8.3 EVALUATING LAND SURFACE FLUX OF METHANE AND NITROUS OXIDE IN AN AGRICULTURAL LANDSCAPE WITH TALL TOWER MEASUREMENTS AND A TRAJECTORY MODEL

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1. Introduction

As the second and third primary greenhouse gases, methane (CH_4) and nitrous oxide (N_2O) have been researched extensively for their global budgets. However, large uncertainties still exist in estimating the strength of the sources and sinks related to agricultural activities. For example, estimates of N_2O emission from agricultural activities, accounting for about a quarter of the total anthropogenic emission, vary in a wide range of 0.6 to 14.8 Tg N yr^{-1} (Mosier et al., 1998).

Each of flux measurement methods for CH_4 and N_2O has its own advantages and drawbacks. The chamber method can be used to quantify the component fluxes from the soil and the plant in an ecosystem, but has difficulty to tackle the heterogeneous nature of the flux on the landscape scale. The eddy covariance method, used for CO_2 flux measurement, has larger footprint than the chamber method, but its application in the measurement of CH_4 and N_2O fluxes is still limited by the detection technique for CH_4 and N_2O concentration. Instrument problems such as signal drift and high frequency loss can lead to bias up to 300% (Kroon et al., 2010a; Kroon et al., 2010b). The flux-gradient method has similar footprint as the eddy covariance and has less stringent requirement for CH_4 and N_2O analyzer in terms of response time and measurement frequency, (Wolf et al., 2008), but requires the assumption that all the scalar quantities are transferred indiscriminately by turbulent eddies.

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The objective of this study was to quantify the CH_4 and N_2O flux in a landscape dominated by soybean and corn cultivation. We used a modified Bowen ratio (MBR) method with the gradient measurement of CH_4 and N_2O on a tall tower. In addition, the fluxes were tested by using Stochastic Time-Inverted Lagrangian Transport Model (STILT) and the concentration observation on 200 m.

2. Methods

The experiment took place at a tall tower site at the University of Minnesota Outreach, Research and Education Park from August to September, 2009. A tunable diode laser (TDL) and an infrared gas analyzer (IRGA) were used to measure the concentrations of CH_4, N_2O, CO_2 and H_2O at the height of 3 m and 200 m above the ground (Griffis et al., 2008). The CH_4 and N_2O concentrations measured by the TDL analyzer were calibrated against pure nitrogen, and a span gas of known CH_4 and N_2O concentrations in every 30 s site cycle. The IRGA was calibrated with a standard CO_2 gas and a dew point generator at the beginning of the experiment.

The CH_4 and N_2O fluxes were calculated with the MBR method (Meyers et al., 1996; Werner et al., 2003). This method assumes that all scalar quantities in air are transferred indiscriminately by turbulent eddies. Under this assumption, the CH_4 or N_2O flux was calculated by

$$F_2 = F_1 \frac{\partial c_2 / \partial z}{\partial c_1 / \partial z}$$

In the equation, F_2 is the flux of CH_4 or N_2O, F_1 is the flux of CO_2, $\partial c_2 / \partial z$ is the gradient of CH_4 or N_2O, and $\partial c_1 / \partial z$ is the gradient of CO_2. Here, the CO_2 flux was the average of the eddy covariance

flux measured simultaneously in a soybean and a corn field near the tall tower (Griffis et al., 2008). The calculated CH_4 and N_2O fluxes were the mean biogenic fluxes of the cultivated cropland, assuming that soybean and corn each accounted for 50% of land area in the tall tower footprint.

We deployed the STILT to obtain the influence function of surface flux on the concentration variation on 200 m of the tower (Lin et al., 2003). The influence function (footprint) was coupled to four types of surface flux data to simulate the concentration variation: 1) constant and homogeneous emission rate; 2) biogenic flux calculated from above method, assuming the region is fully covered by cropland; 3) anthropogenic flux derived from Emissions Database for Global Atmospheric Research (EDGAR) inventory, assuming emission rate constant in time; 4) biogenic flux and anthropogenic flux. The simulated gas concentration variation was noted by ΔC_{con}, ΔC_{bio}, ΔC_{ant}, ΔC_{all}.

