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Technical note

A note on elevated total gaseous mercury concentrations downwind from an agriculture field during tilling

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Abstract

Elevated mercury concentrations were measured at the University of Connecticut's mercury forest flux tower during spring agricultural field operations on an adjacent corn field. Concentrations at the tower were elevated, a peak of 7.03 ng m^{-3} over the background concentration of 1.74±0.26 ng m^{-3}, during times when the prevailing wind was from the direction of the corn field and during periods when the soil was disturbed by tilling. Strong deposition to the forest was recorded at the point of measurement when atmospheric mercury concentrations were elevated. The strongest deposition rate was a 1 hour maximum of −4011 ng m^{-2} h^{-1} following the initial peak in atmospheric concentrations, Analyses of the meteorological conditions and mercury content in agricultural soil, manure and the diesel consumed in the tilling operation indicate that the source of the mercury was from the agricultural tilling operations and it was advected over the tower enriching the atmospheric concentrations above the forest canopy leading to deposition. These results indicate that agriculture operations resulting in a disturbed soil surface may be a source of atmospheric mercury originating from the pool of mercury bound in the soil. This represents a previously undocumented source of mercury emissions resulting from anthropogenic activities.
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Keywords: Natural mercury emissions; Mercury flux measurements; Atmospheric deposition; Agriculture emission; Relaxed eddy accumulation

1. Introduction

The current state of knowledge of Hg emissions from various land covers is summarized in the mercury surface interface model (HgSIM), Bash et al. (2004) which predicts emissions from bare ground agriculture fields as a function of surface temperature following Gillis and Miller (2000). Hg fluxes have been measured, or estimated from air concentrations, over forests (Bash and Miller, in press; Lindberg et al., 1998; Lee, 2000), wetlands (Lindberg et al., 2002) grasslands (Cobos et al., 2002) and desert sites (Lindberg et al., 1999). Engle et al. (2001) and Gustin et al. (2004) measured increased mercury emissions in dynamic flux chambers due to soil disturbance, but none have been made during human disturbance events of measurement sites. We also know of no measurements of Hg fluxes from agriculture crop fields during tilling and manure spreading operations.

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We have been routinely measuring Hg fluxes above a hardwood forest for several years (Bash and Miller, in press). During several days in May 2006 our measured concentrations and vertical fluxes at the site were dramatically higher. A subsequent analyses of wind flows during the period demonstrated that the source of the high concentrations of Hg was an agriculture field, adjacent to our forest measurement site, which was being tilled and planted. The purpose of this note is to document and report this event.

1.1. Site description

The 40 m tall micrometeorology instrument tower is in a Red Maple (*Acer Rubrum*) forest on the University of Connecticut research farm in Coventry Connecticut (Lat. 41° 47′ 30″ N, Long. 72° 22′ 29″ W, 162 m in elevation) shown in Fig. 1. The dates of this event, May 1st to May 10th, 2006, were prior to leaf-out of the hardwood forest, therefore the trees were leafless with no gas exchange in the canopy. The tower is located

