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Mapping and quantifying CO_2 leakage using the Ground CO_2 Mapper.

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Abstract

The standard method for mapping and quantifying CO_2 leakage flux from the ground surface to the atmosphere involves performing numerous point flux measurements using the accumulation chamber technique and then applying geostatistical interpolation to infer spatial distribution and estimate total mass transfer. Monte Carlo simulations using the program MCFlux have recently demonstrated, however, that uncertainty in the resultant estimate can be large if the chosen sample spacing is insufficient to capture the spatial complexity and size distribution of the leakage anomalies. In an effort to reduce this uncertainty we have developed a new tool, called the Ground CO_2 Mapper, that rapidly measures the concentration of CO_2 at the ground surface as a proxy for flux. Recently published results have illustrated the capabilities of the Mapper in terms of sensitivity and spatial resolution, as well as possible influencing parameters such as wind strength. The present work examines the potential of combining Mapper results with point flux measurements (using multivariate geostatistics) to improve data interpretation, with the MCFlux program being used once again to assess uncertainty in the final estimates.

Keywords: CCS; monitoring; geochemistry; near-surface; gas; water.

1. Introduction

The standard method for mapping and quantifying CO_2 leakage flux from the ground to the atmosphere involves performing numerous point flux measurements using the accumulation chamber technique and then applying geostatistical interpolation to infer spatial distribution and estimate total mass transfer [1]. Recently, however, Beaubien et al. [2] developed a probabilistic Monte Carlo software package (MCFlux) and used it to show that this approach can produce large uncertainties that are a function of sampling density versus anomaly size, regardless of the geostatistical technique used. This is because an economically and logistically feasible number of measurement points may yield a sampling density that misses anomalies and/or poorly defines their spatial distribution and magnitude. Additional, higher-density data of a related or proxy parameter would be of great assistance for these types of surveys, as this secondary dataset could be used to choose more significant flux measurement locations or to conduct multivariate geostatistical processing to more accurately estimate total flux distribution and magnitude.

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To this end, we have developed a low-cost, sensitive and rapid tool, the Ground CO_2 Mapper (or "Mapper" for short), to collect high resolution secondary data to help reduce the uncertainty of leakage mapping and quantification [3]. The Mapper measures CO_2 at the ground-atmosphere contact, an interval where leaking CO_2 can accumulate due to its greater density relative to air and, more importantly, reduced wind mixing due to surface friction effects [4]. As greater CO_2 flux will yield greater concentrations at this boundary, the latter represents a proxy for the former. In addition, as concentration can be measured at the ground surface much more rapidly than flux there is the potential that this parameter can yield more detailed spatial datasets.

The present work, which is on-going, uses multivariate geostatistical methods to combine point flux data (the low resolution, quantitative primary dataset) with Mapper survey results (the high resolution, semi-quantitative secondary dataset) to better represent the true CO_2 leakage distribution. Initial tests described here used co-kriging on a limited dataset, whereas future work will focus on using "kriging with an external drift" (KED) because it is more adapted to the automatic processing of large numbers of simulations. MCFlux will be modified so that we can assess the uncertainty of total flux estimates using this newly developed approach; the field work and data processing necessary to accomplish this goal will be completed in the early fall of 2022.

2. Methods

2.1. MCFlux

MCFlux is a program written in Visual Basic 6 to conduct Monte Carlo simulations that assess the impact of sample spacing and strategy on the probability of finding CO_2 leakage anomalies and on the uncertainty of their total flux estimates [2]. The core of the algorithm is built around a very high resolution flux dataset (created completely artificially or using real data as the initial foundation) that has 1 m node spacing and a user-defined total size that is only limited by computer capabilities (up to 1 km^2 has been tested thus far). The program subsamples the synthetic data a statistically relevant number of times for each chosen sample spacing (or total number of samples), thus producing a series of unique but equivalent realizations that can be used to define the probability distribution. When looking at the probability of finding an anomaly, up to 5000 realizations can be performed for each test because run times are short. For simulations conducted to assess flux estimate uncertainties, however, it is only realistic to perform 100 to 500 realizations due to the higher computational costs. External software is called to conduct the spatial interpolation necessary for these simulations, with the commercial program Surfer (Golden Software) being used for kriging and the freely available geostatistical software package GeoLIB [5] being used for sequential Gaussian simulations and kriging with an external drift. A total of five different sampling strategies are available, including regular grids (square, offset, triangular), distributed random and purely random sampling.