3. Results and Discussion

The MBR results are given in Fig. 1. The results suggest that the landscape emitted both CH_4 and N_2O at night (18:00-06:00 LST) at an average rate of 0.02 μmol m^{-2} s^{-1} and 5×10^{-4} μmol m^{-2} s^{-1} and appeared to take up these two gases during the day (07:00-17:00 LST).

The nighttime CH_4 and N_2O fluxes calculated by the MBR method might be questionable, as the MBR assumption usually does not work at night. But several lines of evidence have been found to support the calculated fluxes: 1) The diffusivities for CO_2 and H_2O, calculated from their concentration gradients measured on the tall tower and the fluxes measured in the soybean and corn fields, were linearly correlated, with the correlation coefficient of 0.42 (number of observations 694). That the slope of the regression was close to unity suggests that the assumption that scalars are transported discriminately was a good approximation. 2) Midnight (23:00-04:00 LST) block average of the CH_4 and N_2O concentration was positively and linearly correlated with the midnight block average of the CO_2 concentration (Fig. 2). This correlation suggests that the CH_4 and N_2O accumulated similarly near the ground as CO_2.

The negative daytime fluxes for both CH_4 and N_2O were supported by the modeling result. We first tested the model on CO_2. Driven by a diurnally varying biogenic surface flux that matches the observed diurnal composite flux of the soybean and corn fields, the STILT model reproduced the diurnal phase of the CO_2 concentration observed at the 200 m height on the tall tower, but the magnitude of modeled variation is larger than the observation. The overestimation of the CO_2 concentration variation may be caused by the arbitrary assumption that the landscape is all covered by soybean and corn cultivation. We found that by reducing the diurnal amplitude of the surface flux to 35% of the observed value (optimization coefficient LC = 0.35), the modeled CO_2 concentration produced the best fit with the observation (Fig. 3).

The same method was applied to CH_4 and N_2O (Fig. 4, Fig. 5). The correlation coefficients between the observed concentration variation (ΔC_{obs}) and modeled concentration variation (ΔC_{con}, ΔC_{bio}, ΔC_{ant}, ΔC_{all}) are summarized in Table 1.

Forced with a constant emission value according to the observed nighttime flux, the modeled CH_4 and N_2O concentration variation (ΔC_{con}) at 200 m has poor agreement with the observation,. In comparison, the simulation with the biogenic flux (ΔC_{bio}) agrees reasonably well with the observation; here the surface flux was prescribed according to the observed diurnal pattern shown in Fig. 1. The result suggests that the diurnal pattern of the biogenic flux for CH_4 and N_2O, especially the daytime uptake, exists and dominates the atmospheric concentration variation of CH_4 and N_2O over the agricultural landscape. In the third simulation, the model was forced by the gridded inventory data from EDGAR without any diurnal variation. The original 1°by 1°data were interpolated to 0.2° by 0.2° resolution. The poor correlation between this simulation (ΔC_{ant}) and ΔC_{obs} indicates that either the spatial heterogeneity of the anthropogenic emissions is not well defined in the EDGAR Inventory, or the influence of the anthropogenic emission on the atmospheric CH_4 and N_2O concentrations is not important in the agricultural landscape.

With the optimization coefficient of LC (LC $_{CO2}$=0.35; LC $_{CH4}$=1; LC $_{N2O}$=0.35), the final set of simulation (ΔC_{all}) has the diurnal variation patterns and the magnitude of variations that are comparable with ΔC_{obs}. In this simulation, the surface flux was the sum of the gridded EDGAR inventory flux and the observed biological flux according to Fig. 1 but adjusted by LC. The LC value of 0.35 seems reasonable because the

cultivated cropland, which has much higher productivity than other vegetation types, accounts for only 50% of the footprint of the observation point at the 200 m height. The bigger optimization coefficient LC for CH_4 than CO_2 and N_2O indicates that some important natural sources, such as wetland and lakes, have not been included in the simulation. With the support of the National Land Cover Database (NLCD 2001), we will further improve the inventory in order to obtain a better fit of the modeled concentration and the observation.