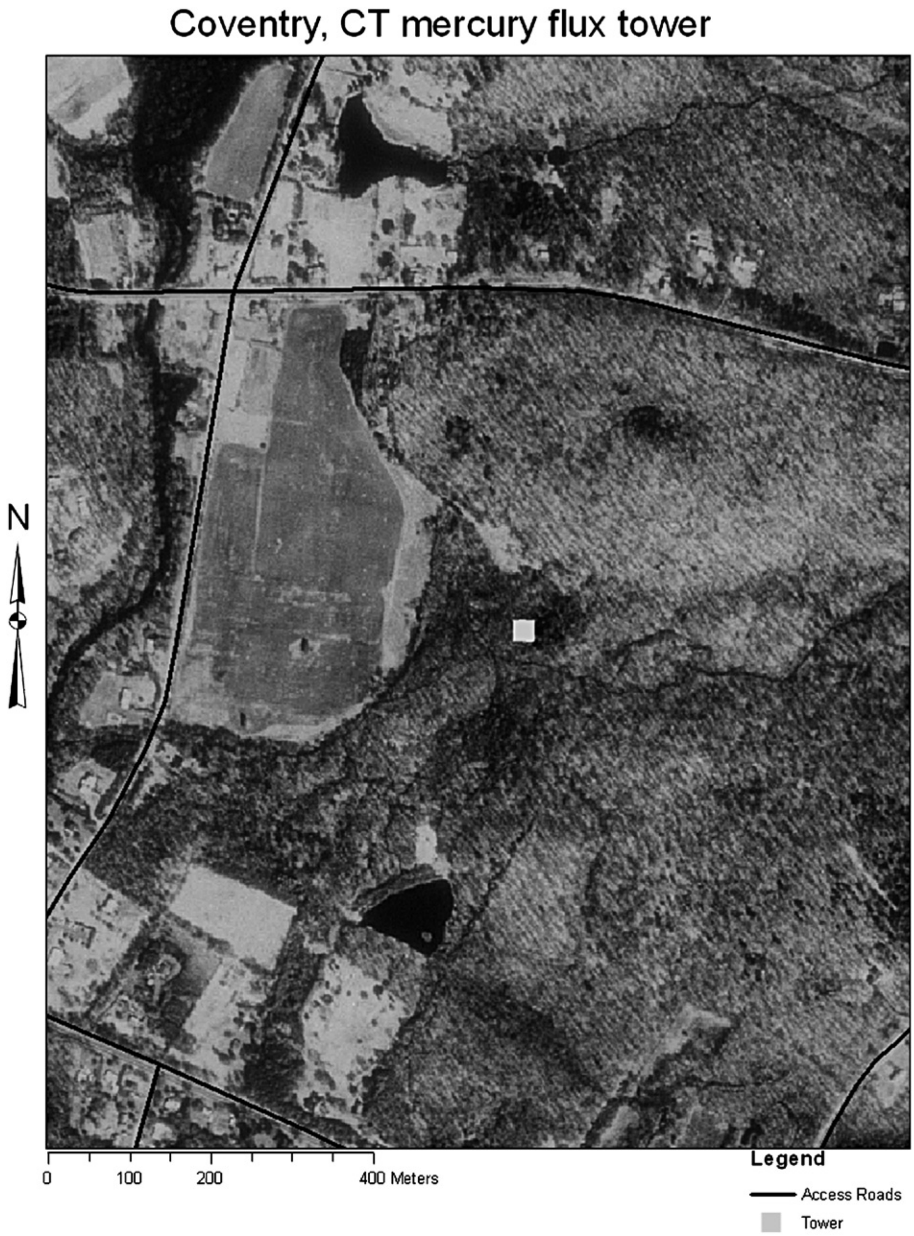

Fig. 1. University of Connecticut experimental forest, micrometeorology tower site and adjacent corn field located west of the tower site.

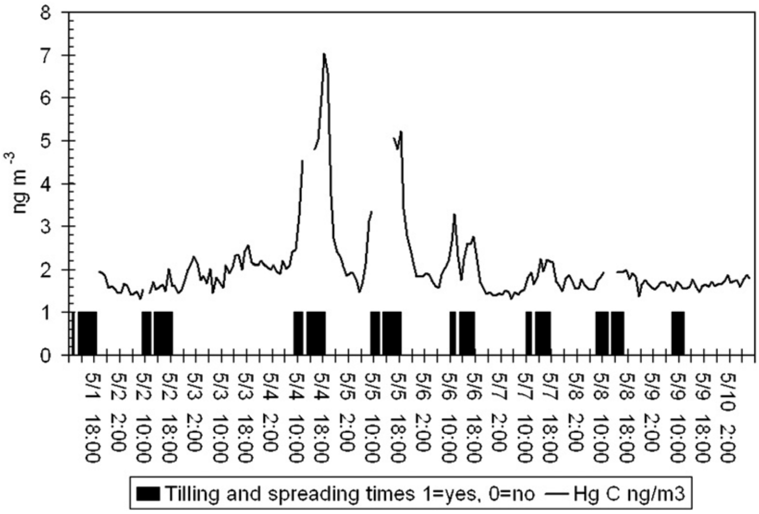

Fig. 2. Mercury concentrations above the downwind forest and periods of agriculture field preparation.

130 m from the edge of the forest, 173 m from the nearest edge of the field and between 334 and 413 m from the tilling operations observed on May 4th. The tower is downwind of the field when the wind direction is westerly, between Southwest and west (225° to 270°). The adjacent agricultural field has been in continuous cultivation for most of the past century and has been continually used by the University of Connecticut to grow silage corn for the past 40 years. Prior to the acquisition of the field by the University of Connecticut, the soil in the field has been amended by ashes from a nearby glass factory which closed before 1900.

1.2. Measurements

Continuous mercury concentration measurements were taken with a Tekran model 2537A mercury analyzer on the tower at 6 m above the 21 m tall forest. Three-dimensional winds and high frequency temperature measurements were made with a Campbell Scientific CSAT3 sonic anemometer at the same location.

