Atmospheric Environment 47 (2012) 554–559

Contents lists available at SciVerse ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Short communication

Examination of mercury inputs by throughfall and litterfall in the Great Smoky Mountains National Park

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ARTICLE INFO

Article history:
Received 26 April 2011
Received in revised form
30 September 2011
Accepted 10 October 2011

Keywords:
Total mercury deposition
Throughfall
Litterfall
Mercury deposition

ABSTRACT

Throughfall and litterfall represent two major pathways which comprise the bulk of the mercury that reaches the forest floor. In an effort to quantify total mercury deposition to forests, throughfall and litterfall samples were collected at three elevations in the Great Smoky Mountains National Park (GSMNP) between April and November in 2008 and 2009. The mean (±standard error) volume-weighted throughfall total mercury concentration across all three sites ranged from 13.62 ng L^{-1} (±0.86) to 18.23 ng L^{-1} (±0.77) with a slight trend of higher throughfall mercury concentration at the high elevation spruce-fir sites relative to the low elevation mixed-hardwood site. The mean (±standard error) throughfall total mercury deposition across all sites ranged from 2.07 μg m^{-2} (±0.09) to 4.09 μg m^{-2} (±0.38). Mean (±standard error) mass-weighted litterfall concentration ranged from 37.2 ng g^{-1} (±1.36) to 62.87 ng g^{-1} (±1.94) and statistically higher ($p = 0.02$, $p < 0.05$) mean concentrations were observed at the higher elevation sites. The mean (±standard error) litterfall mercury deposition across all three sites ranged from 10.34 μg m^{-2} (±0.69) to 29.30 μg m^{-2} (±0.44). Although, no statistically ($p = 0.05$, $p < 0.05$) significant difference was observed between the sites for either throughfall or litterfall deposition, both increased from 2008 to 2009 at the lower elevation site. Mean litterfall deposition (17.93 μg m^{-2}) dominated Hg input for the two years studied over mean throughfall deposition (3.20 μg m^{-2}). Meanwhile, the mean throughfall concentration (17.8 ng L^{-1}) dominated the mean open area precipitation input (6.03 ng L^{-1}) at the high elevation site, suggesting evidence for a dry deposition component from the forest canopy. While the study was limited in scope, the results do provide some basic data that adds to understanding Hg inputs to the terrestrial ecosystem in general and in the GSMNP specifically.

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1. Introduction

Atmospheric mercury (Hg) deposition is an area of interest due to human health and safety concerns from the dietary consumption of fish and seafood, and associated bioaccumulation of mercury in aquatic ecosystems. Understanding the input of mercury into waterbodies via runoff from terrestrial ecosystems is complex. Therefore, an understanding of how mercury is deposited and internal cycling and dynamics of mercury availability are important for a full understanding and quantifying of mercury biogeodynamics. Atmospheric mercury may exist as gaseous elemental mercury (Hg (0)); oxidized gaseous mercury (HgII also known as reactive gaseous mercury); and particulate-bound mercury (Hg_p) (Lindberg et al., 2007). Terrestrial ecosystems are exposed to atmospheric pollutants through wet (precipitation) and dry deposition, litterfall (deposition of foliage), throughfall (water passing through the canopy onto the soils below), and cloud deposition at higher elevations. Once atmospheric mercury is deposited onto the land and surface waters, complex changes and cycling throughout the ecosystem may occur. The fate and mobility of atmospherically deposited mercury to terrestrial environments is poorly understood. Preliminary results from the METAALICUS (Mercury Experiment To Assess Atmospheric Loading in Canada and the US) study suggest that terrestrial systems impose a time lag for the delivery of atmospheric deposition to lakes via runoff (Hintelmann et al., 2002). The presence of organic matter in soils influences watershed mercury cycling, as mercury has been shown to form strong complexes with organic matter (Grigal, 2002; Gabriel and Williamson, 2004). Several studies have demonstrated foliar exchange of gaseous Hg (0) through leaf stomata (Hanson et al., 1997; Lindberg et al., 1998) and subsequent loading to the forest floor via litterfall.

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1352-2310/$ – see front matter Published by Elsevier Ltd.
doi:10.1016/j.atmosenv.2011.10.017

Throughfall–which is precipitation that reaches the forest floor after interacting with the forest canopy–and litterfall are two pathways through which the bulk of the Hg reaches the forest floor (Rea et al., 1996; Grigal, 2002; St. Louis et al., 2001). Numerous studies have examined Hg input in forest ecosystems (Demurs et al., 2007; Choi et al., 2008; Bushey et al., 2008). Much of the information concerning Hg deposition in forest ecosystems has been learned from low elevation sites, and in North America from mostly northern latitude forests, although some studies have been done on low elevation southern forests (Lindberg et al., 1994). Few published studies have focused on Hg deposition in high elevation sites and it is thought that high elevation sites in the Great Smoky Mountains National Park could potentially receive more deposition than the lower elevation sites due to cloud deposition (MACTEC, 2010). In addition, studies have also demonstrated that differences in vegetation/canopy type may influence the capture of deposited mercury, where conifer needles tend to high higher concentrations of mercury over deciduous trees (Lindberg, 1996; Lindberg et al., 1994; Kolka et al., 1999; Demurs et al., 2007; Witt et al., 2009).

