

**POLLUTION PREVENTION AND
MANAGEMENT STRATEGIES
FOR POLYCYCLIC AROMATIC HYDROCARBONS
IN THE NEW YORK/NEW JERSEY HARBOR**

September 2007

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with a preface by
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PREFACE

What is new about this report on reducing the burden of polycyclic aromatic hydrocarbons (PAHs) to the New York/New Jersey Harbor—this fifth major report of the Harbor Consortium? Much of the format is the same. But both the technical complexities and the process leading to a consensus agreement to issue this report presented unprecedented challenges.

The Consortium Process. Let's quickly review the Consortium process to date. Selecting the first four contaminants (mercury, cadmium, polychlorinated biphenyls [PCBs], and dioxins) to be addressed by the New York Academy of Sciences' Harbor Consortium had been a relatively relaxed process. The overall purpose of the selection was to be the same each time: to determine how the contemporary introduction of a contaminant into the Harbor could best be prevented. We would examine established criteria to identify substances or compounds that were in some way a significant threat to the Harbor's health. Through mass balances and other environmental measurement techniques, we would explore both historical and contemporary loadings of those contaminants to the Harbor (entering directly or through waterways leading to it). Then, using techniques pioneered or improved through industrial ecology methods, we would track down the sources of those contaminants to the loadings and determine how they could be prevented, slowed, or diverted from entering the Harbor.

These basic steps were to be the technical or scientific mode of operation for the new Harbor Consortium. But, in addition, a fundamentally new process would accompany these steps. It was this: representatives from the diverse bi-state institutions (public, private, nonprofit, quasigovernmental) listed in this report would observe and/or participate in the technical process—and then be in a position to recommend (by consensus, we hoped) who should do what to achieve the pollution prevention purposes.

As the four major Harbor Consortium reports preceding this one attest, both the technical process for evaluating what is coming into the Harbor and the social process of recommending by consensus how best to slow and/or prevent Harbor contamination have worked far better than anyone could have expected.

PAHs—a distinctive challenge. But the fifth contaminant (PAHs) has, in many ways, been the toughest test. First, the Consortium had to select which would be the last of five toxicants to go through its process. Be-

cause it was to be the last, and because it had become clear that key players in the bi-state region really were paying attention to the work of the Consortium (its recommendations were being implemented in a wide variety of ways), there really was something at stake in this selection. To help decide, a paper was written for the Consortium to explore possible choices. Additionally, several newly emerging toxicants of concern were advocated for study by some Consortium members. The Consortium had always handled this selection process by a vote; in the end, PAHs were selected by an exceedingly close margin. The technical experts associated with the Consortium knew at once that the analysis of PAHs would, at the least, pose new technical challenges.

Why? First, PAHs make up a collection of more than 100 different chemicals. These chemicals as a class are believed to be carcinogenic and to have other harmful effects on human health, as well as having adverse ecological effects. But the toxicity of the various chemicals clearly varies significantly. PAHs break down in soil and water at quite different rates that are affected by temperature and other environmental factors; this made tracking and evaluating them even more difficult. PAHs are ubiquitous and have both natural and anthropogenic sources, primarily combustion activities. They are produced or formed during incomplete combustion, not just in coal, oil, and gas in stationary and mobile sources, but also in combustion of garbage and diverse other organic substances. However, their distribution is not limited to emissions to the air that are deposited in the Harbor, since they, like other substances with which the Consortium has worked, may be deposited on land and move to the Harbor by means that are both diverse and contested.

Further, as the Consortium was soon to learn, some of the major sources of PAHs that proved to be of greatest concern for the Harbor had nothing to do with local combustion processes, but were found, for example, in manufactured goods such as coal tar, creosote, and motor oil. To what extent would PAHs found in these materials reach the Harbor from their diverse applications in products such as treated wood and driveway sealants?

It certainly was not clear at the outset that a coherent and persuasive account of the burden to the Harbor from PAHs could be developed. With the active support of some of the better academic and public sector environmental scholars in the bi-state region and beyond, the Harbor Consortium staff went to work to

bring the same discipline to this highly complicated technical challenge as they had brought to what now seemed the far easier task of tracking mercury or cadmium, the subjects of the initial two Consortium reports. The report on PAHs you are about to read demonstrates again that “our” staff is just very good. It has always been made up of Academy employees, and has been headed for the past three years by Marta Panero, Ph.D. In this report’s case, the lead staff effort is the excellent work of Sandra Valle. Their two university-based consultants, professors Leslie Shor and Lisa Rodenburg, helped carry out this work. You will find a much more sophisticated account of the fate and transport process by which PAHs from various sources reach the Harbor than we had been able to develop for the contaminants in the earlier reports. In fact, it is likely that the Academy team have developed methods and data that belong in the peer-reviewed literature, as well as in this report. But I encourage the reader to take time to read this quite extraordinary account, because the weaving of this technical fabric into a quite clear picture of which PAHs reach the Harbor and how they potentially affect the Harbor’s general health is, in my view, a significant achievement.

The technical account is just the half of it. As indicated above, the goal of the Consortium is not just to know what reaches the Harbor, but to know it well enough to recommend what we, who live and work in this complex watershed, ought to be doing to protect it. If the initial presumption of many Consortium members had been right and the major sources of PAHs reaching the Harbor had been combustion sources, then it is likely that the recommendations in this report would largely be a repeat of those we made on “poor combustion” processes when examining dioxins, to which we would add recommendations on the better-regulated sources (mobile and stationary). But the analysis clearly suggested that our focus should turn to the use of two key manufactured product types that have been of concern to environmental regulators for some time: coal tar sealants (such as those found on driveways and parking lots) and creosote-treated woods in diverse applications (including marine ones). Both product types slowly emerged as very important sources of PAH contamination to the Harbor.

Consequently, as the recommendations for pollution prevention began to focus on these two product types, those associated with their manufacture became regular attendees at a series of Consortium workshops

and meetings. In one sense, there was nothing new in the Consortium being asked to examine—and to enter into a dialogue with producers and even recyclers of—products that contained worrisome substances that might reach the Harbor. Indeed, the Consortium had found it useful numerous times in the past six years to draw those associated with either the production or use of such products into the Consortium discussion, so that it could decide on the most effective recommendations for protecting the Harbor. The Consortium had found quite effective ways earlier to conduct its inquiries and to make its decisions when addressing the consequences of production and use, even when there were active public processes involved (including remedial design, litigation under CERCLA, etc.). But in this case, the Consortium found itself host to players who were actively involved in the full range of both technical and political struggles concerning the future use of creosote and coal tar. In one case, the products had been prohibited from use in one local city, and other jurisdictions were considering similar action. In another case, the state legislatures in both New York and New Jersey were in the midst of a several-year legislative process to control the use of the product, even as the Consortium process continued.

Achieving consensus again. The challenge to the Consortium was to keep focused on its technical base, to evaluate the challenges to its analyses that these players from several sides were making, and to continue on a path to the soundest recommendations it could make. In the midst of that kind of controversy, the Consortium members could still find consensus. It took an extra and, in my view, extraordinarily careful Consortium meeting in June of this year to allow the diverse Consortium participants, in a very open discussion, to understand the staff conclusions and to hear the views of those affected parties who differed with the staff on a variety of technical issues and, of course, on the recommendations. In the end, the story that unfolds in this report is the one developed by the staff, and the Consortium again reached a consensus on the entire technical report and the resulting recommendations. The report here provides the reader with an opportunity to review key elements of that debate.

What is still missing. As always, there were important lessons in this effort that may not be reflected fully in the report itself. The most important to me, as the Chair of the Consortium, was to recognize how

far we, as a society, are from having the institutional means to help our citizenry sort out what a commitment to being a “green” consumer actually means. As we struggled hard with the issues of what is involved in creating and maintaining something as simple as a driveway or parking lot, the diverse claims of those who wanted to limit use of sealant products and those who wanted to use or market the product proved exceedingly hard to evaluate. The staff did limited research to evaluate the claims and counterclaims—and ended up with recommendations on limiting use of certain PAH products that it viewed as a threat to the Harbor. But the development of broad criteria or the arraying of data that would enable the clear recommendation of one product to replace another lay beyond the scope of our work. The same was true for creosote-treated wood in marine environments, where work to test alternative products is under way. The claims and counterclaims of those advocating or criticizing the alternatives are not fully resolvable by a Consortium with the limited resources and scope of this one. Yet it is patently clear that credible independent institutions capable of making such evaluations are needed. As a Consortium we could responsibly say, “limit or divert this contaminant from getting to the Harbor where it may/will cause harm, and to do so, we unanimously recommend limiting Z use of X to do task Y.” What we could not do, in many or even most cases, was to say persuasively that we are confident that “task Y can best be done by using/doing W.” In discussions with diverse Consortium members and many others, I conclude that the establishment of an institution or institutions capable of doing that work is the sine qua non of greening as an effective societal goal.