Mercury flux measurements were made using the CSAT3 sonic anemometer together with the Tekran model 2537A mercury analyzer for Relaxed Eddy Accumulation (REA) measurements. The REA system is described in detail in Bash and Miller (in press). REA combines fast response vertical anemometry to sense upward and downward air motions, with fast switching of intake air to isolate the air from the upward and downward motions. The mercury vapor carried in the isolated upward and downward moving air is then accumulated in separate sampling lines. The mercury concentrations in the sampling lines are measured with the available slow response instrumentation, in this case the Tekran model 2537A mercury vapor analyzer. Flux is calculated following Businger and Oncley (1990), Eq. (1). The measured fluxes were corrected for density perturbations caused by vapor density fluctuations in the air flow through the thermal mass flow meters used in the system (Webb et al., 1980; Pattey et al. 1992; Lee, 2000).

$$F_{\mathrm{Hg}} = \beta\sigma_w\left(C^{+}_{\mathrm{Hg}} - C^{-}_{\mathrm{Hg}}\right) \tag{1}$$

1.3. Farming operations

Manure spreading in the corn field was conducted on May 1 and 2, 2006. It rained on May 3. Plowing and disking was conducted on May 4, 5 and 6, starting at the south end of the field and moving north. Disking was continued on May 7, 8 and 9.

Twelve tons of dry manure per acre were spread and disked into the soil. The manure was composed of 30% saw dust, 5–8% feed refusal, and 62–65% pig, sheep, cow, and horse manure. The mean mercury concentration of the manure was 6.5 ± 2.9 ng g^{-1} with a mean pH of 7.6 ± 0.39. The mean plowing depth of the soil was approximately 16.5 cm. The mean Hg concentration in the soil in the plow layer was 0.573 ± 0.606 µg g^{-1} ($n=20$) with a pH of 6.04 ± 0.32 ($n=20$) and a total carbon content of $3 \pm 1.8\%$ ($n=4$) Mercury