Total Hg in wet deposition is measured weekly across the United States as part of the Mercury Deposition Network (MDN, http://nadp.sws.uiuc.edu/) providing data useful for examining spatial and temporal trends of Hg as it enters terrestrial system as precipitation. Dry deposition of gaseous Hg (II) to foliage and the capture and oxidation of Hg (0) by the forest canopy are sources of Hg in throughfall (Lindberg et al., 1994). Supporting this are data showing total mercury concentrations to be higher in throughfall than in precipitation in several studies (Kolka et al., 2001; Grigal, 2002 references therein; Driscoll et al., 1994; Rea et al., 1996). Forest canopy characteristics have also been shown to influence the volume and deposition of Hg as foliage surface areas differ among species (Kolka et al., 1999; Demurs et al., 2007; Witt et al., 2009; Lindberg, 1996). Given the sources of Hg in throughfall and the capacity of Hg to bind to organic matter, Hg concentrations in litterfall also represent a pathway of Hg to the forest watershed and to soils (Grigal, 2002). In addition, total Hg deposition in litterfall has been measured as being greater than in throughfall or open site precipitation (Grigal, 2002; Kahl et al., 2007; Bushey et al., 2008; Demurs et al., 2007; Rea et al., 1996).

To determine the magnitude of Hg inputs to the forest ecosystem, throughfall and litterfall samples were collected at three elevations in the Great Smoky Mountains National Park (GSMNP) between April and November in 2008 and 2009 to examine deposition inputs that may influence internal Hg cycling dynamics and the contribution of seasonal and elevation differences in total Hg deposition.

2. Methods

2.1. Study site

The study of Hg inputs via throughfall (TF) and litterfall (LF) was carried out in the Great Smoky Mountains National Park (GSMNP) at three sites (Fig. 1); two high elevation sites - Clingmans Dome (CD) at 2024 m and Noland Divide (ND) at 1700 m and a low elevation site - Noland Creek (NC) at 592 m. Great Smoky Mountains National Park is located in eastern Tennessee and western North Carolina and covers 231,075 ha. The forest canopy at the CD site is composed of primarily red spruce (*Picea rubens* Sarg.) trees $<$ 30 cm diameter at breast height (dbh), with some Fraser fir (*Abies fraseri* (Pursh) Poir.). Overall canopy height was 6–8 m. The canopy at the ND site is composed of primarily large (dbh $>$ 60 cm) dominant red spruce with a minor understory hardwood component of yellow birch (*Betula lutea* Mixchx. F.), American ash (*Sorbus americana*), and striped maple (*Acer pennsylvanicum*). The average canopy height of the red spruce at ND was approximately 20–25 m. The NC site is dominantly a mixed-deciduous forest type composed primarily of oaks (*Quercus* sp.) and tulip poplar (*Liriodendron tulipifera*). Canopy height at the NC was estimated at 15–20 m.

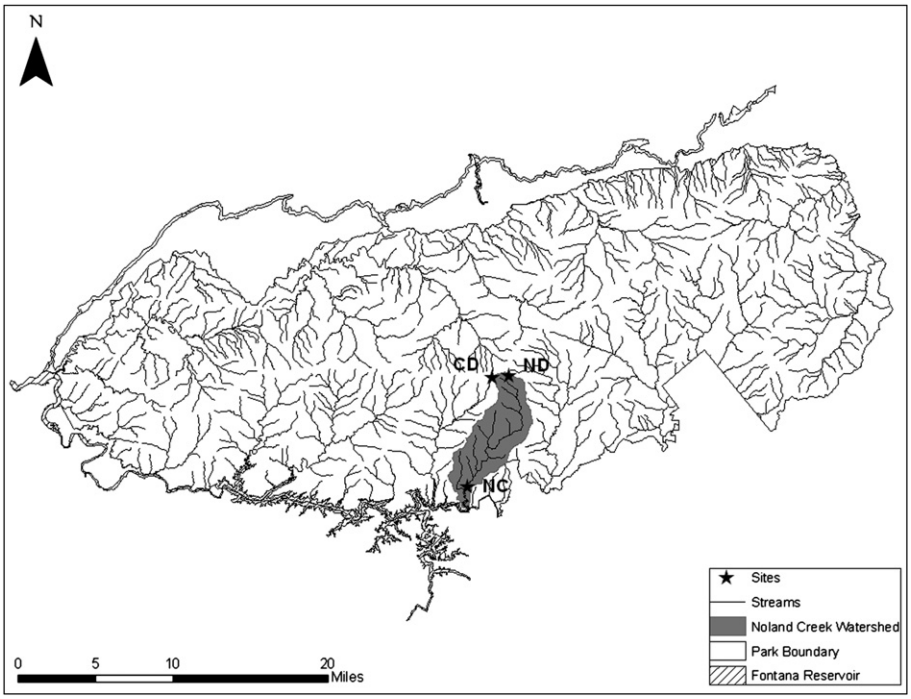

Fig. 1. Study site locations for throughfall and litterfall measurements of total mercury in Great Smoky Mountains National Park.