint simulation of multiple variables with a Markov-type coregionalization model: Mathematical Geology, v. 26, no. 5, p. 565-588.
- Bergfeld, D., Evans, W.C., Howle, J.F., and Farrar, C.D., 2006, Carbon dioxide emissions from vegetation-kill zones around the resurgent dome of Long Valley caldera, eastern California, USA: Journal of Volcanology and Geothermal Research, v. 152, no. 1-2, p. 140-156.
- Carras, J.N., Day, S.J., Saghafi, A., and Williams, D.J., 2009, Greenhouse gas emissions from low-temperature oxidation and spontaneous combustion at open-cut coal mines in Australia: International Journal of Coal Geology, v. 78, no. 2, p. 161-168.
- Hower, J.C., Henke K., O'Keefe J.M., Engle M.A., Blake D.R., and Stracher G.B., 2009, The Tiptop coal-mine fire, Kentucky: Preliminary investigation of the measurement of mercury and other hazardous gases from coal-fire gas vents: International Journal of Coal Geology v. 80, no. 1, p. 63-67.

### References - II

- Kolker, A., Engle, M., Radke, L., Hower, J., O'Keefe, J., Heffern, E., ter Schure, A., Stracher, G., Prakash, A., Román-Colón, Y., and Olea, R., 2009, Measuring CO<sub>2</sub> emissions from spontaneous coal fires in the U.S.: Proceedings, Annual International Pittsburgh Coal Conference, Pittsburgh, PA, September, 2009, 7 p.
- O'Keefe, J.M.K., Henke, K., Hower, J.C., Engle, M.A., Stracher, G.B., Stucker, J., Drew, J.W., Staggs, W.D., Murray, T.M., Hammond, M.L., III, Adkins, K.D., Mullins, B.J., and Lemley, E.W., 2010, CO<sub>2</sub> CO, and Hg emissions from the Truman Shepherd and Ruth Mullins coal fires, eastern Kentucky, USA: Science of The Total Environment v. 408, no. 7, p. 1628-1633.
- Remy, N., Boucher, A., and Wu, J., 2009, Applied Geostatistics with SGeMS: A User's Guide: Cambridge University Press, 284

### References - III

Stracher, G.B., Finkelman, R.B., Hower, J.C., Pone, J.D.N., Prakash, A., Blake, D.R., Schroeder, P.A., Emsbo-Mattingly, S.D., and O'Keefe, J.M.K., 2009, Natural and anthropogenic coal fires, *in* the Encyclopedia of the Earth, http://www.eoearth.org/article/Natural\_and\_anthropogenic\_coal\_fires

