

# Release of Airborne Polychlorinated Biphenyls from New Bedford Harbor Results in Elevated Concentrations in the Surrounding Air

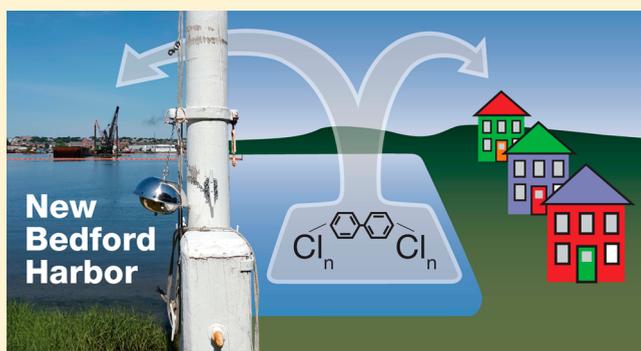
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**S** Supporting Information

**ABSTRACT:** Qualitatively and quantitatively, we have demonstrated that airborne polychlorinated biphenyl (PCB) concentrations in the air surrounding New Bedford Harbor (NBH) are caused by its water PCB emissions. We measured airborne PCBs at 18 homes and businesses near NBH in 2015, with values ranging from 0.4 to 38 ng m<sup>-3</sup>, with a very strong Aroclor 1242/1016 signal that is most pronounced closest to the harbor and reproducible over three sampling rounds. Using U.S. Environmental Protection Agency (U.S. EPA) water PCB data from 2015 and local meteorology, we predicted gas-phase fluxes of PCBs from 160 to 1200 μg m<sup>-2</sup> day<sup>-1</sup>. Fluxes were used as emissions for AERMOD, a widely applied U.S. EPA atmospheric dispersion model, to predict airborne PCB concentrations. The AERMOD predictions were within a factor of 2 of the field measurements. PCB emission from NBH (110 kg year<sup>-1</sup>, average 2015) is the largest reported source of airborne PCBs from natural waters in North America, and the source of high ambient air PCB concentrations in locations close to NBH. It is likely that NBH has been an important source of airborne PCBs since it was contaminated with Aroclors more than 60 years ago.



## INTRODUCTION

Although New Bedford Harbor (NBH), MA, is one of the largest polychlorinated biphenyl (PCB) Superfund sites in the United States,<sup>1</sup> it has not been studied as an important source of airborne PCBs. In 1983, NBH was placed on the National Priorities List of Superfund cleanup sites because of the extremely high levels of PCBs in the sediments. Aroclors 1242 and 1016 were discharged into the harbor for more than 30 years (1940s–1970s).<sup>2–4</sup> PCB concentrations of ≤1 μg g<sup>-1</sup> in flounder and ≤10000 μg g<sup>-1</sup> (dry weight) in sediments have been reported.<sup>2,3,5</sup> These high PCB levels motivated release of seafood consumption advisories starting in 1979<sup>5</sup> and sediment dredging since 1994.<sup>2</sup>

Concentrations of PCBs in air have been measured and reported by the U.S. Environmental Protection Agency (U.S. EPA) since 1999,<sup>6,7</sup> and although concentrations are elevated, the magnitude of the harbor as an emission source is unclear and the potential risk by inhalation caused by emissions is unknown. While remediation is driven by PCB levels in the sediments, PCBs are mobilized from sediment to overlying water and air,<sup>8–10</sup> contributing to human exposure via inhalation. It is important to understand the specific contribution of NBH to local levels of airborne PCBs as part of the risk-based decision making regarding remediation.

We recently developed a strategy for predicting concentrations of airborne PCBs as a function of emissions from contaminated water.<sup>11</sup> We measured PCBs in water, calculated the emissions as gross volatilization flux, and predicted dispersion into the surrounding region. In northwest Indiana, we found that PCBs released from the Indiana Harbor and Ship Canal (IHSC) accounted for 15% of the observed PCB concentrations in the adjoining neighborhoods. The community surrounding the IHSC is one of the most industrially dense regions in the United States and has a long history of environmental contamination. Using this two-pronged strategy of calculated emissions and atmospheric dispersion modeling, we concluded that there were many sources of airborne PCBs in this region in addition to IHSC.<sup>11</sup>

PCB concentrations in NBH water are at least 10 times higher than in IHSC, and its area is 3 times larger than that of IHSC, which could dramatically increase the contribution of airborne gas-phase PCBs to the local atmosphere. Therefore, we hypothesized that PCB emissions from NBH explain

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ambient air concentrations of PCBs in the surrounding communities.

