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Endosulfan wet deposition in Southern Florida (USA)

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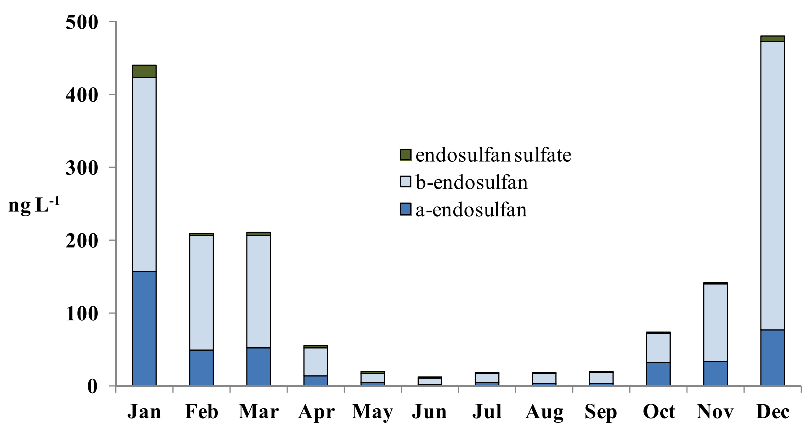
HIGHLIGHTS

- Endosulfan measured in rain for 4 years at three sites in Southern Florida
- High concentrations and wet deposition rates found within an area of high use
- Everglades and Biscayne National Parks border use areas
- Atmospheric transport to and wet deposition within these parks observed
- Potential for adverse ecological impact was indicated.

GRAPHICAL ABSTRACT



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ABSTRACT

The atmosphere is an important transport route for semi-volatile pesticides like endosulfan. Deposition, which depends on physical–chemical properties, use patterns, and climatic conditions, can occur at local, regional, and global scales. Adverse human and ecological impact may result. We measured endosulfan wet deposition in precipitation over a 4-year period within an area of high agricultural use in Southern Florida (USA) and in nearby Biscayne and Everglades National Parks. Endosulfan's two isomers and degradate, endosulfan sulfate, were detected at high frequency with the order of detection and concentration being β-endosulfan > α-endosulfan > endosulfan sulfate. Within the agricultural area, detection frequency (55 to 98%) mean concentrations (5 to 87 ng L^{-1}) and total daily deposition (200 ng m^{-2} day^{-1}) exceeded values at other sites by 5 to 30-fold. Strong seasonal trends were also observed with values at all monitored sites significantly higher during peak endosulfan use periods when vegetable crops were produced. Relatively high deposition in the crop production area and observations that concentrations exceeded aquatic life toxicity thresholds at all sites indicated that endosulfan volatilization and wet deposition are of ecotoxicological concern to the region. This study emphasizes the need to include localized volatilization and deposition of endosulfan and other semi-volatile pesticides in risk assessments in Southern Florida and other areas with similar climatic and crop production profiles.
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1. Introduction

After more than six decades of intensive use, endosulfan (6,7,8,9,10,10-hexachloro-1,5,5a,6,9,9a-hexahydro-6,9-methano-2,4,3-benzodioxathiepin-3-oxide) continues to be an effective insecticide. The technical product is a mixture of two diastereoisomers known as α-endosulfan (or I) and β-endosulfan (or II) with α- and β-isomers typically in a 70:30 ratio, respectively. Endosulfan is valued for control of many crop damaging pests, potential for use in rotation with other insecticides for resistance management and for its relatively low cost and toxicity to pollinators (U.S.EPA, 2002; Janssen, 2011). Notably, neonicotinoid insecticides that are increasingly used in place of endosulfan are highly toxic to bees and have been implicated in the decline of bee populations in the USA and Europe (EFSA, European Food Safety Authority, 2013).

Recent risk assessments have indicated that likely exposures to both endosulfan isomers and endosulfan sulfate, through normal use in crop production, present unacceptably high risks to farm workers and wildlife (UNEP-POPRC, 2009; U.S.EPA, 2010a). In most studies, the three forms are considered to be equivalent in terms of potential toxic impact and are summed to evaluate exposure risk (Weber et al., 2010). This sum is identified as "total endosulfans" in discussions that follow. Other concerns linked to exposure potential are endosulfan atmospheric transport, persistence of the parent isomers and degradate endosulfan sulfate in soil, water, and sediment, and bioaccumulation (UNEP-POPRC, 2009). These issues led to the classification of endosulfan as a persistent organic pollutant by the Stockholm Convention on Persistent Organic Pollutants and to plans for phased termination of all uses globally (UNEP-POP, 2011). The Convention agreement terminated most endosulfan uses in 2012. India was granted an exemption for use of 14 crops until 2017. Continued use of selected crops in China, at least until mid-2016, is also anticipated (U.S.EPA, 2013).