2.2. Ground CO_2 Mapper

The Mapper was developed to rapidly map in high resolution the distribution of CO_2 concentration at the ground-atmosphere interface, given that leaking dense CO_2 can accumulate in this poorly mixed interval (known as the roughness height or aerodynamic roughness length). To conduct a survey the Mapper is moved across an area in a uniform pattern and at a constant walking pace. Drawing air from a tube dragged on the ground surface, the Mapper analyses the CO_2 content with a small-volume NDIR (Non-Dispersive Infrared) sensor and associates the resultant value with a precise location determined using a Differential Global Positioning System (DGPS). The Mapper response is rapid (T_{67}=0.75 seconds), has little memory due to rapid wash-out of the sensor volume and has a low level of noise (1σ<10ppm). This results in a high level of sensitivity, despite being a mobile platform, and yields high-resolution, detailed maps that are spatially accurate. Along track sample spacing is about 20 cm at a walking speed of 2.5 km hr^{-1} thanks to a 4 Hz sampling frequency, while it is recommended to maintain a between-track spacing of 2 to 5 m. According to controlled experiments and mixing model results, Graziani et al. [3] predict that the Mapper has a 60% probability of defining an intersected 2m-wide anomaly having a peak flux of 75 g m^{-2} d^{-1} in a background field of 20 g m^{-2} d^{-1} under the tested wind and vegetation conditions. During measurements conducted in a 4000 m^2 field in which numerous natural leaks of geogenic CO_2 occur, these same authors note that an 80-point flux survey with an average 10 m node spacing took > 4 hours to complete whereas a Mapper survey conducted with 2 m track spacing and a walking speed of 2.5 km hr^{-1} only took about 35 minutes and yielded much more detailed results.

2.3. Sample collection

Field work was conducted near the town of Ailano, central Italy, where large volumes of geogenic, deep-origin CO_2 is naturally leaking to the atmosphere from various locations throughout the valley [6]. Measurements for preliminary tests using co-kriging (see below) were performed in October 2018 in an essentially flat, ca. 4000 m^2 agricultural field that had 2-3 cm cut grass at the time of the surveys. A total of 80 flux measurements were made using the accumulation chamber technique, with 60 points along a regular, 10 m spacing square grid pattern and 20 at additional in-fill points chosen to better define some anomalies. Various Mapper surveys were performed during the same period using a walking speed of either 2.5 or 5 km hr^{-1} and with between-track distances ranging from 2-4 m. Instead, Mapper surveys and high density flux measurements (ca. 2.5 m spacing) in a 100x100 m field are planned for early September, 2022, to create the dataset necessary to perform Monte Carlo simulations using kriging with external drift.

2.4. Multivariate Geostatistics

Multivariate spatial prediction techniques [7] describe a class of geostatistical methods that combine more than one variable to better estimate the distribution of a parameter of interest, with bivariate methods clearly representing the case where only two parameters are considered. For a valid application of these methods there must be clear physical relationship and a strong cross-correlation between the parameters. In general, the spatial distribution of a primary but sparsely sampled variable (in our case, CO_2 flux) is estimated with the help of a more densely sampled secondary variable (in our case, ground CO_2 concentration measured by the Mapper) in order to reduce the prediction variance and uncertainty [8]. Two methods are used in this study, co-kriging (CoK) and kriging with an external drift (KED).

CoK, which was chosen for preliminary tests given its wide-spread use in the literature (e.g., [9]), involves estimating the autocorrelation for each variable as well as all cross-correlations between them, resulting in more variography, modelling and computation time compared to kriging. Care must be taken when conducting CoK, as any variability introduced during these additional steps may minimize any hoped-for gains in precision. KED, which is mathematically equivalent to Universal Kriging and Regression Kriging, combines regression between the dependent and secondary variables with simple kriging of the regression residuals [10]. For proper and valid use of KED it is necessary that the secondary variable varies smoothly in space and it must be known at all locations of the primary variable and at all locations to be estimated [5]. This method produces maps that reflect the trends of the secondary variable due to the assumptions of the algorithm, however care must be taken as this is not necessarily proof that the primary variable has that same trend. KED requires less variography compared to CoK and thus it represents a better choice where automation linked to Monte Carlo simulations is required.