4. Reference

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Table 1 A summary of correlation coefficient for modeled and observed concentration variation.

	CO_2	CH_4	N_2O
ΔC_{con} **and** ΔC_{obs}	-0.17	-0.40	-0.07
ΔC_{bio} **and** ΔC_{obs}	0.47	0.40	0.13
ΔC_{ant} **and** ΔC_{obs}	-0.04	-0.41	-0.06
ΔC_{all} ***and** ΔC_{obs}	0.53	0.19	0.14

*LC $_{CO2}$=0.35; LC $_{CH4}$=1.20; LC $_{N2O}$=0.35

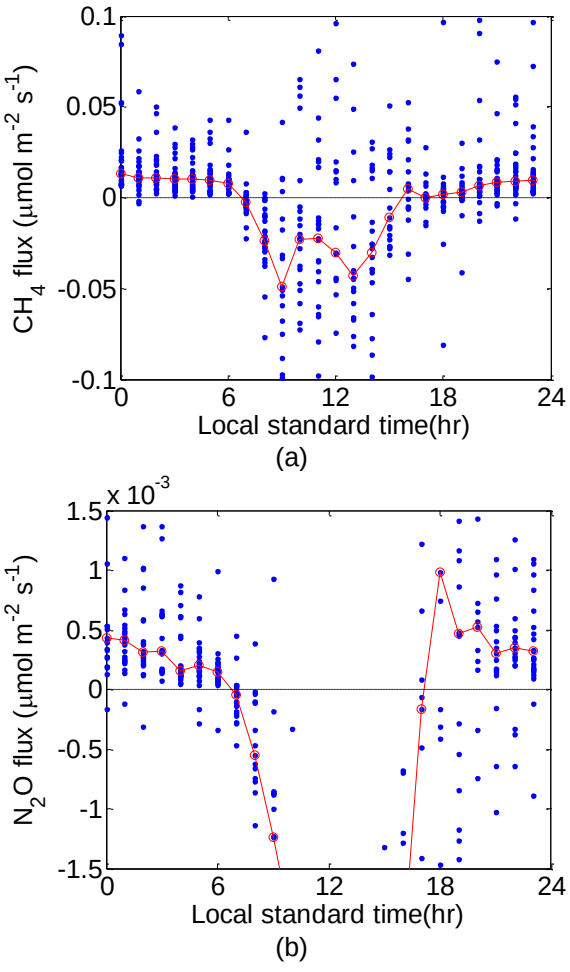

Figure 1: Ensemble diurnal variation of CH_4 (a) and N_2O (b) fluxes calculated from the tall tower gradient measurement, August ~September 2009: blue dot – hourly flux value; red circle – median value for each hour of the day

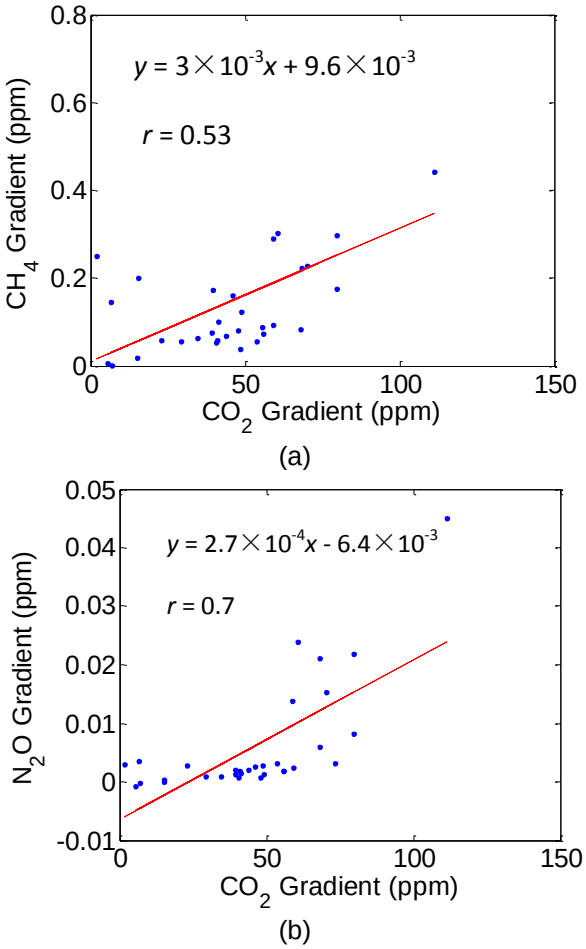

Figure 2: Linear relationship between the night CH_4(a), N_2O(b), and CO_2 gradients, August ~September 2009: blue dot – the block average between 23:00-04:00 LST; red line- linear regression