In the USA, an earlier agreement defined plans to terminate all endosulfan uses by the end of 2016 and within the highest use state, Florida, in 2014 (U.S.EPA, 2010a). USDA data collected in, 2011 indicated that endosulfan use in Florida accounted for more than 50% of all uses nationwide. A very high use area is around the city of Homestead in the southern-most part of the state (Fig. 1). An estimated six to eleven metric tons are applied annually to the approximately 14,000 ha in vegetable and nursery crop production in Homestead's agricultural area (HAA) (Stone, 2013). HAA is surrounded by Everglades National Park (EVR) on the west and south, Biscayne National Park (BNP) on the east, and the urbanized area of southern Miami on the north (Fig. 1). Environmental monitoring within HAA over the past two decades has documented the presence of endosulfan residues in surface water, sediment, and biota (Fulton et al., 2004; Harman-Fetcho et al., 2005; Pfeuffer, 2011; U.S.EPA, 2010a). Data have indicated potential for adverse ecological impact to freshwater and marine ecosystems (Fulton et al., 2004; Rand et al., 2010; Scott et al., 2002).

A principal concern is endosulfan's high toxicity to aquatic organisms. Water quality criteria established by U.S. EPA (2012) for protection of the most sensitive freshwater indicator species are 0.220 and 0.056 ug L^{-1} for acute and chronic exposures respectively. The corresponding criteria for marine species are 0.034 and 0.0087 ug L^{-1}. Bioaccumulation has also been observed. Endosulfan and the toxic degradate, endosulfan sulfate, were detected in tissues of small demersal fish in Southern Florida at levels that may adversely impact wading birds and other organisms that use these fish as food sources (Rand et al., 2010). Since residues were found in fish captured in areas that were not adjacent to land used for crop production, a link to endosulfan atmospheric transport and deposition was indicated.

These findings motivated our effort to evaluate atmospheric transport and wet deposition of endosulfan's two isomers and endosulfan sulfate within the HAA and the National Parks. A summary of ground

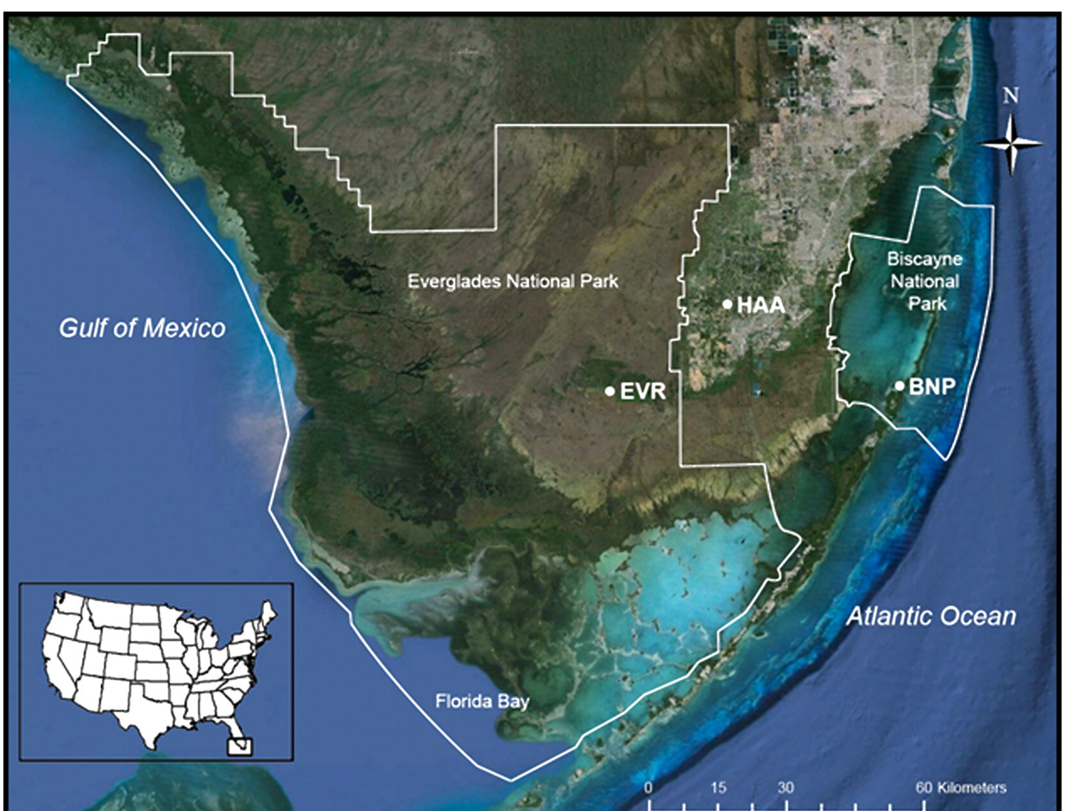

Fig. 1. National Park boundaries and locations of wet-deposition collectors in the Homestead Agricultural Area (HAA), Everglades National Park (EVR), and Biscayne National Park (BNP) (Google Earth, 2013).