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# Deforestation and cultivation mobilize mercury from topsoil

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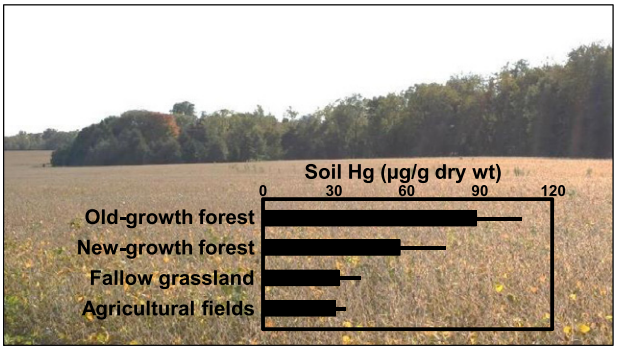
## HIGHLIGHTS

- Deforestation is linked to decreased concentrations of mercury (Hg) in Ohio soil.
- Reforested soils remain depleted in Hg after 60–80 years of new growth.
- Mercury losses from deforested Ohio and Amazon soils are proportional.
- Deforestation may have mobilized 550 Mmol of Hg from soil during the past 200 years.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Terrestrial biomass and soils are a primary global reservoir of mercury (Hg) derived from natural and anthropogenic sources; however, relatively little is known about the fate and stability of Hg in the surface soil reservoir and its susceptibility to change as a result of deforestation and cultivation. In southwest Ohio, we measured Hg concentrations in soils of deciduous old- and new-growth forests, as well as fallow grassland and agricultural soils that had once been forested to examine how, over decadal to century time scales, man-made deforestation and cultivation influence Hg mobility from temperate surface soils. Mercury concentrations in surficial soils were significantly greater in the old-growth than new-growth forest, and both forest soils had greater Hg concentrations than cultivated and fallow fields. Differences in Hg:lead ratios between old-growth forest and agricultural topsoils suggest that about half of the Hg lost from deforested and cultivated Ohio soils may have been volatilized and the other half eroded. The estimated mobilization potential of Hg as a result of deforestation was 4.1 mg $m^{-2}$, which was proportional to mobilization potentials measured at multiple locations in the Amazon relative to concentrations in forested surface soils. Based on this relationship and an estimate of the global average of Hg concentrations in forested soils, we approximate that about 550 Mmol of Hg has been mobilized globally from soil as a result of deforestation during the past two centuries. This estimate is comparable to, if not greater than, the amount of anthropogenic Hg hypothesized by others to have been sequestered by the soil reservoir

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since Industrialization. Our results suggest that deforestation and soil cultivation are significant anthropogenic processes that exacerbate Hg mobilization from soil and its cycling in the environment.



## 1. Introduction

Terrestrial vegetative biomass and soils are a primary reservoir of mercury (Hg) derived from natural and anthropogenic sources (Smith-Downey et al., 2010; Mason et al., 2012; Driscoll et al., 2013). Atmospheric Hg is both wet- and dry-deposited to the Earth's surface (Miller et al., 2005), and absorbed in the gas phase by foliage through stomata (Ericksen et al., 2003; Fay and Gustin, 2007; Bushey et al., 2008). The uptake of gaseous elemental mercury ($Hg^0$) by foliage, as well as leaves functioning as a site of $Hg^0$ oxidation and deposition, results in litterfall being the greatest flux of Hg to soils in deciduous forests (St. Louis et al., 2001; Demers et al., 2007; Risch et al., 2012; Juillerat et al., 2012). Organic-rich surface soils are estimated to contain about 1200 Mmol of Hg globally (Smith-Downey et al., 2010), which accounts for more than a third of the Hg pool actively cycling between the soil–ocean–atmosphere system (Mason et al., 2012). Deforestation may diminish the size of the soil Hg reservoir by removing litterfall, and potentially equally important throughfall (Demers et al., 2007) inputs as well as exacerbating its mobilization from soil, either by erosion (Fostier et al., 2000; Povari et al., 2003; Mainville et al., 2006) or volatilization (Magarelli and Fostier, 2005; Gustin et al., 2006; Kuiken et al., 2008; Briggs and Gustin, 2013; Mazur et al., 2014; Carpi et al., 2014). Net losses of Hg from tropical soils are evidenced by decreased concentrations following deforestation of rainforests (Fostier et al., 2000; Lacerda et al., 2004; Almeida et al., 2005; Mainville et al., 2006; Comte et al., 2013). However, the fate and stability of Hg in temperate surface soils and its susceptibility to change as a result of man-made deforestation and cultivations are largely unknown (Carpi et al., 2014; Mazur et al., 2014).