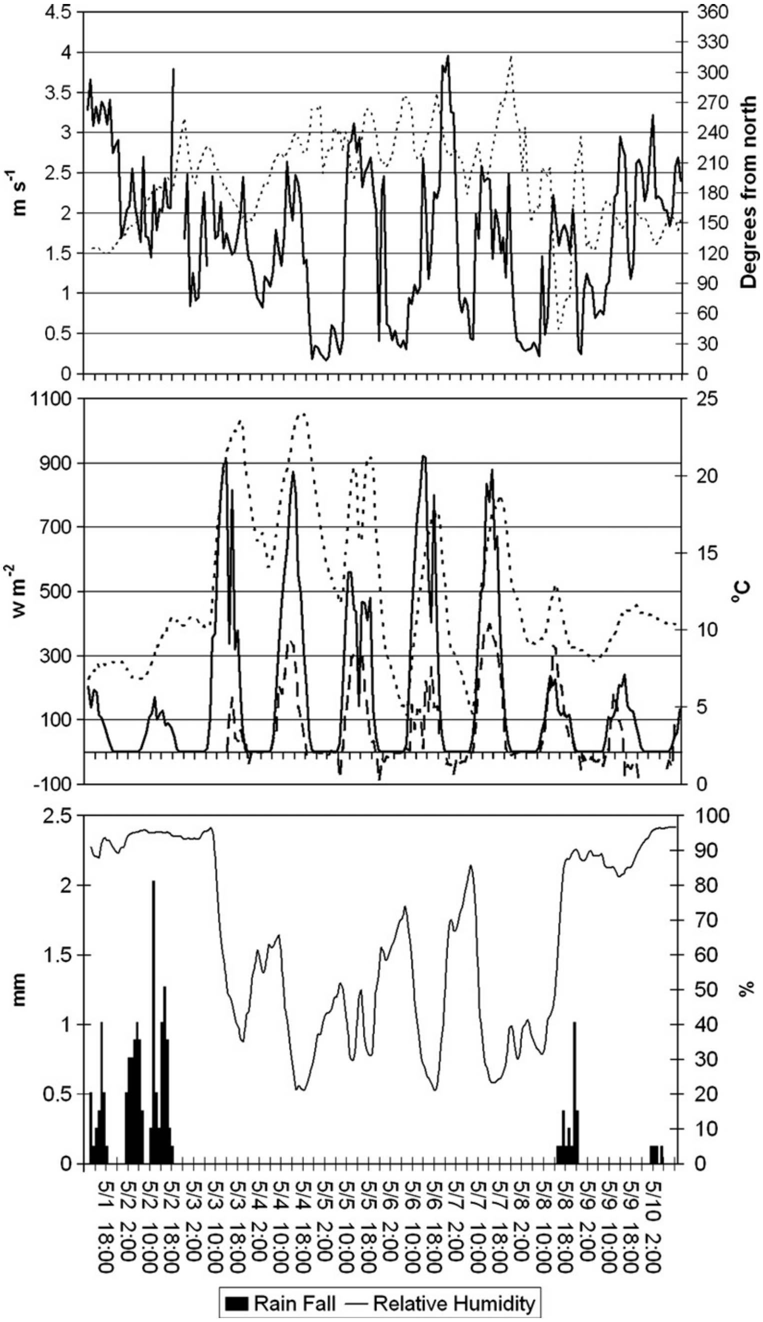

Fig. 3. Hourly meteorological observations during the period of agricultural field preparation, wind speed and wind direction (top), incoming solar radiation, sensible heat flux and ambient temperatures (middle), and precipitation and relative humidity (bottom).

concentrations in the field plow layer have a much higher horizontal variability and little vertical variability when compared to samples taken in the adjacent woods which averaged 0.26±0.01 μg g^{-1}, (n=4).

655 l of diesel fuel were used in the operation with an estimated mercury concentration range of 0.073±0.044 ng g^{-1} (Landis et al., in press) to 0.4 ng g^{-1} (Liang et al., 1996) in the fuel, assuming a density of 0.85 kg l^{-1} (Nelson, 1958). Approximately 199 l of diesel was used in tilling operations while 209 l was used in spreading operations. During manure spreading operations diesel was consumed at an approximate rate

Table 1
Atmospheric conditions at times of tilling

	5/1	5/2	5/3	5/4	5/5	5/6	5/7	5/8	5/9
	Spreading center and NW	Spreading center and NW	None	Tilling SW	Tilling SW and SE	Tilling SE and NW	Tilling NW and NE	Tilling	Tilling
U (m s^{-1})	3.23	2.21	1.73	1.92	2.69	1.62	1.91	1.51	2.26
Udir (deg)	120	185	172	226	232	260	248	103	149
S.D. udir (deg)	29	39	38	32	30	37	41	38	34
Solar radiation (W m^{-2})	134	105	528	544	407	660	591	149	140
Rain (mm)	2.79	6.35	0	0	0	0	0	0.64	0
T (C)	15.38	9.68	11.36	21.70	22.99	19.73	14.82	15.41	11.97
z/L	−0.0699	0.0517	−0.112	−0.349	−0.172	−1.09	−1.34	−0.791	−0.0376

U is wind speed, Udir is wind direction, T is temperature and ζ is the stability parameter, $\zeta = z/L$, where z is height and L is the Obukov length [$L = -(\rho\ c_p\ T\ u_*^3)/(\mathrm{k\ g\ H})$], where ρ is the air density, c_p is the specific heat of air, T is the air temperature, u_* is the friction velocity, k is the von Karman constant (0.4), g is the acceleration due to gravity, and H is the sensible heat flux (Stull, 1988).

of 9.5 l h^{-1}, and during tilling operations diesel was consumed at an approximate rate of 7.6 l h^{-1}.

2. Results

2.1. Hg concentrations and Hg fluxes

The air mercury concentrations observed at the tower increased during the first 3 days of tilling the field but were at background values during manure spreading operations, Fig. 2. Since the field was west of the tower, emissions from the field should only be measured on the tower during periods of westerly wind. On May 1 and 2, during manure spreading, the wind was southerly and the concentrations at the tower were at the background level, Figs. 2 and 3. On May 3 the wind had turned westerly in mid-day but the measured concentrations on the tower remained at background levels, Fig. 3 and there was no activity in the field due to rain the night before. On May 4, 5, and 6 the wind was westerly during plowing and disking of the southwester portion of the field, and very high concentrations of Hg were registered at the tower, Fig. 3. On May 7 the wind was still southwesterly and disking continued on the North