To address our hypothesis, in response to and in collaboration with community and environmental organizations, we launched a field effort in 2015 to evaluate the effect of emissions from NBH on airborne PCBs. We calculated congener-specific PCB emissions from NBH as a function of reported water concentrations, chemical properties, and local meteorology. We used an atmospheric dispersion model to predict gas-phase PCB concentrations in the region surrounding NBH. In addition, we compared our predictions to measured values from our own samplers and those measured by the U.S. EPA. Lastly, we examined the long-term trends in PCB emissions and air concentrations for 2006–2015 using historical data reported by the U.S. EPA.

## MATERIALS AND METHODS

**Airborne PCB Concentration Measurements.** Airborne PCBs were measured using polyurethane foam passive air samplers (PUF-PAS) as previously described.<sup>12–15</sup> The PUF-PAS collects both gas and particle phases, but because PCBs are mostly in the gas phase (~90%), the values reported here are assumed to be the gas phase. Samplers were placed at the same 18 locations for three consecutive periods, from July to November of 2015 in New Bedford, Fairhaven, Dartmouth, and Acushnet, MA (Figure S1), except for one location where we sampled two rounds. The sampling locations were selected by community members following discussions about study objectives and the need for the spatial distribution of PCB monitors. All but two selected locations are near residential homes.

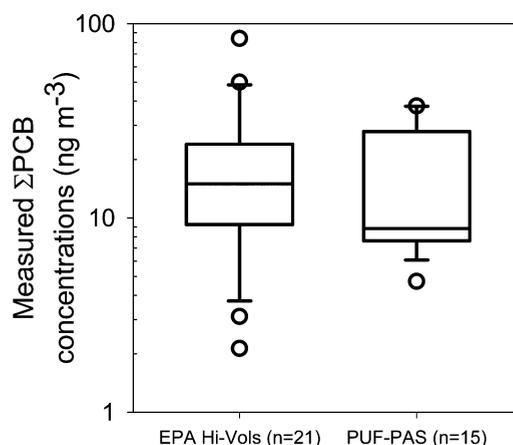
Prior to placement, PUF disks were cleaned in a Soxhlet apparatus for 24 h with hexane, followed by 24 h with acetone, and finally with a 1:1 (v/v) hexane/acetone solution for 24 h. PUFs were dried for 1 h in a ventilated fume hood, wrapped in combusted aluminum foil within Ziploc bags, and stored at  $-4^{\circ}\text{C}$ .<sup>14,15</sup> Hourly sampling rates ( $R$ ) specific to each sampler and PCB congener were modeled from local meteorology, ranging from 2 to 3  $\text{m}^3 \text{day}^{-1}$ . These sampling rates were used to calculate effective sampling volumes ( $V_{\text{eff}}$ ) for each sampler and PCB congener, ranging from 25 to 110  $\text{m}^3$ .<sup>13</sup> We used these volumes to calculate the concentration of airborne PCB from the mass collected in the passive samplers.

**Analytical Methods and QA/QC.** The laboratory methods have been described previously.<sup>12–14,16</sup> Additional details, including the QA/QC for the airborne PCB measurements, airborne PCB emission calculations, air dispersion model AERMOD, and meteorological data utilized in this investigation, are provided in the Supporting Information. PCB concentrations in water and from high-volume air sampling were reported elsewhere, and details are also provided in the Supporting Information.

## RESULTS AND DISCUSSION

**Airborne PCB Measurements.** Concentrations of airborne  $\Sigma\text{PCB}$  ranged from 0.4 to 38  $\text{ng m}^{-3}$ , with a geometric mean of  $3.1 \pm 3.8 \text{ ng m}^{-3}$ . The values of  $>10 \text{ ng m}^{-3}$  ( $n = 9$ ) are the highest values reported for outdoor  $\Sigma\text{PCB}$  PUF-PAS samples by our laboratory (Chicago, Cleveland, and East Chicago)<sup>12–14</sup> and also by others (Toronto, ON).<sup>17</sup> Our values did not show a significant difference from U.S. EPA large-

volume air sampler (Hi-Vol) gas-phase measurements<sup>7</sup> for the same months of 2015 (Mann–Whitney;  $p = 0.32$ ) (Figure 1).

