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Evaluation of use of EcoCELL technology for quantifying total gaseous mercury fluxes over background substrates

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Abstract

Total gaseous mercury (Hg) fluxes from large ($7.3 \times 5.5 \times 4.5$ m, $L \times W \times D$) climate-controlled gas exchange mesocosms (Ecologically Controlled Enclosed Lysimeter Laboratories or EcoCELLs) containing tallgrass prairie soil–plant monoliths were measured from 2002 to 2005. EcoCELL Hg fluxes (calculated based on the difference in air Hg concentrations inside mesocosms and in incoming air, soil area of the monoliths, and airflow through the system) indicated a net annual emission of 102 $\mu g\,m^{-2}$, while soil Hg fluxes measured simultaneously using a dynamic flux chamber were an order of magnitude lower. Since Hg fluxes measured from empty EcoCELLs in winter and when housing the soil–plant monoliths at the same time of year were similar, we hypothesized that the Hg signal generated by the tallgrass prairie soil–plant monoliths was too low to be detected using the EcoCELL technology. Because mesocosm Hg exchange was correlated with solar radiation and temperature, with the largest emissions occurring at midday and in the summer, we also hypothesized that the flux from mesocosm infrastructure would change over time. Limited by the ongoing experiment, the EcoCELLs were manipulated to test the above hypotheses. When monoliths were completely covered and excluded from the exchange with the surrounding air, mesocosm Hg exchange was unaffected. Furthermore, removal of vegetation at the end of each growing season did not affect mesocosm Hg fluxes. Tests with changing mesocosm airflow also indicated that the signal from the tallgrass prairie monoliths was not being measured. These results suggest that, although EcoCELLs performed well in a study using Hg contaminated soils and have been successfully applied to understand processes controlling Hg fluxes, there are limitations of this technology for quantifying Hg exchange from background substrates. Prior to the use of similar systems the detection limit and Hg exchange from an empty system need to be carefully quantified.

Keywords: Mercury flux; Mesocosms; Chamber blank; Background system

1. Introduction

Landscape level total gaseous mercury (Hg) exchange encompassing canopy, litter, and soil fluxes over large areas (~10–200 m^2) has been assessed using micrometeorological gradient measurements (i.e. Modified Bowen Ratio; Kim et al., 1995; Lindberg et al., 1995; Meyers et al., 1996). Fluxes attributable to individual component surfaces such as soil, plant, and litter are typically measured using dynamic flux or gas exchange chambers (cf. Xiao et al., 1991; Zhang et al.,

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2005). Micrometeorological measurements require certain assumptions that are often hard to meet (homogenous gas flux, horizontally uniform surface), while chambers may alter environmental conditions (light wavelengths and intensity, temperature, air mixing, air-surface boundary layer) and plant physiology (transpiration, photosynthesis), and are limited in scale (surface areas typically $<1\,m^2$). Use of environmentally controlled large gas exchange mesocosms could allow for measurement of Hg fluxes over undisturbed surfaces and under-controlled conditions.

The Ecologically Controlled Enclosed Lysimeter Laboratories (in further text referred to as EcoCELLs or mesocosms) at the Desert Research Institute (Reno, Nevada) are large climate-controlled plant growth chambers ($7.3\times5.5\times4.5\,m$, $L\times W\times D$) that were applied in this study for the measurement of air–ecosystem Hg exchange. These mesocosms have been demonstrated to quantify fluxes of carbon dioxide and water vapor with a resolution similar to that of a leaf-level gas exchange system (for detailed description, operating parameters, calibration and assumptions, see Griffin et al., 1996), and have been used to quantify net ecosystem production, net ecosystem exchange and evapotranspiration on an ecosystem level in several studies (cf. Obrist et al., 2003; Verburg et al., 2004, 2005). The mesocosms have also been used for study of Hg behavior and cycling in a system with enriched substrate (Ericksen et al., 2003; Gustin et al., 2004; Johnson et al., 2003).

This project was an ancillary component of a multi-year interannual climate variability study (2001—still ongoing as of 2006) that focused on effects of an anomalously warm year on ecosystem processes using intact soil–plant monoliths of tallgrass prairie contained in mesocosms (Arnone et al., in preparation; Verburg et al., 2005), and complimentary field experiments (Zhou et al., 2006). Data collected from October 2002 to October 2003 using EcoCELLs suggested that the low-Hg containing tallgrass prairie was a net source of Hg to the atmosphere, annually emitting approximately $60\,\mu g\,Hg\,m^{-2}$ (estimate based on measured mesocosm-level Hg fluxes adjusted for Hg exchange measured from empty EcoCELLs in November 2001 just prior to installing the tallgrass prairie monoliths) (Obrist et al., 2005). This annual Hg flux compares well with data reported in some studies (Lindberg et al., 1992, 1998, 2002; Lindberg and Meyers, 2001), while Hg emissions presented in other studies of background vegetated ecosystems are much lower (Schroeder et al., 2005), or indicate net deposition from the atmosphere (Obrist et al., 2006; Poissant et al., 2005; Zhang et al., 2005).

Because of the timing of experiments, Hg exchange from empty EcoCELLs has not been determined for a variety of environmental conditions, nor has the system been calibrated by any sort of standard addition of a known amount of Hg. The only two assessments of Hg exchange from empty mesocosms were during a three-day period in December 1999 and a 12-day period in November 2001 (not a part of the multi-year study). Both of these were from the time of year when light and temperature are of low intensity. Since Hg fluxes are driven by solar irradiance and air temperature (Bahlmann and Ebinghaus, 2003; Gustin et al., 1997; Gustin et al., 2002; Lindberg et al., 1979; Poissant et al., 1999; Zhang et al., 2001) it is possible that the differential between the air Hg concentration inside mesocosms and in incoming air (ΔC) from an empty EcoCELL would be higher if measured during warmer times of the year. We hypothesized that the available assessments of Hg exchange from empty EcoCELLs could not appropriately be applied as a correction for EcoCELL-level Hg fluxes measured from the substrate of interest year round. Furthermore, mesocosm-level ΔC's measured in 1999 and 2001 when EcoCELLs were empty (Obrist et al., 2005) versus when containing tallgrass prairie monoliths for the same months in 2002–2004 were very similar (see below), and we hypothesized that the Hg signal from the tallgrass prairie monoliths housed inside mesocosms was too small to be resolved relative to that occurring from the mesocosm infrastructure. Constrained by the ongoing experiment in mesocosms, we could not measure the Hg exchange from empty EcoCELLs under a variety of environmental conditions and directly test the outlined hypotheses. This paper reports the results of a series of tests conducted to indirectly determine whether mesocosm Hg exchange measured during sunny and warm conditions was associated with the EcoCELL infrastructure alone or could reflect that of the soil–plant monoliths housed within as well. Namely, the effect of EcoCELLs airflow on calculated mesocosm Hg flux was assessed, and Hg fluxes from mesocosms with substrate of interest excluded from exchange by covering, and before and after vegetation clipping were examined. The results provide important considerations for those involved