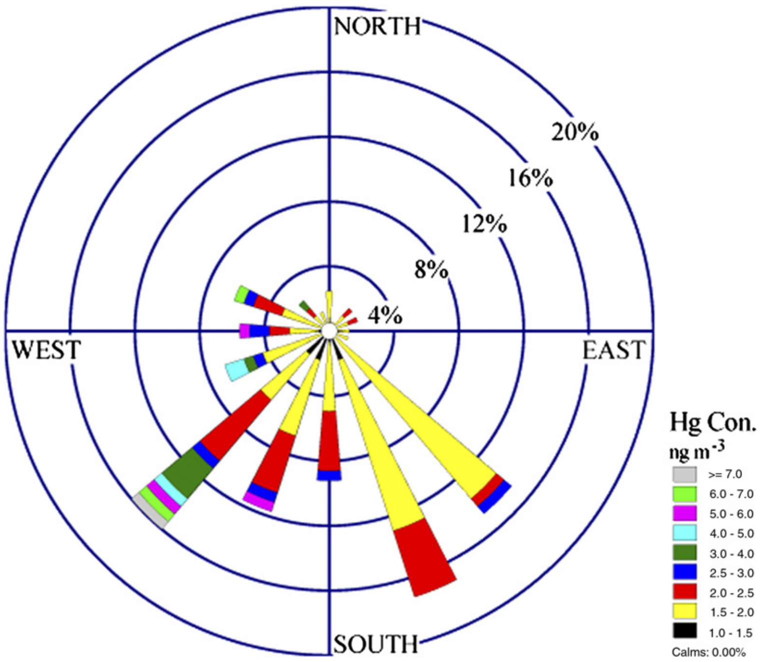

Fig. 4. Mercury atmospheric “concentration rose” during the period of agricultural field operations.

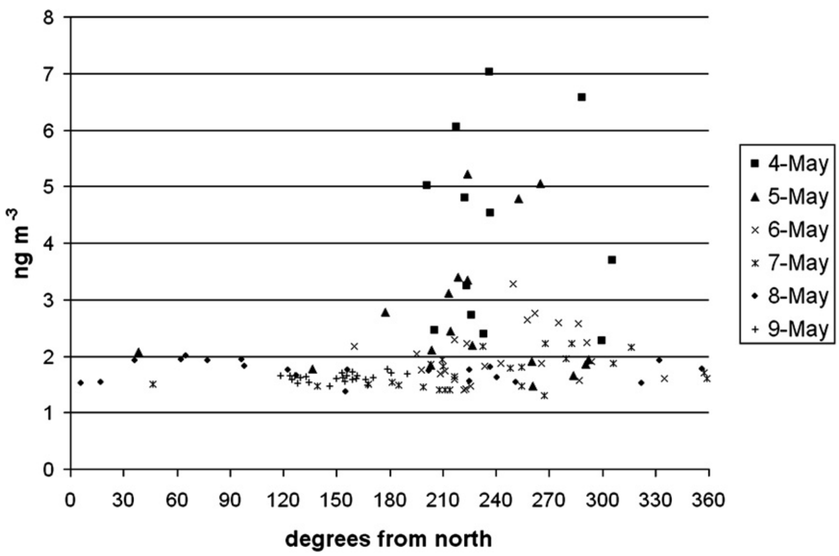

Fig. 5. Hourly average mercury concentrations in the air at the tower during as a function of wind direction during periods of plowing and disking.

portion of the field and the ambient concentrations were slightly elevated at the tower as shown in Figs. 2 and 3. On May 8 and 9, the wind was southerly again, during disking and no increases in Hg concentration can be observed, Figs. 2 and 3. Thus it appears that large increases in gaseous mercury were emitted from the field during the plowing and disking operations, which were recorded at the tower when the southern part of the field was worked due to the prevalent wind direction during those days. On May 7th, when the wind was still southwesterly only small increases in concentration were measured because the north portion of the field was being tilled and the southwest wind advected the mercury that may have been released north of the tower.

Table 1 and Fig. 3 summarize the weather conditions during the times of tilling. In general the daytime periods from May 3rd to May 7th were characterized by low winds, high incoming solar radiation and high heat flux (convection), Fig. 3. These meteorological conditions likely caused the emission plumes from the tilled areas of the field to meander, disperse slowly and rise over the adjacent forests. On the overcast days the wind directions were generally from the south while tilling operations were conducted on the northern portions of the field or were not conducted at all. During the manure spreading operations of May 1st and 2nd there was frequent rainfall, the field was then left to dry on May 3rd before it was tilled from the 4th through the 9th, Fig. 3. The influence of the tilling operations on the adjacent field is clearly seen in a “concentration rose”, Fig. 4. The “concentration rose” shows that all the atmospheric mercury concentrations measured over 3.5 ng m^{-3} were during periods when the mean wind direction was from the adjacent field.

Fig. 5 plots all the 1 hour average mercury concentration measurement as a function of wind direction during times when the field was tilled. Note that wind directions from the southwest show maximum concentrations and wind directions below 200° showed concentrations at the back ground level. The highest peaks in the concentrations were observed during the initial tilling operations. The initial tilling operations also coincided with a two day period of strong solar radiation, high temperatures and southwesterly winds, Table 1. On May 8th, 2006, there was a 2 hour period of southwesterly wind during the first 2 hours of tilling however the northern portion of the field was being tilled.