What we have achieved. I want to conclude this preface by reviewing the goals of the Consortium and what this fifth report does to complete the project. In doing so, I do not want to provide a synthesis of our work. A final report with the goal of doing that should be ready for review by the Consortium by the end of the year. Still, I do want to catch the significance of the fact that this fifth Consortium report will be issued exactly 10 years after the formation of the Consortium was recommended at a well-attended workshop I chaired in September 1997. The first sentence of the final paragraph (headed Outcomes) of the report of that workshop, published by the New York Academy Sciences in February 1998, said the following:

The ultimate objective of the Consortium process is to develop the industrial ecology analyses that will frame and inform long-term solutions to the flows of five critical contaminants into the New York Harbor and communication of those solutions such that a broad commitment to their implementation is realized. (*Industrial Ecology and the Environment: Applications to the New York Harbor*, p. 40)

That precise work, and much more, has been done. I want to thank those who have supported this Consortium process financially, donated Consortium member time and talent, and provided the Consortium with an institutional home. Finally, and especially, I want to thank our talented and persistent staff. It has been an unprecedented process.

Charles W. Powers
Chair

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The ongoing contributions of our Harbor Consortium, led by its Chair, Charles W. Powers, merit special mention. The Consortium has been at the center of this project, playing a key role in overseeing the research and the pollution prevention deliberation process. We are grateful for the guidance of all its members, their continued involvement in the project, and their generous sharing of time and expertise.

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LIST OF ACRONYMS

| | | | |
|--------------|--|---------------|--|
| AFUE | Annual fuel utilization efficiency | HW | Hazardous waste |
| APCD | Air pollution control device | IRIS | Integrated Risk Information System |
| BAP | Benzo[a]pyrene | kg | Kilogram |
| BMP | Best Management Practices | Kow | Octanol–water partition coefficient |
| BOF | Basic oxygen furnace | lb | Pound |
| BTU | British thermal unit | LTO | Landing and take off |
| BUR | Built-up roof | MES | Mild extract solvents |
| CAFE | Corporate Average Fuel Economy | mg | Milligram |
| CARP | Contaminant Assessment and Reduction Project | MHF | Multiple hearth furnace |
| CDD | Chlorinated dibenzo-para-dioxin | Mi | Mile |
| CDF | Chlorinated dibenzofuran | MISLE | Marine Information for Safety and Law Enforcement |
| CDT | Clean diesel technology | MS4 | Municipal Separate Storm Sewer System |
| CHE | Cargo handling equipment | MSW | Municipal solid waste |
| CHP | Combined heat and power | MW | Molecular weight |
| cm | Centimeter | NEI | National Emissions Inventory |
| CO | Carbon monoxide | NJ | New Jersey |
| CSF | Cancer slope factor | NJADN | New Jersey Atmospheric Deposition Network |
| CSO | Combined sewer overflow | NOAA | National Oceanic & Atmospheric Administration |
| DIFM | Do-it-for-me | NOEL | No observed effects level |
| DIY | Do-it-yourself | NOx | Nitrogen oxide |
| DOC | Dissolved organic carbon | NRC | National Response Center |
| DOC | Diesel oxidation catalyst | NY | New York |
| DOD | Department of Defense | OWB | Outdoor wood boilers |
| DPF | Diesel particulate filters | PAH | Polycyclic aromatic hydrocarbon |
| EGR | Exhaust gas recirculation | PCB | Polychlorinated biphenyls |
| EIA | Energy Information Administration | PCD | Port Commerce Department |
| ESP | Electrostatic precipitator | p-DCB | Para-dichlorobenzene |
| EU | European Union | PM | Particulate matter |
| FAA | Federal Aviation Administration | ppm | Parts per million |
| FB | Fluidized bed | R-EMAP | Regional Environmental Monitoring and Assessment Program |
| FF | Fabric filter | Rfd | Reference dose |
| ft | Feet | RMW | Regulated medical waste |
| FUDS | Formerly used defense sites | RTA | Railway Tie Association |
| FWPCA | Federal Water Pollution Control Act | | |
| GLBTS | Great Lakes Binational Toxics Strategy | | |

LIST OF ACRONYMS (CONT'D)

| | |
|-----------------------|---|
| SCR | Selective catalytic reduction |
| SO_x | Sulfur oxides |
| SPCC | Spill prevention control and countermeasure |
| SPDES | State Pollutant Discharge Elimination System |
| SSI | Sewage sludge incinerators |
| TDAE | Treated Distillate Aromatic Extracts |
| TEF | Toxicity equivalency factor |
| TRI | Toxics Release Inventory |
| U.S. DOT | United States Department of Transportation |
| U.S. EPA | United States Environmental Protection Agency |
| USCG | United States Coast Guard |
| VOC | Volatile organic compounds |
| WTE | Waste-to-energy |
| WWPI | Western Wood Preservers Institute |
| yr | Year |

EXECUTIVE SUMMARY

Overarching P2 Recommendations for Major Sources of PAHs in the Watershed Region

A wide spectrum of sources contributes to total polycyclic aromatic hydrocarbon (PAH) releases in the New York/New Jersey Harbor Watershed, ranging from vehicle use, to fuel combustion, to leaking and dumping of petroleum products, as well as industrial and commercial activity. However, of the numerous PAH sources evaluated in this report, 11 major sources are estimated to contribute individually more than 2% to the total emissions released to each primary medium of release (air, water, and land) (Fig. 1).¹

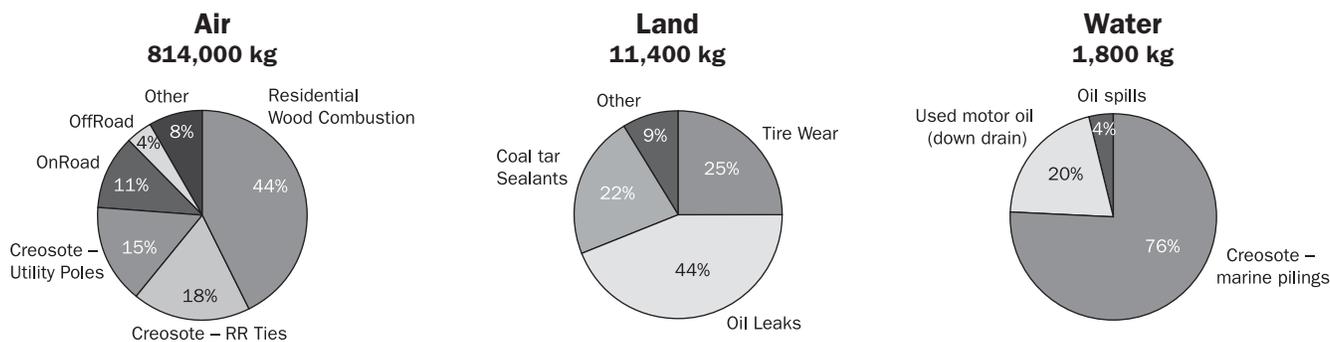
While these major emission sources differ in definition, some share fundamental characteristics. For example, many of these sources share a similar mechanism of release. PAHs released from vehicle exhaust and residential wood heating, as well as several of the minor sources, are all the result of combustion processes, many of which include the combustion of fossil fuel. Furthermore, although vehicle exhaust, tire wear, used motor oil disposal, and oil leaks differ with respect to medium and mechanism of release (i.e., petrogenic vs. pyrogenic), they are all associated with transportation-related activities (TABLE 1). Thus, by systematically grouping sources of PAHs based on common characteristics, overarching pollution prevention recommendations can be made that address multiple sources of PAHs.

In general, PAH emission reductions can be addressed at several points prior to the contaminant reaching the endpoint or environment of concern (in our case the Harbor); these points include the following:

1. at the supply side (e.g., reducing the use of materials containing PAHs in the manufacturing processes, thus generating products with no, or low PAHs);
2. at the demand side (e.g., reducing the demand for fossil fuel consumption through alternative design and material substitution);
3. at the point of release (e.g., increasing the efficiency of the combustion device or implementing BMPs to prevent leakage); and
4. after the PAHs have been released but prior to their reaching the environment of concern (e.g., through stormwater management measures).

Implementation of various measures at all of these points is critical for a comprehensive abatement strategy, which may include pollution prevention (P2) as well as best management practices (BMPs). Below is a summary of overarching P2 and BMP alternatives that address the reduction of PAH releases from combustion, transportation, and material-related PAH sources. For more details on these recommendations, see the specific sector sections presented in the technical section of this report.

Figure 1. Relative releases of PAHs by primary medium of release (air, water, and land)



“Other” air and land releases refer to sources that individually contribute 2% or less to total releases to the respective medium. These include industrial sources, residential and commercial fossil fuel combustion, open burning of household waste and tires, other transportation-related sources (e.g., vessel and personal boats, locomotive, airplane), and PAHs in ash residue that is sequestered in a landfill.

*Average of the estimated range of releases.

1. For some sources, such as creosote-treated railway ties, it is believed that PAHs are released to both air and land; however, air is considered the primary medium of release for creosote-treated railway ties and poles because it is to this medium that releases are more likely to impact the Harbor. Similarly, coal tar sealants release PAHs to land and air; however, we were able to estimate releases only to land.