3. Preliminary Results

Initial tests involved the use of an existing dataset collected from Ailano in 2018 to examine the potential for using CoK to process flux and Mapper data. The linear fit between the complete 80 point CO_2 flux dataset and the average ground CO_2 concentration measured by the Mapper (within a 1m radius around each of those points) shows that these two parameters are highly correlated (R^2=0.907, p<0.0001), which is a fundamental requirement of CoK.

Ordinary Kriging was first conducted directly on the 80-point flux dataset (Fig. 1a). A total of four cokriging tests were performed, with the primary variable consisting of the flux dataset and the secondary variable consisting of the raw data from one of the two, 2.5 km hr^{-1} surveys (Fig. 1b, c) as well as the 3-point running average (3RA) for each, given that small-scale Mapper variability could potentially affect nugget size (maps not shown).

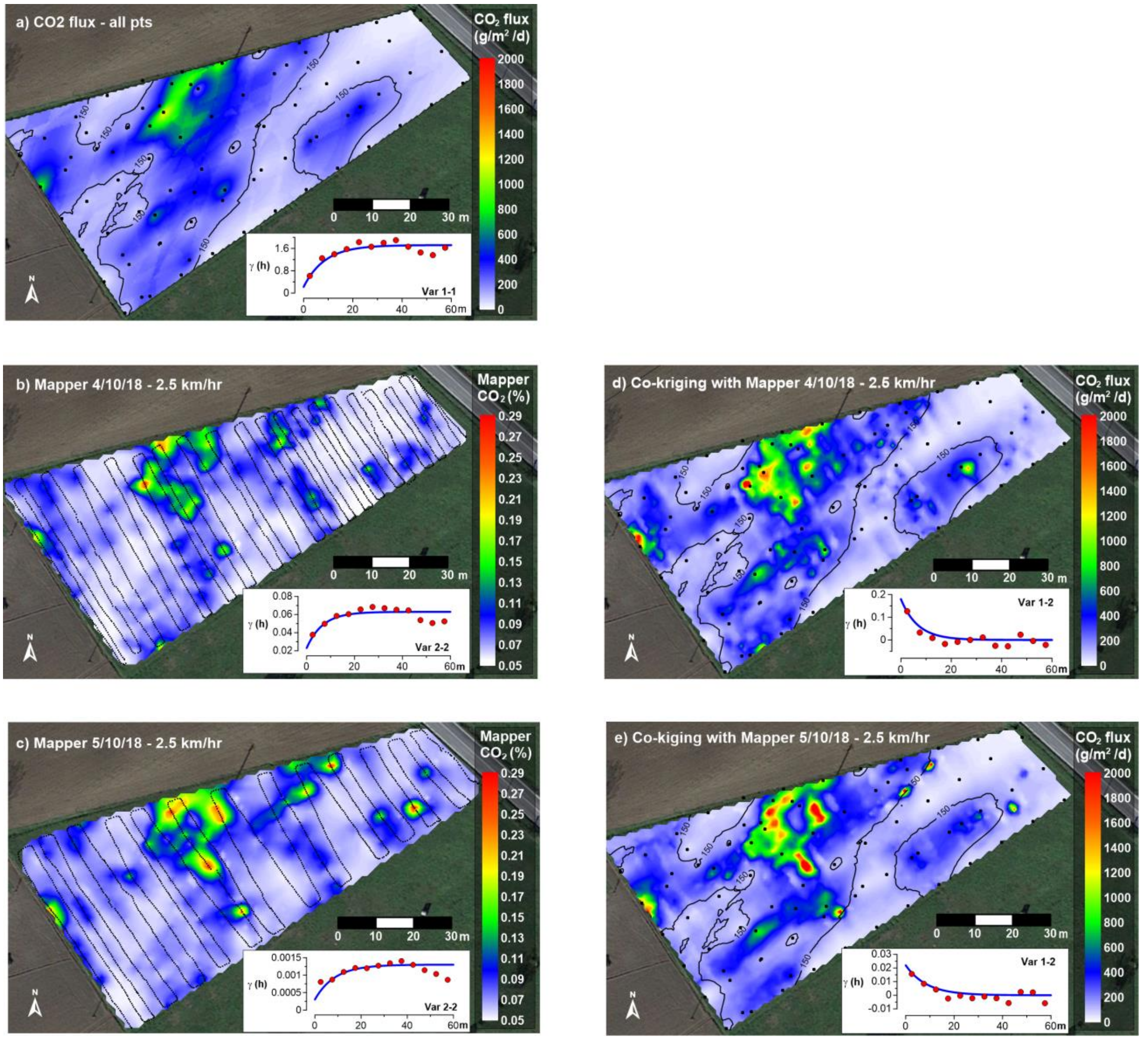

Fig. 1. Contour plots created using kriging for the CO_2 flux field data (a) as well as the Mapper field data collected at a walking speed of 2.5 km hr^{-1} on two successive days (b, c) as well as contour plots created using co-kriging using the flux data together with the first (d) and second (e) Mapper datasets. Black symbols in (b) and (c) show Mapper sample points along the traces, while the 150 g m^{-2} d^{-1} contour line in (d) and (e) are taken from (a) for reference purposes. Calculated variograms are given for each plot.

In general, the analysis of experimental variograms shows that the anisotropy parameters of the primary CO_2 flux dataset (direction ~45° and anisotropy ratio ~ 2) are comparable with those calculated using CoK with all secondary datasets. Kriging and co-kriging results were compared by calculating the Root Mean Squared Standardized Error (RMSSE), a diagnostic statistic that is calculated by dividing each prediction error by its estimated prediction standard error; the closer the RMSSE value is to 1 the lower the error. Table 1 shows how the RMSSE values for all CoK simulations (0.87-0.9) are higher that calculated for the ordinary kriging of the flux data only (0.74), implying that the multivariate geostatistical approach using Mapper data