Vertical fluxes of mercury at the tower measured with the REA are plotted against wind direction, Fig. 6a. Positive fluxes indicate upward diffusion of Hg gas, in other words, evasion to the atmosphere. Negative fluxes mean downward diffusion of Hg or deposition into the forest stand. Then when the wind was from the field toward the forest the fluxes in general were larger in magnitude than the background fluxes to and from the forest floor. The background fluxes showed periods of evasion during the daytimes and little to no flux at night. On days when atmospheric mercury concentrations greater than 3.5 ng m^{-3} were advected above the forest from the field, the mercury flux was typically strongly negative, Figs. 6b and 7. During the peak flux hour on

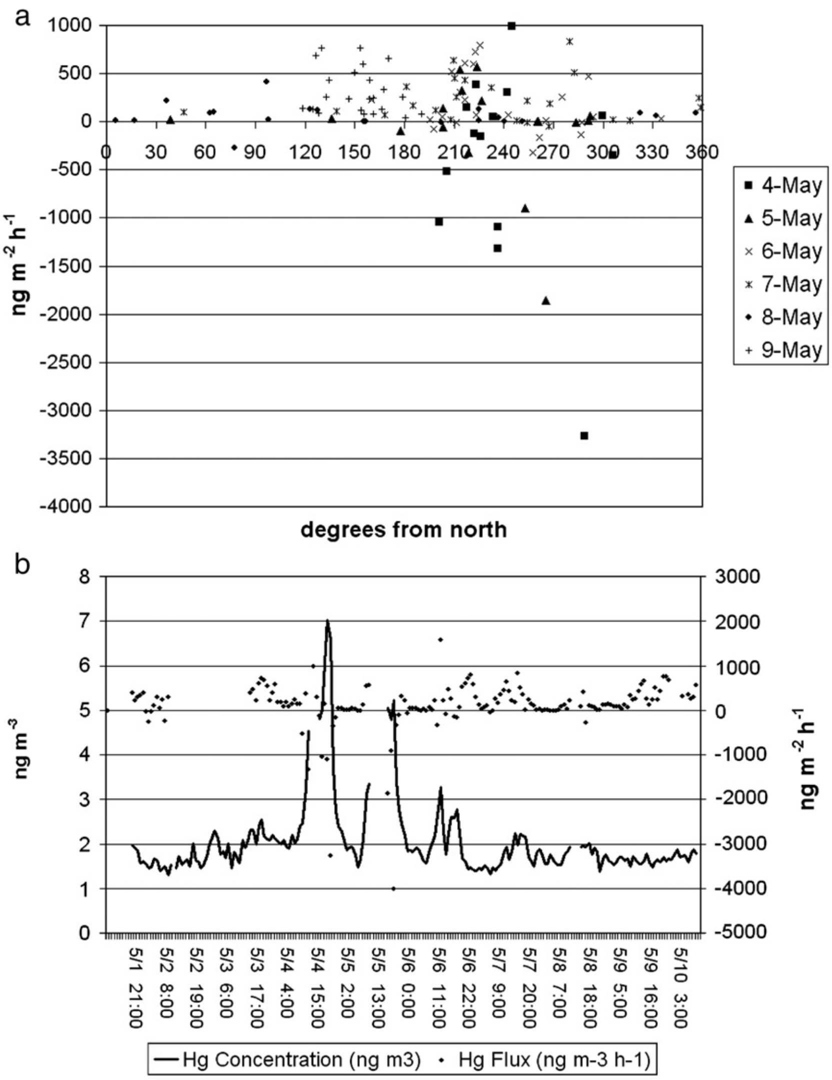

Fig. 6. Hourly mercury fluxes during tilling operations as a function of wind direction (a) and a time series of mercury concentrations and the REA mercury flux at the flux tower during the 10 days of measurements (b).

May 4th, tilling operations were being conducted in the southwestern portion of the field at a distance from 310–440 m at ~250° from north. The maximum hourly mercury deposition to the forest canopy was, −4011 ng m^{-2} h^{-1}, with strong deposition most of the day when the plume advected over the tower location, Fig. 6b. Strong evasion events from the forest were then measured following the initial deposition event of May 4th, Fig. 6. The magnitude of these evasion events may be due to enrichment of the surface media near the flux tower during the previous deposition events; or the mercury vapor on these days could have been advected horizontally through the leafless forest causing higher concentrations below the REA system.