Table 1. Categorization of major sources of PAHs in the Watershed region.

| Combustion | Transportation | Materials containing PAHs |
|--|--|---|
| Mobile Vehicle exhaust Nonroad engine exhaust | Mobile Vehicle exhaust Nonroad engine exhaust Tire wear Used motor oil (down storm drains, leaking) | Mobile Tire wear Used motor oil (down drain, leaking) |
| Stationary Wood combustion Other, minor sources | Infrastructure Coal tar parking lot sealants Creosote railroad ties Creosote marine pilings | Infrastructure Coal tar parking lot sealants Creosote railroad ties Creosote utility poles Creosote marine pilings |
| Other Minor sources | Other Minor sources | Other Minor sources |

Overarching Pollution Prevention (P2) and Best Management Practice (BMP) Recommendations:

Combustion-Related Recommendations

1. Reduce supply-side combustion activities.
 - Promote the generation of power from renewable sources of energy (that do not release PAHs).
2. Reduce demand for combustion activities.
 - Promote energy conservation by means that include increasing the energy efficiency of buildings and homes (e.g., through improved insulation and windows), strengthening building energy standards, improving efficiency standards for heating and cooling units, and the use of energy efficient appliances. Consider implementing these measures through voluntary and educational measures as well as revised energy codes.
 - Promote energy efficient manufacturing operations, services, and products. Identify and encourage consumption of products that are less energy intensive to manufacture, such as products that contain recycled material.
 - (Recommendations for reducing emissions from vehicle fuel combustion are included under recommendations, below.)
3. Reduce PAH emissions from combustion activities.

- Optimize combustion conditions (e.g., high temperatures, adequate oxygen supply) through the adoption of best management practices.
- Promote utilization of the best available pollution control devices to capture and minimize the release of PAHs, such as technologies that capture particulate bound PAHs (e.g., filters, electrostatic precipitators) and atmospheric PAHs (e.g., carbon beds).

Transportation-Related P2 Recommendations

1. Reduce vehicular use and the development of related infrastructure that can release PAHs.
 - Increase infrastructure capacity of public transportation, while establishing incentives for responsible vehicle use (such as taxes, congestion pricing, or incentives for public transport).
 - Enhance the diversity of transportation options by improving public access to mass transit, thus reducing vehicle traffic, congestion, and paved impervious parking areas.
 - Prioritize pedestrian and biking areas when developing or redeveloping communities. Plan communities that connect via pedestrian and bike pathways, and via public transportation.
 - Prioritize construction projects that fill in areas in already developed urban centers (possibly through policy and permitting

practices). Couple this with programs that encourage the settlement in, and development of, urban centers, possibly by providing incentives for urban dwellers and those that choose to live close to towns. Support informational and educational campaigns about the benefits of living in urban centers.

2. Reduce PAH releases from transportation-related activities and infrastructure.
 - Increase the efficiency of combustion engines in vehicles and nonroad equipment while increasing the market share of vehicles and equipment that are not powered by the combustion of fuels that release PAHs. This could be done by aggressively raising the federal efficiency standards for vehicles bought or sold in the U.S.
 - Encourage the use of engines that produce fewer PAHs through incentives such as variable registration fees (i.e., lower

NOTE ON ALTERNATIVE PRODUCTS AND MATERIALS

The purpose of this report is to identify sources releasing PAHs in the region and propose recommendations that will reduce the overall quantity of PAHs in use or released within the NY/NJ Harbor Watershed and thus reduce the amount of PAHs that may eventually reach the Harbor. While for some sectors we identify alternative materials and/or practices that could potentially be implemented and/or used to reduce the quantity of PAHs released in the Watershed, the relative environmental impacts of these alternatives, outside of PAHs, are often undetermined and demand further investigation. Some of the analytical tools that can be used to make a comparison between products and policies are life-cycle assessment and material-flows analysis (specifically burden shifting). A comparative analysis would be optimal, although it goes beyond the scope of our current research. Therefore, we are not in a position to recommend any particular alternative materials or products.

registration fees for engines that are likely to release fewer PAHs).

- Reduce unnecessary emissions and releases of PAHs through proper maintenance practices, such as regularly checking equipment for malfunctions and leaks, and through the enforcement of anti-idling regulations; consider extending anti-idling regulations to all nonroad equipment.

Materials-Related P2 Recommendations

1. Decrease the supply of, and demand for, materials containing PAHs.
 - Explore the potential costs and benefits of using alternative materials that do not contain PAHs. Costs may include price of purchase, time required to implement change or use of materials, and an inferior product, while benefits might include a superior product, healthier work environment, less harm to the environment, and fewer regulations. Educate industry and consumers on the results of these assessments.
 - Investigate the development of substitutes for PAH-containing materials, as well as product and structural designs that eliminate the need for materials containing PAHs. This may be pursued through industry and federally supported studies.
2. Reduce PAH releases from the use of materials that contain PAHs.
 - When feasible, avoid the use of products that contain and release PAHs.
 - Promote the use of best management practices to reduce PAH releases throughout the manufacturing stage and use of products and materials. This could be done through education and outreach.
 - Educate people on the proper disposal of materials containing PAHs, while increasing opportunities to recycle or properly dispose of the materials (e.g., recycling facilities, pick-up services, collection locations at retail establishments).

Stormwater Management Recommendations

The last point at which P2 and BMP recommendations can be made is after PAHs have been released but before

they reach the Harbor. These types of recommendations are relevant to most sources releasing to land (and to the atmosphere if PAHs deposit onto land). For PAH sources that release directly to the Harbor there are no further opportunities to curb loadings.²

The quantity of PAHs reaching the Harbor via stormwater runoff may be reduced by several means:

- Increase the area of pervious surfaces (green infrastructure) that can potentially filter contaminants out of stormwater (e.g., increase vegetated medians, sidewalks, and green roofs; restore abandoned developments into green spaces; use pervious surface material). Consider implementing these measures through education and outreach as well as policy and sustainable-development initiatives.
 - Implement and enforce stormwater management policies for already developed, redeveloped, and newly developed areas statewide. For example, for large development projects that will increase overall impervious surface area, consider requiring on-site stormwater treatment measures, such as stormwater filtration, retention systems, or stormwater capture and reuse. Consider retrofitting existing sites with similar stormwater treatment measures.
 - Educate the public on the importance of keeping stormwater clean and how they can help (e.g., not dumping contaminants down the stormwater drains, reducing the impervious surface area of their property).
 - Conduct further stormwater sampling to confirm the importance of stormwater to total loadings of PAHs to the Harbor.
- Invest in the update/improvement of PAH emission factors, ensuring that, when possible, all 16 U.S. EPA priority PAHs are evaluated. Improved emission factors for sources that have been identified as potentially significant should be developed first.
 - Develop emission factors for sources of PAHs that have been identified but for which data that allow release estimates to be calculated are not available.
 - For activities that have been identified as potentially significant sources of PAHs, collect more accurate and detailed activity data. These data potentially will provide clarity on the nuances of emission rates as they relate spatially and temporally in a region.

Data Gap Recommendations

While efforts were made to use the best available emission factors and source activity information when calculating releases of PAHs, some data were incomplete and/or outdated, and/or there were uncertainties with the data ultimately impacting the uncertainty of the emission estimate. The following recommendations address the need for ongoing improvement of PAH emission factors and data collection. Recommendations addressing specific sectors where data gaps have been identified are included in the technical section of this report.

2. More information on stormwater best management strategies and related policies will be available in a forthcoming Harbor Project report on how to prevent the mobilization of suspended solids before they reach waterways.

SUMMARY OF FINDINGS

The goal of this report is to use an industrial ecology approach to evaluate all potential sources releasing polycyclic aromatic hydrocarbons (PAHs) in the New York/New Jersey Watershed region and to evaluate their potential to reach the Harbor. We have estimated primary releases of PAHs from each source by compound, county, and medium of release (i.e., to air, land, or water). We also estimate the likelihood of each of these emissions to reach the Harbor via fate and transport modeling. Finally, we compare our estimated inputs to the Harbor from this exhaustive industrial ecology approach with an independent mass balance that computed PAH inputs based on environmental monitoring data. Our ability to demonstrate broad consistency between the two approaches helps validate the accuracy of our findings, despite the inherent difficulty of the task.

This section provides the following:

- A. Summary findings of the major sources of PAHs in the Harbor watershed region (including classification by medium of release and source category). Sources are considered major if their emissions individually comprise more than 2% of releases from all sectors evaluated in this report that release to air, water, or land (Fig. 1). Sources that are considered minor to this particular region are discussed in Section 4 of this report; they may be considered major sources of PAHs in other regions;
- B. An explanation of the likelihood of releases to reach the harbor (based on the fate and transport model) and a comparison of our estimates of loadings to the Harbor to the independently developed mass balance assessment; and
- C. A summary of related P2 recommendations to curb releases from the major sources affecting the NY/NJ Harbor.