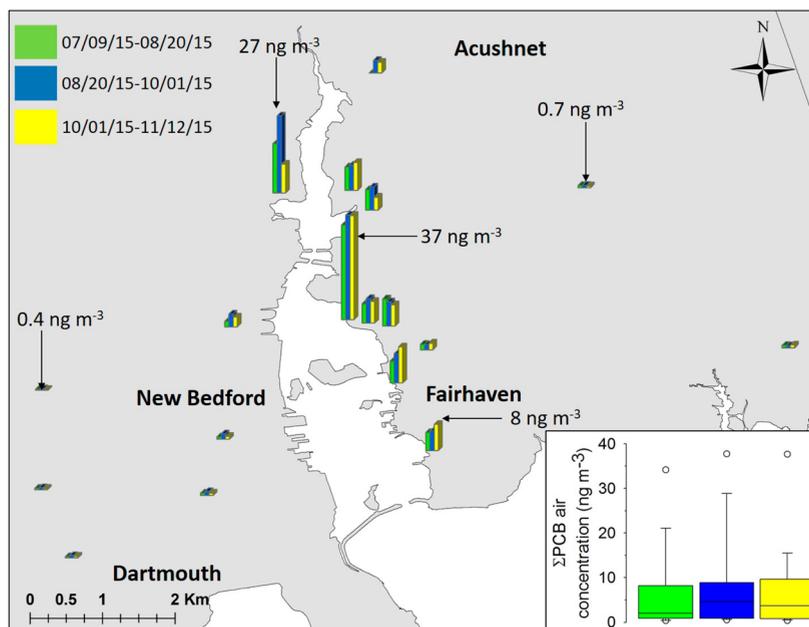


**Figure 1.** U.S. EPA high-volume (Hi-Vols) and PUF-PAS measurements for comparison of the gas phase. Both sampling methods were found from July to November 2015. No significant difference was found between the methods (Mann–Whitney;  $p = 0.32$ ). The locations of our PUF-PAS and U.S. EPA Hi-Vol samplers used in this comparison are given in Figure 3.

A clear and large spatial variability was found, with the highest values located closest to the water (Figure 2). Interestingly, little temporal variability was observed. Indeed, no statistical difference was found among the three sampling period average concentrations (Kruskal–Wallis;  $p = 0.83$ ). Samples from the same location varied by a factor of  $<3$ , whereas PUF-PAS samples in Chicago varied in average  $>6$ -fold for the same location.<sup>13</sup> In addition, the highest value showed the lowest variability (10%), suggesting a single and constant PCB source.

Very similar PCB congener profiles were found in all the samples, dominated by low and middle chlorinated congeners ( $<$ pentachlorobiphenyls). The majority of the congeners found in the samples are present in Aroclor mixtures (Figure S4). The samples have strong Aroclor 1242 and 1016 profiles, consistent with numerous reports of sediment contamination with these two commercial mixtures<sup>2–4</sup> and a consistent congener profile (sample average  $\cos \theta = 0.97$ ).<sup>11,18</sup> Samples collected farther from NBH exhibit a larger relative contribution of non-Aroclor PCB11 (Figure S5). PCB11 is a byproduct of the manufacture of contemporary paint pigments.<sup>19,20</sup> The PCB11 fraction in the samples exhibits one of the highest variabilities (standard error) of the 171 PCB congeners measured, suggesting that NBH is not the major source of this congener. These results suggest a single and continuous source of airborne PCBs, consistent with our hypothesis that the water of NBH is a large source of PCBs to the local atmosphere.

**PCB Emissions.**  $\Sigma\text{PCB}$  gross volatilization fluxes ranged from 160 to 1200  $\mu\text{g m}^{-2} \text{day}^{-1}$  for the 2015 samples. These fluxes result in  $\Sigma\text{PCB}$  emissions from the upper and lower harbor areas ranging from 90 to 140  $\text{kg year}^{-1}$ , with an average of 110  $\text{kg year}^{-1}$ . The only previous PCB emission calculations for NBH were published by Garton et al.<sup>8</sup> Using their data from 1983, we were able to predict a gross volatilization flux of 70  $\mu\text{g m}^{-2} \text{day}^{-1}$ , which is on the same order of magnitude as the lower-end estimates reported here (2015 water data). NBH fluxes are higher than those of other well-known PCB-