Apparently the amounts of Hg emitted from the field when disturbed by plowing were large enough, when advecting (drifting downwind) over the adjacent forest, to cause a very large concentration gradient between the air (high concentration) and the forest (lower concentration) thus, inducing turbulent deposition of the advected Hg to the forest.

3. Discussion

3.1. The background flux footprint

The sources of the Hg flux at the tower instrument site are a combination of emissions from the surfaces

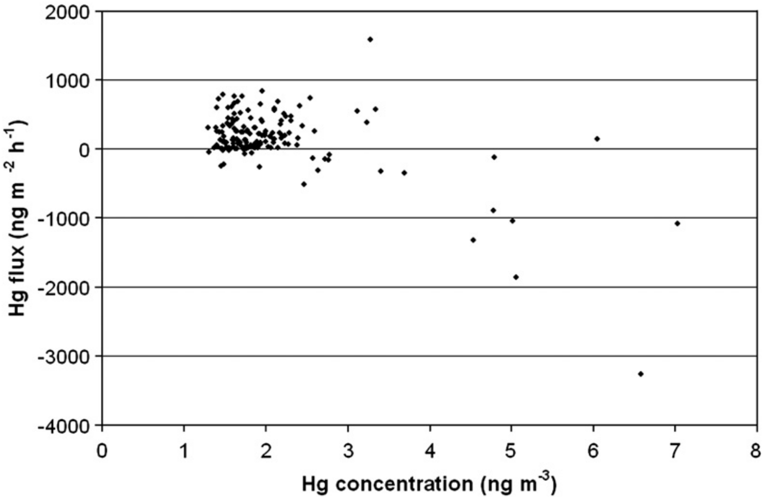

Fig. 7. mercury concentrations (ng m^{-3}) plotted against the REA mercury flux (ng m^{-2} h^{-1}).

upwind of the sensor. An analytic flux footprint model following Amiro (1998) was calculated for the tower site to show the probable sources of the *background* fluxes measured at the tower during the plowing and tilling operations. The footprint is defined as

$$F(x,y,z) = f'(x,z)D_y(x,y) \tag{2}$$

where *F(x,y,z)* is the flux footprint in m^{-2}, x is the streamwise distance, y is the lateral distance, and z is vertical distance. *f′(x,z)* is the vertical crosswind footprint in m^{-1} (Eq. (3)) and $D_y(x,y)$ is the Gaussian dispersion in the lateral direction in units m^{-1}.

The vertical crosswind-integrated footprint model is

$$f'(x,z) = \left(\frac{\Phi}{z_m}\right)\left(\frac{k^2}{\phi_m\left\{\ln\left[\frac{pz}{z_o}\right] - \psi\right\}}\right) \tag{3}$$

where Φ is the normalized crosswind-integrated footprint, z_m is the measurement height, k is von Karman's constant (0.4) and $p \approx 1.55$ following Horst and Weil (1994). The dimensionless wind shear, φ_m, and the diabetic correction factor, ψ, are calculated following Stull (1988) from the sonic anemometer measurements. The normalized crosswind-integrated footprint is calculated following Horst and Weil (1994):

$$\Phi = \left(\frac{z_m}{z}\right)\frac{\bar{u}(z_m)}{\bar{U}(z)}A\exp\left[-\left(\frac{z_m}{bz}\right)^r\right] \tag{4}$$

where $\bar{u}(z_m)$ is the wind speed at the height of the measurements, $\bar{U}(z)$ is the effective speed of wind advection modeled at the mean plume height using the logarithmic wind profile following Stull (1988). A, and b are fit parameters calculated using gamma functions of r following Gryning et al. (1983). r is calculated as a function of stability following Gryning et al. (1983).

During the epochs of the highest ambient enrichment on May 4th, 2006 the wind direction ranged from 205 to 245 degrees from north with a mean standard deviation in the wind direction of 33°. The peak concentration of 7.03 ng m^{-2} h^{-1} occurred at 18:00. During these times the footprint analysis, shown in Fig. 8, predicted that 95% of the background flux originated from the direction of the field, and within approximately 300 m from the flux tower.

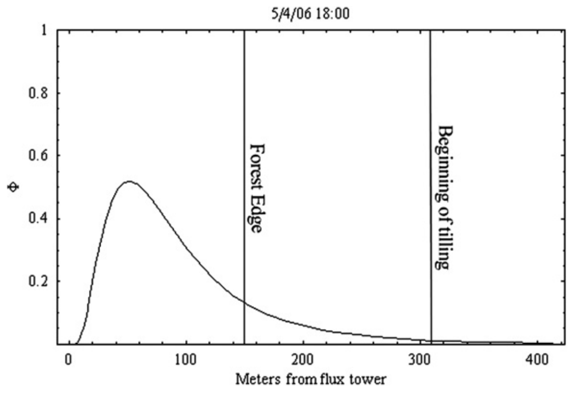

Fig. 8. normalized crosswind-integrated footprint of the flux measured at the flux tower on the 4th of May 2006.