A. Major Sources of PAHs in the Watershed

PAHs can be found naturally in petroleum deposits and are produced naturally through biogenic processes³; they are also the products of the incomplete combustion of organic material. Some PAHs are man-

ufactured for use in dyes, insecticides, and solvents. Historically, anthropogenic point sources were the major source of PAHs; however, due partly to pollution control devices, nonpoint sources have become the dominant source of PAHs. Consequently, the major sources currently releasing PAHs in the Watershed area may be classified into three general categories:

- Combustion processes, mobile and stationary (e.g., wood, fossil fuel)
- Petroleum spills/dumping
- Releases from products made with petroleum or coal (e.g., creosote, coal tar).

Specific sources that fall under these general categories include residential wood combustion, vehicle exhaust, creosote-treated wood, refined coal tar sealants, and petroleum leaks and dumping (TABLE 2).

Releases by Medium

Our estimates of primary releases of PAHs indicate that atmospheric releases are the dominant source of PAHs in the Watershed (FIG. 2). This is partially due to the pervasiveness of combustion activities throughout the Watershed, particularly the combustion of wood in residential heating units and vehicle activity, and partially due to PAH volatilization from creosote-treated wood (FIG. 1).

Releases by Source Category

Several major sources of PAHs in the Watershed fall into the transportation sector, and include vehicle exhaust, motor oil leaks and improper disposal, non-road engine exhaust, and surfaces sealed with refined coal tar sealants. These comprise approximately 15% of total releases to all media in the Watershed.⁴ Wood combustion comprises more than one-third of total releases from major sources; however, the dominant source of PAH releases is creosote-treated wood (including uses in water and on land) (FIG. 3).

B. Potential for PAH Emissions to Reach the Harbor

Not all PAHs that are released into the environment will ultimately reach the Harbor. The potential for

3. PAHs may be synthesized by biochemical processes in both terrestrial and marine organisms, and released into the environment by metabolic activity or decomposition. Given the urbanization of the Watershed area, it is likely that this is a relatively small source.

4. Creosote-treated railway ties and marine pilings could also be considered under the PAH release category Transportation (increasing the percentage of emissions from this group closer to 50%); however, given their collectively large release of PAHs, we chose to present creosote-treated wood emissions in a separate category.

Table 2. Summary of major sources of PAHs within the Watershed

| Source | PAH release in the Watershed | | PAH loadings to the Harbor ^a (kg/yr) | Emission factor/ratios applied | | Level of regional activity |
|--|------------------------------|-------------------|---|--------------------------------|--------------------------|----------------------------|
| | Quantity released (kg/yr) | Medium of release | | Particle-bound or gaseous | Uncertainty ^b | Uncertainty ^c |
| Residential fuel combustion | | | | | | |
| Residential wood stoves and fireplaces | 341,200 | A | 500 | P&G | E | II |
| Materials containing PAHs | | | | | | |
| Creosote, utility poles | 122,200 | A | 300 | G | D | II |
| Creosote, railway ties | 291,600 | A, L | 500 | G | D | I |
| Creosote, marine pilings | 1600 | A, W | 800 | G/P | D | III |
| Refined coal tar sealant | 900–5800 ^d | L | 900 ^e | P | C | I |
| Transportation | | | | | | |
| Vehicle exhaust | 91,500 | A | 300 | P&G | D | II |
| Tire wear | 2800 | L | 1400 | P | B | II |
| Oil leaks | 5000 | L | 2600 | P | D | IV |
| Improper disposal of used motor oil (down drain) | 400 | W | 200 | P | B | IV |
| Nonroad internal combustion | 32,500 | A | 100 | P&G | D | II |
| Oil spills and dumping | | | | | | |
| Oil spills ^f | 70 | W | 70 | P | D | IV |

A= atmosphere, W= water, L=land, P= particle bound, G= gaseous

a Loadings to the Harbor represent the calculated loadings described in The Potential for PAHs to Reach the Harbor found later in this report.

b The U.S. EPA gives a rating to emission factors in their AP-42 database of air pollutant emission factors, ranging from A to E, with A being the best. The rating is a general indication of the reliability or robustness of the factor. When emission factors were not rated by the U.S. EPA or another source, the same metrics used by the U.S. EPA to assign a rating were applied. See Appendix D for a description of rating metrics.

c A rating system similar to that used by the U.S. EPA was created to represent the uncertainty of activity levels used to estimate emissions. See Appendix D for a description of rating.

d This estimate is based on a particulate PAH yield published in Mahler et al. [1] and reflects the range of area that is sealed per gallon of sealant (60–80 square feet) and the length of time between resealings of a surface. (Surfaces may be sealed every 1 to 5 years; therefore, the total area of sealed surface may be proportional to 5 times the annual quantity of sealant consumed.) An unpublished study by the City of Austin estimates an average annual rate at which the sealant is worn away. When this rate is used and it is assumed that the area of sealed surface is equal to 5 times the annual consumption rate, it is estimated that up to 40,000 kg/yr of PAHs are released (to land and air combined) in the Watershed region.

e This estimate is based on yields published in Mahler et al. [1] and the average area sealed per gallon of sealant and resealing rate. When the unpublished City of Austin wear rate is used, and the fate and transport factors are applied, it is estimated that approximately 1600 kg/yr of particulate PAHs are reaching the Harbor.

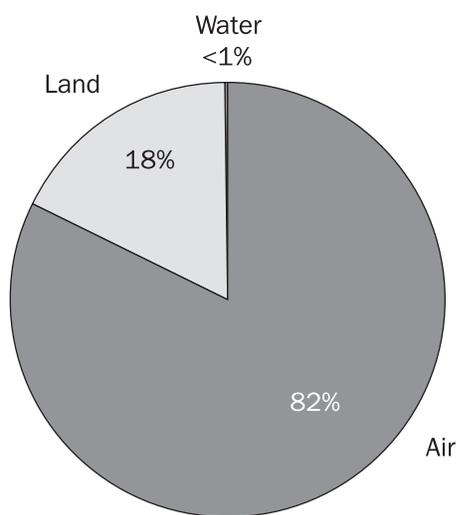
f It should be noted that the quantity of oil spilled varies greatly from the year to year. The value presented here is an average of spills that reached the water in the Harbor area reported in 2001–2004.

PAHs to reach the Harbor depends on the characteristics of the particular PAH compounds and the medium of release. It also depends on several factors regarding the point of release, including the proximity of the point of release to the Harbor; the distribution of pervious versus impervious land surfaces; and the hydrodynamics of stormwater, sewers, and surface water features, among many other factors. In an attempt to understand the potential for different PAH emissions (by source, compound, and location) to reach the Harbor, a simple fate and transport model was constructed to estimate transmission of each PAH compound from major emission sources in each county in the region.

A large number of simplifying assumptions were required to make this task tractable, and the selection of “typical” fate and transport parameters is necessarily somewhat arbitrary. These assumptions will influence the calculated transmission efficiency of each PAH from each source to each medium from each county, and, thereby, the resultant prioritization of PAH emission sources most impacting the Harbor. For a more detailed discussion of the fate and transport analysis, see APPENDIX C of this report.

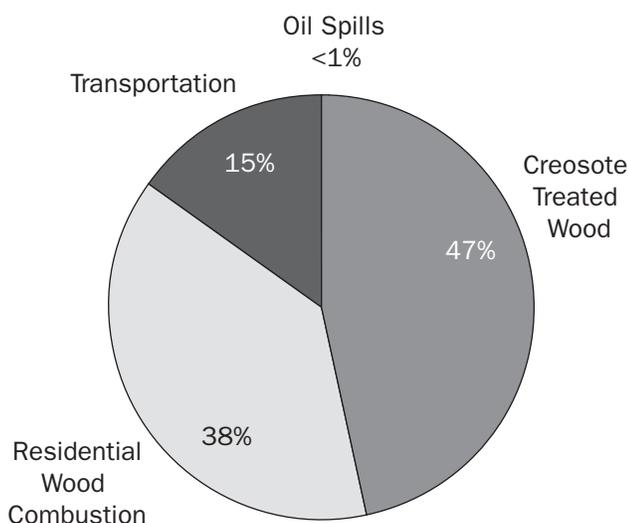
The largest primary PAH emission sources in the region are very different from the sources that con-

Figure 2. Estimated relative PAH emissions from the major sources of PAHs in the Watershed to air, water, and land



Land releases include releases from creosote-treated railway ties, and the average estimated release from coal tar sealed surfaces using yields from Mahler et al. [1].

Figure 3. Estimated relative contribution to total releases from the major PAH sources in the Watershed



Releases from creosote-treated railway ties to land are included, as well as the average estimated release from coal tar sealed surfaces using yields from Mahler et al. [1].

tribute most to PAH loadings to the Harbor waters⁵ (FIG. 4). Recall that the dominant sources of primary PAH emissions are atmospheric sources, including wood combustion, volatilization from creosote-treated wood, and vehicle exhaust. Even accounting for wet and dry deposition of particles, gross gas absorption of PAHs by surface water, and runoff of atmospheric fallout in stormwater into streams and sewers, only a very small percentage of primary atmospheric emissions ever reach Harbor waters. By contrast, sources involving emission of PAHs directly onto impervious land surfaces in counties nearest the Harbor are readily transported to harbor waters, and dominate PAH loadings to the Harbor. These sources include oil leaks, tire wear, and the use of coal tar sealants.⁶

Our estimates indicate that transportation-related activity (i.e., on-road and off-road engine exhaust, tire wear, and motor oil disposal and leakage) is the source category contributing most greatly to total loadings of PAHs (FIG. 5). Releases from coal tar sealed surfaces could also be categorized under transportation, in which case, transportation-related activity would be responsible for over 70% of estimated PAH loadings to the Harbor.