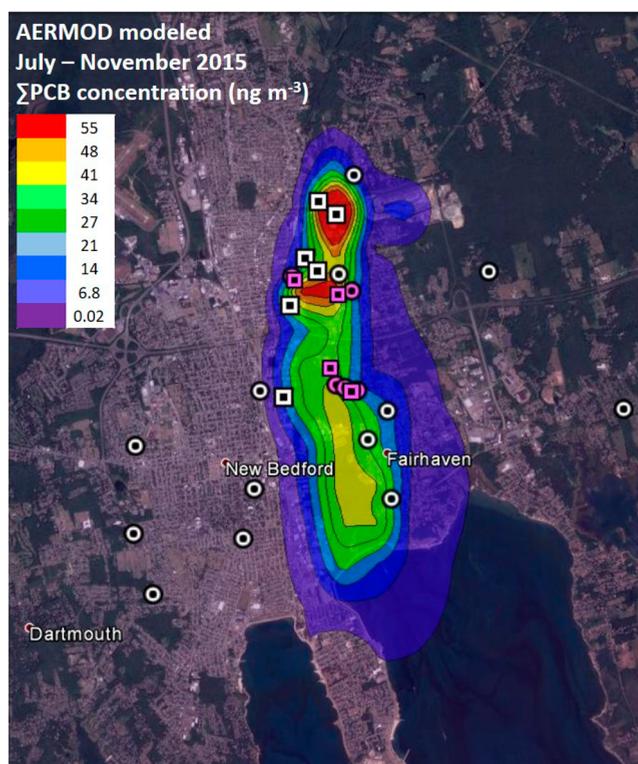
**Figure 2.** Spatial and temporal distributions of airborne  $\Sigma$ PCB (nanograms per cubic meter) in New Bedford Harbor. Sampling round 1 (green) from July 9, 2015, to August 20, 2015, sampling round 2 (blue) from August 20, 2015, to October 1, 2015, and sampling round 3 (yellow) from October 1, 2015, to November 12, 2015. The inset shows the box-and-whisker plot for the concentration distribution of the three sampling periods. Map source: Office of Geographic Information (MassGIS), Commonwealth of Massachusetts, MassIT.

contaminated water systems in the United States. For example, gross volatilization fluxes from Green Bay in 1989 ranged from 0.2 to  $5.3 \mu\text{g m}^{-2} \text{day}^{-1}$ .<sup>21</sup> New York Harbor in 1989 yielded  $\sim 3 \mu\text{g m}^{-2} \text{day}^{-1}$ .<sup>22</sup> The values for the Hudson River Estuary in 1999–2001 ranged from 0.05 to  $0.9 \mu\text{g m}^{-2} \text{day}^{-1}$ .<sup>23</sup> The values for the Delaware River in 2001–2003 ranged from 0.2 to  $2.5 \mu\text{g m}^{-2} \text{day}^{-1}$ .<sup>24</sup> IHSC in 2006 yielded  $7 \mu\text{g m}^{-2} \text{day}^{-1}$ .<sup>9</sup> Although it is very difficult to compare PCB fluxes between different studies, all these fluxes are at least 2 orders of magnitude lower than the 2015 NBH values. NBH fluxes are a result of the notably high concentrations of dissolved PCBs measured in NBH, which in some cases were in the hundreds of nanograms per liter (Table S1). Indeed, some values are only a factor of 100 below the water solubility limit.

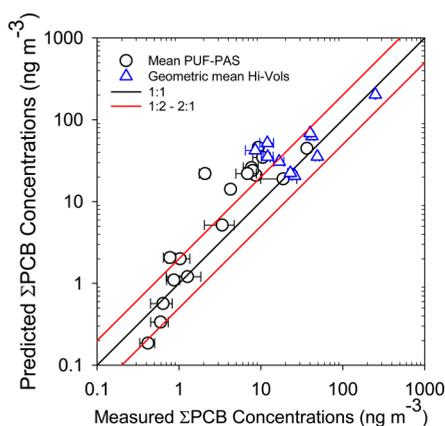
We estimated the historical trend in emissions using U.S. EPA measurements of dissolved PCBs in water collected since 2006. We excluded from consideration any water samples that were collected while there was active dredging, as indicated in the U.S. EPA water quality reports. The resulting  $\Sigma$ PCB emissions ranged from 7 to  $26000 \mu\text{g m}^{-2} \text{day}^{-1}$ . Although some values are extremely high, a clear decrease in emission over time was observed (Figure S6). This reduction is consistent with the observed decline in air and sediment PCB concentrations over time.<sup>5–7,25–34</sup>

**Modeled PCB Concentrations.** AERMOD is an EPA model for predicting the dispersion of airborne pollutants from a source, in this case NBH. We applied AERMOD to compare the measured PCB concentrations to those predicted as a function of our calculated emissions. The AERMOD predictions exhibit a PCB spatial distribution consistent with our field measurements and validate our hypothesis that NBH waters are the source of PCB in the nearby surrounding air (Figure 3).