Mercury deposited to soils is thought to be relatively immobile as a result of its high affinity for organic matter (Meili, 1991; Yin et al., 1996; Hintelmann et al., 2002; Demers et al., 2007; Mauclair et al., 2008; Obrist et al., 2011). Concentrations of Hg are often correlated strongly with the organic content of soils and sediments (Johansson et al., 1991; Biester et al., 2002; Hammerschmidt et al., 2004; Obrist et al., 2011; Juillerat et al., 2012; Richardson et al., 2013; Šípková et al., 2014). Consequently, Hg tends to accumulate in the litter and upper soil horizons that often contain more organic matter than deeper horizons (Biester et al., 2002; Demers et al., 2007; Smith-Downey et al., 2010; Obrist et al., 2011). However, changes to the organic content of soil may affect its capacity to retain Hg. Soil disturbances, such as cultivation, can significantly reduce the organic content of soil (Johnson, 1992; Robertson et al., 1993; Compton and Boone, 2000; Mainville et al., 2006); new-growth forests and cultivated soils can contain about 15% and 30–50%, respectively, less organic matter than old-growth forest soils (Compton and Boone, 2000; Flinn and Marks, 2007). Soil organic content can remain at reduced levels for at least 50 years after cultivation has ceased and natural plant growth is restored (Johnson, 1992; Koerner et al., 1997; Compton et al., 1998; Compton and Boone, 2000).

Given the significance of the soil reservoir in the global Hg cycle and its potential susceptibility to changes in Hg storage capacity as a result of changes in land use, we examined the Hg content of soils from old- and new-growth forest stands, as well as fallow grassland and agricultural fields in southwest Ohio, USA. This region was mostly deciduous forest prior to European settlement in the late 18th century, but most of the land has been deforested and put into cultivation during the past 200 years (Hendricksen, 1933). Our hypothesis was that deforestation and cultivation would exacerbate Hg loss from these temperate soils and, by extension, forested soils would have greater Hg concentrations than nearby cultivated fields. We also examined soil concentrations of lead (Pb), another chalcophilic metal, from which we inferred differences in the transport of Hg.

## 2. Materials and methods

### 2.1. Study sites

The effect of deforestation and cultivation on Hg in soil was examined by comparing concentrations among old-growth forest (>200 year old stand), new-growth forest (60–80 years old), agricultural fields, and fallow areas either on or near the campus of Wright State University (WSU), in Greene County, southwest Ohio, USA (Fig. 1). The area has a mean annual temperature of 11.1 °C and receives, on average, 1.04 m of precipitation annually. Atmospheric deposition of Hg is presumed to be homogeneous throughout the relatively small study area (110 $km^2$; http://nadp.sws.uiuc.edu/mdn/), which is distant from large combustion sources (Konkler and Hammerschmidt, 2012), although Hg deposition to soils in forested areas can be enhanced by through- and litter-fall fluxes (St. Louis et al., 2001; Demers et al., 2007). All sampling locations were more than 30 m from roadways to minimize potentially anomalous loadings of Pb from automobile emissions (Rodríguez-Flores and Rodríguez-Castellón, 1982; Nabulo et al., 2006).

The WSU Campus Woods (0.8 $km^2$; Fig. 1), a temperate deciduous forest, was an ideal location to examine soil Hg concentrations related to stages of forest growth, because it has well-defined stands of new and old growth, which have been identified by vegetation species, diversity, and aerial photography (Runkle et al., 2005). The Woods are undisturbed natural habitat, other than a few walking trails. Tree species in

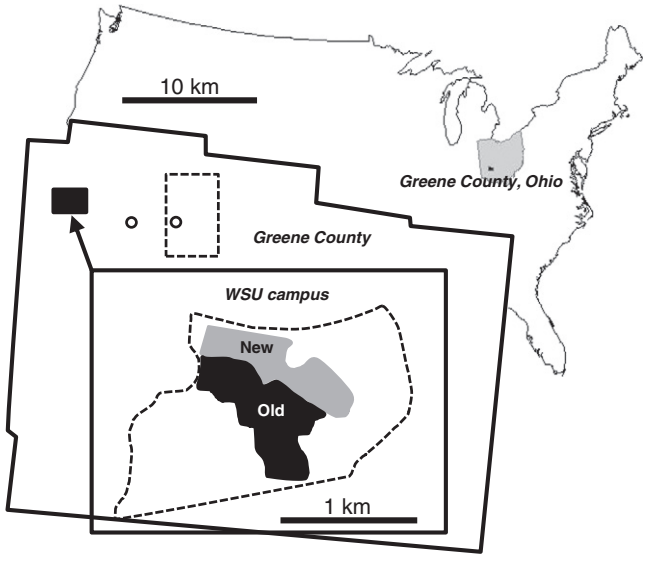


**Fig. 1.** Soil sampling locations in Greene County, Ohio, USA. Sampling sites were in cultivated agricultural fields (open dashed box), fallow grassland (open circles), and old- and new-growth forest on the campus of Wright State University (WSU, filled rectangle). The inset shows the outline of the WSU campus (dashed line) and size and proximity of the old- and new-growth forest stands on campus. The 10-km scale is for Greene County and 1-km scale is for the campus inset.