B.1 Comparison of Estimated PAH Loadings with Mass Balance

Our industrial ecology and modeling approach allows us to follow primary releases across environmental compartments throughout the region and to prioritize primary PAH sources by their ultimate contribution to PAHs in Harbor waters. In this section, we demonstrate the consistency of our estimated loadings using this approach with results of an independent analysis calculating loadings from environmental monitoring data, referred to as the PAH mass balance. While consistency between the two independent approaches by no means guarantees the accuracy of either, it does provide some reassurance that the analysis is likely to be on (or near) target.

The mass balance for 14 of the U.S. EPA's priority PAHs was developed using data primarily from the Contaminant Assessment and Reduction Project (CARP) [2], the Regional Environmental Monitoring and Assessment Program (R-EMAP) [3], and the New Jersey Atmospheric Deposition Network (NJADN) [4].⁷ The goal of the mass balance was to understand the flux of PAHs entering and leaving the Harbor

5. Our focus is on PAH pollution sources reaching Harbor waters. Other emission sources may still impact other geographic locations or environmental compartments.
 6. The average estimate of loadings from surfaces sealed with refined coal tar-based sealants is presented in Figure 4. The upper end of the estimated coal tar-sealant loadings is approximately 2000 kg/yr.
 7. The PAHs included in the mass balance are naphthalene, acenaphthene, fluorene, phenanthrene, fluoranthene, pyrene, benz[a]anthracene, chrysene, benzo[a]pyrene, perylene, benzo[b+k]fluoranthene, benzo[ghi]perylene, indeno[1,2,3-cd]perylene, and dibenz[ah]anthracene.

Figure 4. Estimated releases (kg/yr) of the major sources of PAHs in the Watershed (white) compared with estimated loadings (black)

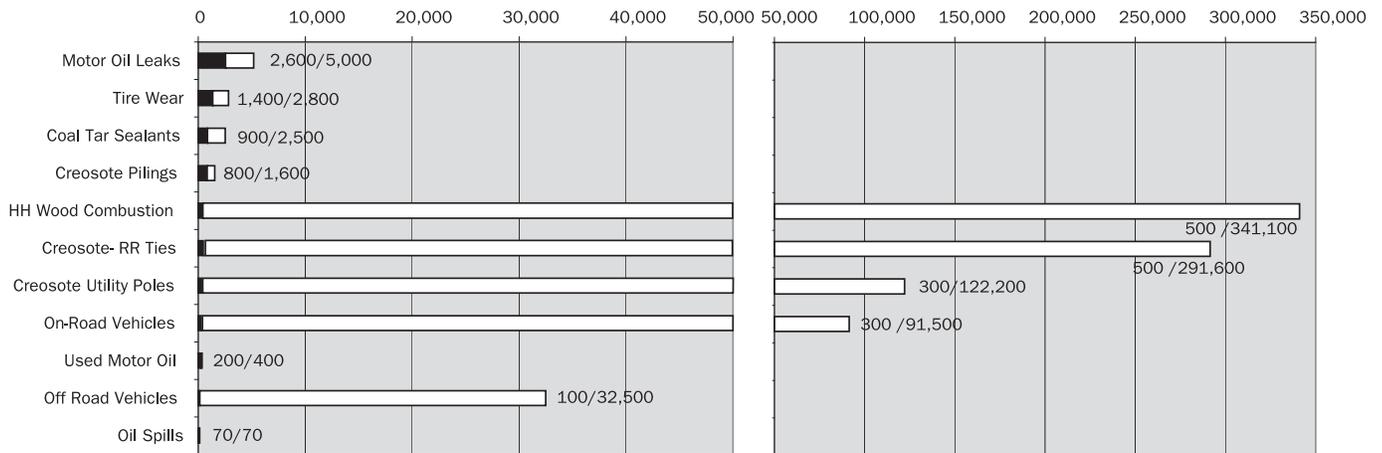
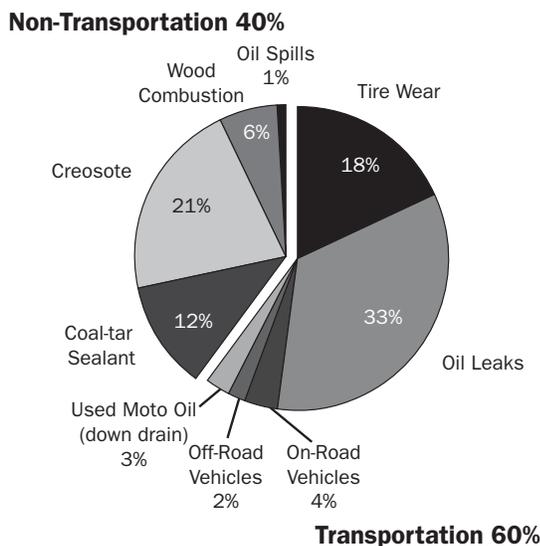


Figure 5. Estimated relative loadings of PAHs to the Harbor from major PAH sectors in the Watershed



through various transport mechanisms. This mass balance considers inputs of PAHs from tributaries, atmospheric deposition, wastewater treatment plant discharges, combined sewer overflows (CSOs), stormwater runoff, and oil spills. It also considers outputs of PAHs via advection of dissolved or suspended sediment-bound PAHs into the coastal Atlantic Ocean or Long Island Sound, volatilization of dissolved PAHs into the atmosphere, and removal of sediment-bound PAHs via disposal of dredged sediments outside the

NY/NJ Harbor. A summary of the mass balance can be found in APPENDIX B. Additional details are expected to be available in a future publication. Information can now be obtained from Lisa Rodenburg.⁸

Although the PAH mass balance for the Harbor identified six loading mechanisms, most of the loadings are due to stormwater/CSO (53%) and tributaries (23%). We therefore compare our loadings, by loading mechanism (as described in the POTENTIAL FOR PAHs TO REACH THE HARBOR section of this report), with the loadings from stormwater, CSOs, and tributaries presented in the PAHs mass balance. A range of loading values is given by the mass balance to account for variability in the monitoring data and in estimation of flowrates.

Loadings were estimated by applying fate and transport factors to estimated primary releases of PAHs to air, land, and water for each of the 16 priority PAHs for each county in the region.

Our estimated loadings from stormwater and tributaries are consistent with the range of inputs calculated in the mass balance (FIG. 6). Our total estimated loadings via stormwater are in the middle of the range of inputs presented in the mass balance. We have not estimated Harbor loadings via wastewater, including the wastewater contribution to CSO outflows, which are included in the mass balance CSO loading estimate. Estimated loadings from the tributaries are also consistent with the mass balance, although the industrial ecology–transmission approach estimate is at or near the bottom of the range for the mass balance inputs.

8. Lisa Rodenburg, Rutgers, The State University of New Jersey, totten@envsci.rutgers.edu.

Estimated loadings to stormwater, CSOs, and surface water are dominated by transportation-related activity and releases from surfaces sealed with coal tar sealants (FIG 7). Loadings from transportation-related activity are dominated by tire wear and oil leaks. These sources are relatively low in total emissions, especially relative to atmospheric emissions, but have

high transmission efficiency, since they all involve direct emissions to an impervious surface, allowing high stormwater transport potential. The releases directly to the Harbor are similar in quantity to estimated loading from the tributaries; however, these loadings are dominated by emissions from creosote-treated pilings.