AERMOD predicted  $\Sigma$ PCB within a factor of 2 of our PUF-PAS measurements 50% of the time (9 of 18 samples) (Figure 4). Interestingly, all the locations where the prediction



**Figure 3.** AERMOD prediction map for mean  $\Sigma$ PCB concentrations (nanograms per cubic meter) from July to November 2015. Circles represent our PUF-PAS samplers, and squares represent High-Vol samples placed by the U.S. EPA. Pink circles and squares represent the samplers used for comparison of methods (Figure 1). Both PUF-PAS and Hi-Vol samplers were active between July and November 2015, although the PUF-PAS samplers were continuously collecting while the Hi-Vol samplers were sampling in 24 h periods. Map source data copyright 2016 Google.

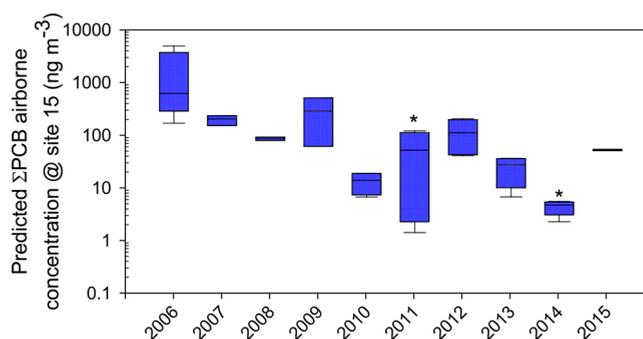


**Figure 4.** AERMOD predictions  $\Sigma$ PCB vs field measurements from July to November 2015. Circles represent mean PUF-PAS measurements ( $n = 3$ ), and triangles represent geometric means of Hi-Vol measurements from the U.S. EPA ( $n \geq 5$ ). Error bars represent one standard deviation for the PUF-PAS samplers and one geometric standard deviation for the Hi-Vol samplers. The black line represents the 1:1 line, and the red lines represent the 1:2 and 2:1 lines (i.e., factor of 2).

exceeded the measurements by a factor of  $>3$  were located close to the water and east of NBH. In general, the predictions exceeded our field measurements by an average factor of 2.6, ranging from 0.5 to 11. The U.S. EPA has been monitoring airborne PCBs in NBH since 1999 using Hi-Vol samplers.<sup>6,7</sup> Their Hi-Vol measurements for the same period of time were somewhat closer to the predicted concentrations with an average factor of 2.1, ranging from 0.7 to 5, with only one location with an average factor of  $>4.0$ . Given that the range of PCB concentrations is large and a factor of  $\sim 100$ , we conclude that the modeling approach we used is appropriate and accurately predicts the effect of NBH emissions on ambient PCBs in the air of the surrounding communities.

**Implications.** The results of this study support our working hypothesis: PCB emissions from NBH explain nearby air concentrations. Specifically, we cite three independent findings. First, we note the large spatial variation in measured airborne PCBs, with much higher concentrations close to the shoreline. These measurements were reproducible. Second, we found that the profiles of PCB congeners in the air samples are remarkably similar, and also similar to those of the commercial mixtures Aroclor 1016 and Aroclor 1242. The similarities are strongest for air samples collected close to the shoreline. Third, we found that our predicted and measured air concentrations exhibit similar ranges of values and similar spatial distributions, both decreasing in magnitude with distance from the water. To the best of our knowledge, this is the first study to show that a PCB-contaminated waterway is responsible for the nearby measured PCBs.

It is likely that PCBs have been emitted from NBH water for many years. Using the same modeling approach, we predicted airborne PCB concentrations from U.S. EPA water measurements since 2006. Our calculations illustrate a decrease in PCB emissions as well as airborne PCB concentrations (Figure 5). Our findings indicate that NBH is one of the largest ongoing sources of airborne PCBs in the United States and the cause of the highest concentrations in the neighborhoods surrounding the harbor. It is likely that NBH has been an important source of airborne PCBs in the New Bedford area since it was contaminated with Aroclors.



**Figure 5.** AERMOD predicted airborne  $\Sigma$ PCB concentrations at site 15 using U.S. EPA water concentrations from 2006 to 2015. Predictions were calculated for each month, depending on the available U.S. EPA water data. Asterisks indicate a significant difference between 2006 predicted concentrations and 2011 and 2014 (Mann–Whitney;  $p = 0.004$ ).

## ■ ASSOCIATED CONTENT

### 📄 Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.estlett.7b00047.

Details of the analytical methods, PCB emission calculations, water PCB concentrations, and maps (PDF)

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### Notes

The authors declare no competing financial interest.

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