3.2. The disturbance effect

During the tilling operations on the first 2 days with the wind blowing from the agricultural field, a mean TGM deposition rate of −311 ng m^{-2} h^{-1} to the leafless forest canopy and a mean atmospheric TGM concentration of 4.59 ng m^{-3} were measured. The average evasive flux during this period when there was no tilling of the field was 29.4 ng m^{-2} h^{-1} and the mean mercury concentration during the previous 3 days before plowing was 1.74±0.26 ng m^{-3}. These observations of flux direction changing with ambient concentration levels are in line with the range of compensation points in the vegetative mercury flux described by Ericksen and Gustin (2004) and Hanson et al. (1995). We were unable to quantify the mercury emission rates from the field directly from the flux measurements because the REA flux system was too far downstream of the tilling operations.

Frequent rainfall, May 1st, 2nd, 8th, and 10th, and the low pH of the rain (measured at 4.6 for these precipitation events) may have enhanced the mobility of divalent mercury bound in the soil during the tilling operations, Fig. 3. The strong solar radiation and higher ambient temperatures following the rain events and during periods of tilling may have further enhanced the evasion of mercury through photoreduction of divalent mercury bound to soil particles and in the soil water solution. We believe the large emissions during periods of tilling were due to the enhanced exposure of the soil by the disturbance of the soil. Downwind advection of this emitted mercury resulted in elevated concentrations at the flux tower and resulted in strong local deposition events. The bulk of the Hg emitted was stored in the surface soil. An eight hour day of disking at this field used approximately 76 l of diesel. Diesel fuel has a density of approximately 0.85 kg l^{-1} (Nelson, 1958) and a mercury concentration range of approximately 0.073±0.044 ng g^{-1} (Landis et al., in press) to 0.4 ng g^{-1} (Liang et al., 1996). Thus, roughly 4.7±2.8 to 25.84 µg of mercury were emitted during each eight hour day of tilling assuming 100% of the mercury in the diesel was emitted during the combustion process. Diesel engines run using an air to fuel mass ratio ranging from approximately 15:1 to 100:1. These concentrations in the diesel and the amount of diesel burned are not large enough to explain the elevated concentrations measured from the field. The measured soil mercury concentration was 0.573±0.606 µg g^{-1} (n=20) and approximately one hectare a day was tilled to a depth of ~16 cm. This concentration over the plow depth results in a pool of mercury of 109.5 mg m^{-2} in the soil, a much larger potential source than that of the diesel consumed by tilling operations. Furthermore there was no increase in the ambient mercury concentrations during spreading operations, which also used diesel fuel, on May 2nd which had consistent southwesterly wind flow, Figs. 2 and 3. These observations strongly suggest that mercury is emitted from the soil during agricultural tilling operations which presents a large seasonal source of mercury emissions at this site.

It is likely that the emission processes from agricultural tilling operations are similar to those from undisturbed soil, but at a much higher rate due to the disturbance of the soil. Surface temperature (Carpi and Lindberg, 1998), solar radiation (Carpi and Lindberg, 1998; Poissant and Casimir, 1998), and soil moisture (Gillis and Miller, 2000; Gustin and Stamenkovic, 2005; Lindberg et al., 1999) are all environmental factors that effect emissions from undisturbed soils. The much larger magnitudes of the emissions here are likely caused by the mixing and aeration of the soil which releases the Hg gas already in the soil pores and increases the surface area exposed to the atmosphere and the ambient solar radiation which increases the photochemical release of Hg at the surface of the newly exposed soil surface area.

4. Conclusions

A number of studies have shown that Hg from atmospheric deposition and leaf litter fall is bound in the organic layers of the topsoil (Biester et al., 2002; Seehan et al., 2006). We conclude that atmospheric deposition is the most likely source of Hg stored in the agriculture field soil and large amounts evade to the atmosphere when the soil is disturbed. Thus it is possible that a significant evasion event of mercury in the spring season is added to the atmospheric load due to agricultural tilling and planting operations. HgSim (Bash et al., 2004) and other models of natural or anthropogenic emissions do not currently include this source. So potentially a large seasonal source of mercury is unaccounted for in current atmospheric mercury models. In light of this we believe that the contributions of agriculture land to the atmospheric loads of Hg need to be examined with further field experiments and with modeling.

Acknowledgements

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