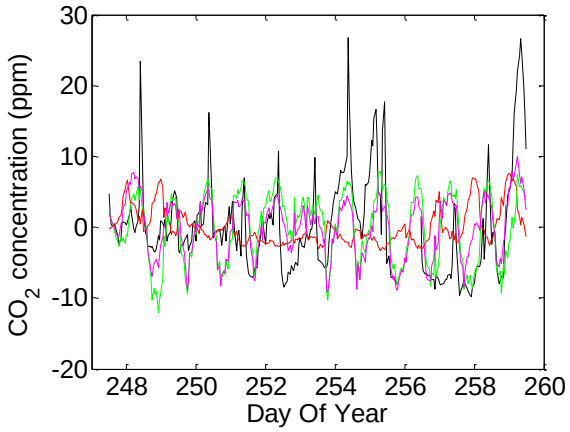

Figure 3: Time series of observed and modeled CO_2 concentration variation. Black line- CO_2 concentration variation from observation; green line- CO_2 concentration variation modeled from biogenic flux; red line- CO_2 concentration variation modeled from EDGAR anthropogenic inventory; magenta line- CO_2 concentration variation modeled from biogenic flux and EDGAR anthropogenic inventory.

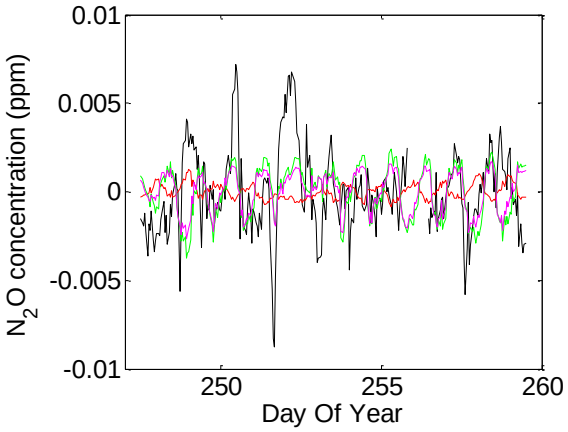

Figure 5: Time series of observed and modeled N_2O concentration variation. Black line- N_2O concentration variation from observation; green line- N_2O concentration variation modeled from biogenic flux; red line- N_2O concentration variation modeled from EDGAR anthropogenic inventory; magenta line- N_2O concentration variation modeled from biogenic flux and EDGAR anthropogenic inventory.

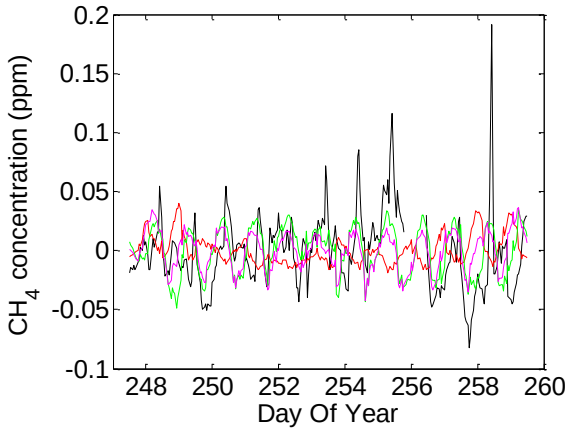

Figure 4: Time series of observed and modeled CH_4 concentration variation. Black line- CH_4 concentration variation from observation; green line- CH_4 concentration variation modeled from biogenic flux; red line- CH_4 concentration variation modeled from EDGAR anthropogenic inventory; magenta line- CH_4 concentration variation modeled from biogenic flux and EDGAR anthropogenic inventory.