Figure 6. Estimated PAH loadings (solid bars) vs. range of PAH loadings estimated in the mass balance (hollow bars)

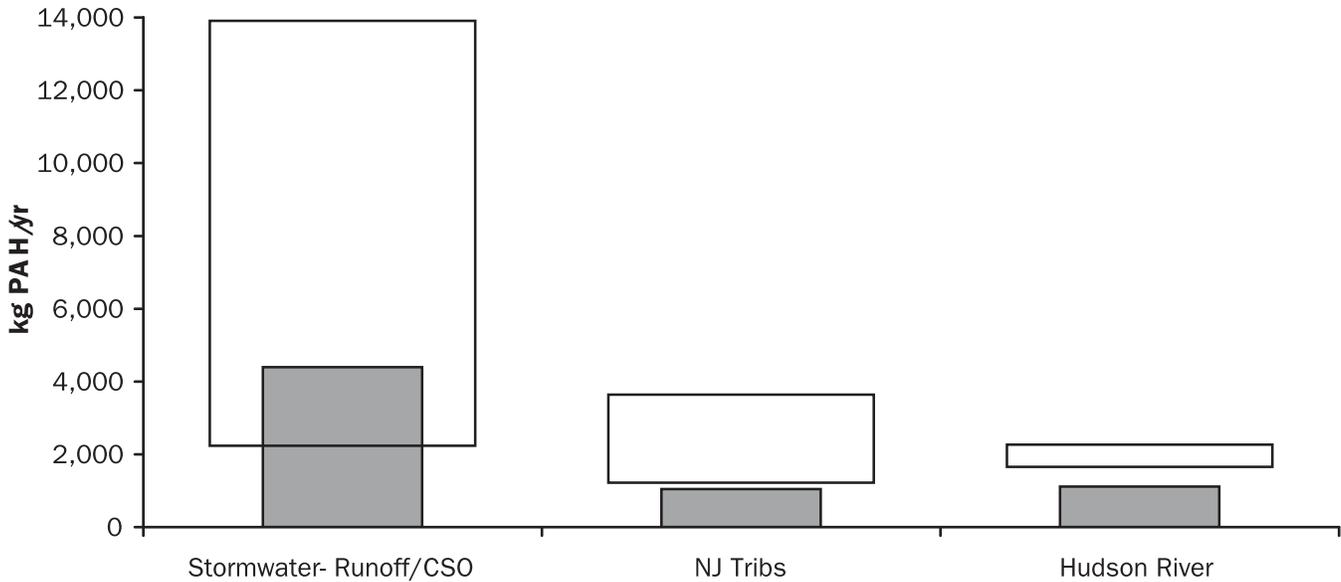
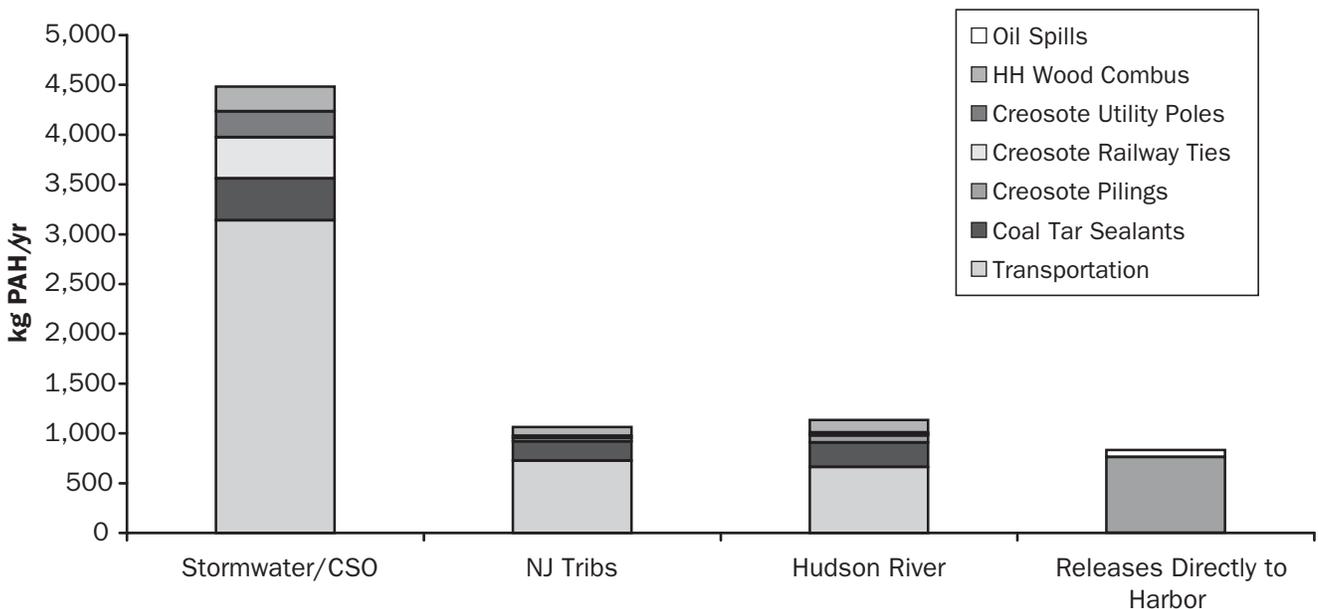


Figure 7. Estimated loadings of PAHs from sources contributing PAH stormwater, tributaries, and directly to the Harbor



Note on Major Sources of Benzo[a]pyrene

Many PAH compounds have been found to cause adverse effects on humans and ecosystems. However, benzo[a]pyrene (BAP), along with a few other PAHs such as naphthalene, are considered possible or likely human carcinogens, and have been targeted as priority compounds for which releases should be reduced. BAP does not break down easily in the environment, and, like many hydrophobic organic contaminants, can accumulate up the food chain in fatty tissues. BAP is included in the U.S. EPA's Persistent Bioaccumulative and Toxic (PBT) initiative.⁹ Some regions have taken direct action to reduce releases of BAP, such as the Great Lakes Binational Toxics Strategy: Canada–United States Strategy for the Virtual Elimination of Persistent Toxic Substances in the Great Lakes (GLBTS). The GLBTS is an ongoing program seeking to reduce releases of persistent toxic substances including BAP.

Our analysis indicates that major primary BAP emission sources in the Harbor region are household wood combustion and the use of coal tar sealants. Taking fate and transport factors into account, the three largest sources of BAP loadings to Harbor waters are the use of coal tar sealants, tire wear, and household wood combustion (FIG. 8).

C. Summary of Findings and Recommendations for Sectors Affecting the Harbor

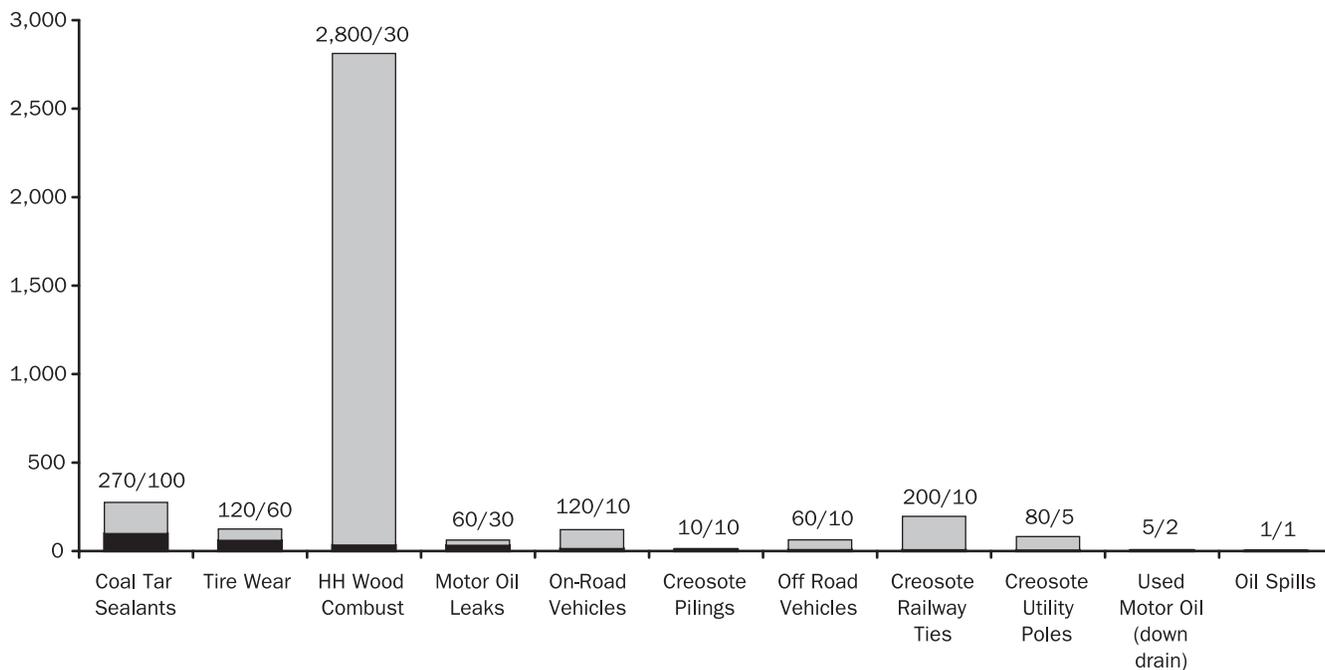
Below is a summary of our findings for the major sources of PAHs in the Watershed region, listed in order of total quantity of PAHs released per year, and specific P2 and BMP recommendations to curb releases from these sources.

C.1. Combustion Sources

Residential Wood Combustion

The major residential heating fuels in the U.S. are natural gas, fuel oil, electricity, coal, wood, and kerosene. In 2001, the Energy Information Administration reported that residential wood combustion was approximated only 6% of total energy consumed for heating. However, in the Watershed, releases of PAHs from residential wood combustion are almost 30 times larger than releases of PAHs from residential gas, oil, and coal combustion combined. Wood is typically combusted in a fireplace, wood stove, or outdoor wood boiler (OWB). While fireplaces and woodstoves may have pollution control devices, OWBs are not regulated.