is valid and can potentially improve accuracy and reduce uncertainty of CO_2 leakage mapping and quantification. Table 1 also reports the calculated total flux for these tests, with kriged flux yielding around 1.26 T_{CO2}/d compared to around 1.4 T_{CO2}/d for all CoK results. The lower value for the former may be linked to the well-known fact that kriging tends to smooth results which leads to underestimation of peak values.

Table 1. Summary of quantification tests performed using flux data (kriging) and flux plus Mapper data (cokriging). "-raw" = original Mapper data; "-3RA" = three point running average; RMSSE = Root Mean Squared Standardised Error.

Test	*Primary dataset*	*Secondary dataset*	*Method*	*RMSSE*	*Total flux (kg_{CO2}/d)*
1	fCO_2-80pts	-	Krig	0.736	1,257
2	fCO_2-80pts	041018a-raw	CoK	0.867	1,470
3	fCO_2-80pts	051018c-raw	CoK	0.865	1,456
4	fCO_2-80pts	041018a-3RA	CoK	0.870	1,379
5	fCO_2-80pts	051018c-3RA	CoK	0.903	1,422

4. Summary and ongoing work

The preliminary tests reported here indicate the potential for using multivariate geostatistics to combine point flux measurements and Ground CO_2 Mapper surveys in order to reduce uncertainty in leakage flux mapping and quantification. For Carbon Capture and Storage (CCS) applications, the resultant greater spatial resolution and leakage estimate accuracy will improve stakeholder confidence, site safety, and any eventual carbon credit auditing accuracy. In the immediate future a highly detailed sampling campaign is planned, with the goal of creating a high-resolution dataset that can be used to conduct Monte Carlo simulations using sub-sampled flux values together with Mapper survey results. For this work Kriging with an external drift will be used instead of co-kriging due to the fact that KED requires less variography and thus is better adapted for the automated processing of large numbers of Monte Carlo simulations. It is foreseen that this probabilistic approach will allow for a more quantitative assessment of the approach and its impact on overall uncertainty, and may also indicate how the Mapper can help reduce the number of flux measurements needed without sacrificing overall accuracy and precision.

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