Figure 8. Estimated releases of BAP (kg/yr) from the major sources of PAHs in the Watershed (gray) compared with estimated loadings (black)



9. For more information on PBTs, visit the EPA web site at <http://www.epa.gov/pbt/>.

In the Watershed, the percentage of homes using wood combustion as their primary heating source is less than 1%. However, PAH emissions from this source contribute to over one-third of the total releases from the major sources of PAHs in the Watershed. We estimate that approximately 341,200 kg of PAHs are released every year by the combustion of wood in fireplaces and wood stoves, most of which occurs in conventional wood stoves that do not utilize pollution controls. An emission factor for PAHs released from OWBs was not available at the time of this report. It is probable that releases of PAHs from OWBs are a significant source of PAHs in the Watershed region.

Summary of Recommendations for Residential Wood Combustion

- Reduce the consumption of heating fuel (wood) by properly sizing the heating unit for the heated space; by implementing residential energy efficiency measures such as increased insulation, sealing cracks, and passive solar design; and by the use of non-PAH-releasing heating fuel. Consider using outreach as well as regulations to promote these measures.
- Optimize combustion conditions by educating the community on the proper use and maintenance of wood combustion units.
- Consider increasing emission standards for wood burning units to reflect the best available control technology. Promote the penetration of these units in the market through changeout and incentive programs.
- It is likely that outdoor wood boilers release a significant amount of PAHs, given that they are typically larger, combust more material, and are not regulated. Research should be conducted on the quantity of PAHs emitted from combusting wood in outdoor wood boilers.

C.2. Materials Containing PAHs

Creosote-Treated Wood

Creosote, a distillate of coal tar, is a probable human carcinogen and is commonly used to waterproof and preserve wood. The only U.S. EPA registered use of creosote-treated wood is in commercial applications. Approximately half of creosote-treated wood manufactured in the U.S. is used for railroad ties, while 30% is used for utility poles, and only a small fraction is used for marine pilings (0.17%) [5]. Creosote is ap-

proximately 80% PAHs by weight (and approximately 20% priority PAHs, as defined by the U.S. EPA). PAHs are released from creosote-treated wood through leaching and volatilization.

Railroad Ties: In the Watershed region, most of the PAHs released from creosote-treated wood are released from on-land applications, railway ties in particular (as opposed to in-water applications, such as marine pilings). Reportedly 95% of ties in use are treated with creosote, with only a small proportion of track segments using concrete ties. We estimate that approximately 291,600 kg of PAHs are released from railway ties in the Watershed per year. The exact proportion of PAHs released to air and land from railway ties is largely unknown. Given the aqueous solubility of individual PAHs, the quantity of precipitation typical for our region, and the geometry and installation of railroad ties, we assumed that 50% of PAH emissions are volatilized to the atmosphere and 50% are leached to pervious land surfaces.

Utility Poles: An estimated 13% of the utility poles in service nationwide are treated with creosote. However, in two New York counties, approximately 40% of the poles in service are treated with creosote. Given the typical geometry and installation of utility poles, and the relatively low water infiltration and air exchange rates expected in densely packed soil versus coarse granular railroad ballast, we have assumed PAHs are lost only from the fraction of a pole's length that extends above the ground surface, and have attributed 100% of those emissions to the atmosphere. Based on these assumptions, we estimate that approximately 122,200 kg of PAHs are released to the air every year as volatile emissions from creosote-treated poles.

An informal phone survey of electric distributors in the Watershed, conducted by the Harbor project staff, revealed that approximately 9% of distributors are replacing retired utility poles with creosote-treated poles. Most distributors are replacing poles with pentachlorophenol-treated poles; pentachlorophenol is a compound associated with dioxins. Most retired phone poles and railway ties are combusted in a cogeneration facility. However, some are donated or given to the public where they cannot be traced and managed.

Marine Pilings: These pilings are exposed to land, water, and air, and the rate at which PAHs are released is influenced by each of these media. The quantity of

pilings in the Watershed is difficult to estimate, given the many types and sizes of aquatic facilities (e.g., private, public, commercial marinas). Based on the best available information, it is estimated that approximately 1600 kg of PAHs are released from marine pilings to air and water every year in the Watershed.

Summary of Recommendations for Creosote-Treated Wood

While this report was under development, New York and New Jersey passed legislation banning the sale and use of creosote-treated wood state wide, with exemptions for creosote-treated wood used in railway and power pole/utility applications. The laws also ban the combustion and disposal in an unlined landfill¹⁰ of all creosote-treated products.¹¹ The recommendations presented below reflect the conclusions of the Harbor Consortium that were reached based on the data presented in this report, and are separate from the legislation described above. They may also be applicable to other marine environments.

Because our focus is on the reduction of all PAHs reaching the Harbor, we make the following recommendations:

- Where feasible (in terms of cost, availability, and performance), avoid the use of creosote-treated wood. As noted earlier, we are not in a position to recommend alternatives, although further research comparing alternatives, such as a life-cycle assessment, would be optimal.¹²
- If creosote-treated wood is used for aquatic applications, use wood that has been treated, transported, and installed according to the Best Management Practices (BMPs) for the use of treated wood in aquatic and other sensitive environments developed by the Western Wood Preservers Institute (WWPI), Wood Preservation Canada, the Southern Pressure Treaters Association, and the Timber Piling Council.
- Consider using BMPs (similar to those referenced above) if creosote-treated wood is used for land applications (railway ties and utility poles). (The WWPI BMPs for creosote-treated wood were developed specifically for wood

used in aquatic and other sensitive environments. It is likely, however, that these BMPs are also appropriate for wood used in terrestrial applications.)

- Do not distribute retired treated wood to the general public.
- Educate the public and, particularly, utility and railway employees who may distribute wood to the public on why it is recommended that wood not be distributed to the public (i.e., hazards associated with burning treated wood in uncontrolled combustion units, and potential exposure of humans and animals to the treated material).
- Evaluate the costs and benefits of removing dilapidated structures constructed with creosote-treated wood. In addition to removing material containing and releasing PAHs, removing dilapidated wood from waterways may improve navigational routes.

Refined Coal Tar Parking Lot Sealants

Many parking lots and driveways are made of asphalt pavement and may be sealed with either asphalt or refined coal tar sealants. Although there is no statistical or quantifiable evidence that pavement sealers extend the life of the pavement, they typically are applied to asphalt substrates every one to five years to protect the pavement from degradation caused by weather, motor oil spills, and traffic. These dark black sealants are also applied for aesthetic purposes. Refined coal tar sealants containing up to 20% total PAHs¹³ are primarily used in the eastern U.S. PAHs may be released from the sealed surface primarily through volatilization (especially the lower molecular weight PAHs) and through abrasion of the sealant from vehicular traffic. Asphalt sealants contain up to 0.66% total PAHs and are used in the western U.S. Pavement sealers, refined coal tar sealers particularly, have only recently been identified as a potentially significant source of PAHs; however, the sale and use of refined coal tar sealants has already been banned in Austin, Texas, and Dane County, Wisconsin. In addition to regional regulations, home improvement retailers such as Lowe's and

10. Such as a landfill that is not properly lined to prevent groundwater contamination.

11. The New York legislation also prohibits the manufacture of products containing creosote and has an exception for wood burned in a permitted facility.

12. The Creosote Council, a participant in the development of this report, did not agree with all of the report's recommendations (see footnote to recommendations on creosote-treated wood in the Technical Report). Nevertheless, all the members of the Consortium approved the recommendations by consensus.

13. Total PAH refers to the following compounds: naphthalene, 2-methylnaphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[a]anthracene, chrysene, benzo[a]pyrene, and dibenz[ah]anthracene.

Home Depot have discontinued the sale of this type of sealant at their stores nationwide.

Based on the estimated volume of refined coal tar-based sealant sold in the region, and the quantity of sealant typically applied to a square meter of surface area, we estimate that between 7.8 and 10.4 million square meters are sealed every year. However, a given area may be resealed as seldom as once every five years, so it is possible that regional sales correspond with an area of sealed surface of up to 50 million square meters, or approximately 0.1% of the harbor drainage basin land surface area. Based on the yields calculated in a peer-reviewed study by Mahler et al. [1] and the estimated range of sealed surface area in the Watershed, we estimate that between 900 and 5800 kg of particulate-bound PAHs are released per year from surfaces sealed with coal tar sealants in the Watershed. Using a wear rate provided by the City of Austin (unpublished), we estimate that between 8000 and 40,000 kg of PAHs are released per year (to land and air, combined) from surfaces sealed with coal tar sealant in the Watershed. Because the City of Austin study was not published at the time of this report's publication, throughout our report we use only estimates that are based on the published yield rates.

PAHs from coal tar sealants are released directly onto impervious surfaces, making the transmission of PAHs to the Harbor much more efficient.

Summary of Recommendations for Refined Coal Tar Sealants

- Implement urban-form measures or policies that reduce the need for parking lot areas, such as increasing mass transit services, increasing pedestrian and biking areas, and providing incentives for current and future drivers that reduce or eliminate miles driven.
- Advise architects, developers, homeowners, and decision makers to avoid the use of sealants containing PAHs and to consider alternative designs and paving materials for certain surfaces.
 - Residential driveways. Alternatives include no- or low-PAH sealants, gravel, or pervious concrete. These efforts should be focused on suburban areas that typically have more driveways and parking lots.
 - Commercial parking lots. Alternatives include no- or low-PAH sealants, concrete, pervious concrete, placing parking lots

beneath structures where they will not be exposed to stormwater transport, and reducing the overall paved area of the project.

- Determine through scientific studies whether pavement sealants extend the life of the asphalt parking lot/driveway and other surfaces onto which sealants are applied. Research the benefits and environmental impact of using alternative products such as concrete driveways, no- or low-PAH sealants, or asphalt-based sealants versus using no sealants.
- Promote the development of a specification for the manufacture of asphalt-based sealants that results in a consistently lower concentration of PAHs **AND** a minimum level of performance (e.g. a lifetime of at least five years).

C.3 Transportation-Related Releases

PAHs are released by transportation-related activities through combustion of fossil fuels and from the materials that are used during the operation of the vehicle. Refined coal tar sealed parking lots could also be considered under this category of sources, although for this discussion we have chosen to include sealants in the "Materials Containing PAHs" category.

Approximately seven million passenger vehicles are registered in the Watershed. In New York alone, vehicle miles driven are expected to increase by 14% from 2002 to 2010. As a group, transportation-related releases are the third largest source of PAHs in the Watershed region, with most of the releases due to vehicle exhaust (FIG. 9).

Summary of Transportation Recommendations (Overall)

The Watershed region, like many other areas, relies on the efficient transport of goods and people from place to place for a vibrant economy and quality of life. This mobility, however, is not without environmental consequence. Reducing PAH releases from this source will require pollution prevention measures for transportation demand as well as for PAH point of release (e.g., fuel combustion). The following are general P2 recommendations aimed at reducing vehicular transportation:

- In dense urban communities, increase infrastructure capacity of public transportation, while establishing measures that deter vehicle use and generate revenue for improved mass

transit options, such as pollution taxes or congestion pricing. Promote the development or redevelopment of communities that provide pedestrian access to surrounding communities via pedestrian and bike pathways.

- In rural and suburban areas, design communities that foster public transportation, bicycling, and walking, possibly through the development of smaller economic centers in tandem with incentives to live close to the center of town.
- In urban and rural communities, prioritize infill projects that capitalize on existing municipal infrastructure and do not require the development of undeveloped land.

Vehicle exhaust: PAH emissions from vehicle exhaust are the result of the incomplete combustion of gasoline or diesel. The quantity of PAHs released depends on the fuel combusted, vehicle class (e.g., light duty, heavy duty, or motorcycle), and the mode in which the vehicle travels. For example, a recent study (Shah et al. [6]) indicates that the average PAH emission rate for diesel vehicles driving in congestion is approximately an order of magnitude higher than for diesel vehicles cruising.

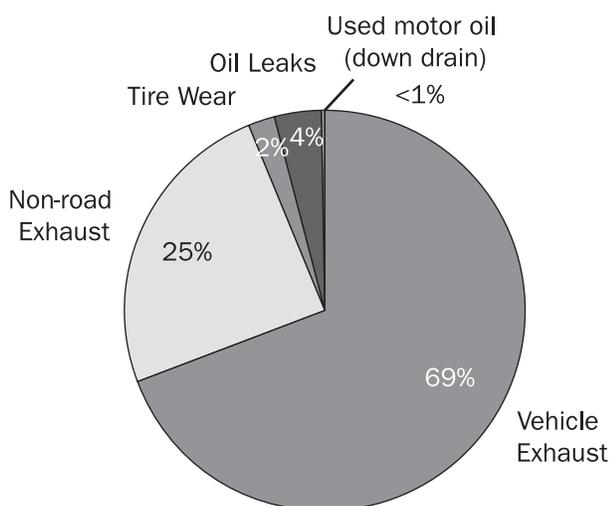
We estimate that approximately 91,500 kg of PAHs are released in the Watershed every year from vehicular exhaust. Although vehicles travel all over the Watershed,

approximately one-third of the activity takes place in the Watershed counties directly bordering the Harbor. Given the population density of this area, vehicles are often traveling in stop-and-go traffic, potentially releasing more PAHs than if the miles were traveled on uncongested highways. Our emission estimates indicate that gasoline vehicles are the vehicle class contributing the most PAHs from this source category

Summary of Recommendations for Vehicle Exhaust

1. Reduce fuel combustion by establishing higher fuel efficiency standards for vehicles; transforming the fleet of vehicles in the Watershed (public and private) to more fuel efficient and/or hybrid models through a combination of legislation, incentives (e.g., free parking for hybrids, tax breaks for the purchase of fuel efficient vehicles), and education; enforcing current anti-idling laws; and educating the community on the environmental burdens associated with vehicle exhaust.
2. Improve vehicle performance by promoting and investing in innovative vehicle design, such as reduced drag aerodynamic designs and lightweight materials, and by promoting clean diesel technology research.

Figure 9. Estimated contribution of PAH emissions from transportation-related activity in the Watershed



Tire wear: Vehicles release PAHs through particulate matter that is worn away from tires as the vehicle is driven.¹⁴ PAHs in tires can be attributed to naphthenic and aromatic extender oils that are added to improve stress resistance, and that contribute to tire tread grip, wear, and endurance qualities. Tire tread can contain 17 to 357 mg PAHs per kg of tire tread, or approximately 160 to 500 mg of PAHs per tire. The quantity and size of particles released depend on vehicle type, terrain over which the vehicle travels, how the vehicle is driven, and tire qualities. Tire wear rates can range from 96 mg of tire particulate per mile traveled (for motorcycles) to 656 mg of tire particulate per mile traveled (for heavy-duty vehicles). It is also possible for PAHs to volatilize from the tire, especially when the tire comes in contact with high temperature surfaces. So far, however, only low levels of gaseous PAHs have been observed [7] (as cited in [8]).

We estimate that approximately 2800 kg of PAHs from tire wear are released to the land surface of the Watershed every year. Because the miles driven are

14. Brake dust is also a source of PAHs; however, an emission factor was not available.

expected to increase, it is likely that releases from this source will also increase. The European Commission has decided to restrict the PAH content in tires; starting in 2010, they will require extender oils used to manufacture tires to have a maximum concentration of 10 mg PAHs¹⁵ per kg or approximately 5 to 7 mg PAHs per tire. Companies manufacturing tires both in the EU and in the U.S. will likely play a role in the market penetration of lower-PAH tires in the U.S. once the appropriate extender oil alternatives have been established in the EU.

Summary of Recommendations for Tire Wear

Research substitutes for high-PAH extender oils that are currently used in tires (potentially capitalizing on the research that has begun in the European Union) while continuing to meet tire safety standards.

- Reduce tire wear rates by educating the driving community on proper tire maintenance, including proper tire inflation, vehicle alignment, and the importance of allowing adequate time to brake. Support the investigation of advanced tire designs that reduce tire wear rate.

Motor oil leaks: Motor oil leakage from vehicles is typically released onto pavement, and although some of the motor oil released may be absorbed into the pavement, some of it will be transported readily across impervious surfaces. There is limited information on the number of vehicles leaking motor oil in the U.S; therefore, there is uncertainty in our estimated releases. The American Petroleum Institute estimates that approximately 70% of motor oil consumed results in used motor oil. The remaining 30% is either leaked or lost to combustion (another source of PAHs, although not quantified in this report). One source reports that 46% of vehicles leak hazardous fluids, including motor oil [9].

We estimate that approximately 5000 kg of PAHs are released per year to the region's land surface from leaking passenger vehicles in the Watershed. Leaks from commercial trucks and vehicles are not included in this estimate and likely contribute to releases of PAHs from this source as well.

Summary of Recommendations for Motor Oil Leaks

- Reduce undetected oil leaks through education and outreach to the driving community

on how to check for oil leaks; incorporate oil leak testing and remediation into the standard vehicle emissions testing currently required by the state.

- Reduce the need for motor oil through innovative vehicle design that requires less lubrication.

Improper disposal of used motor oil: In 2004, the U.S. demand for motor oil was approximately one billion gallons. Of this, approximately 70% results in used motor oil (see above). Motor oil that has been used in a gasoline engine dramatically increases in PAH content (up to 4% to 8% polyaromatic plus 2% to 5% diaromatic). Survey data indicate that approximately 16% of the population who change the oil in their own cars (do-it-yourselfers) do not dispose of used motor oil properly. Improper disposal includes dumping it on the ground, down sewer or sink drains, or placing it in the garbage where it will end up in landfill.

We estimate that the following quantities of PAHs are released in the Watershed every year from improper disposal of used motor oil: 1800 kg to landfill, 400 kg to the ground, and 400 kg down drains. Overall, our estimates indicate that approximately 8% of the used motor oil generated in the Watershed is disposed of improperly.

Summary of Recommendations for Used Motor Oil

- Increase collection of used motor oil by elevating awareness of existing used motor oil collection services through outreach and education campaigns that target vehicle and nonroad equipment users as well as educational institutions. Materials to be distributed should include information on local collection facilities (e.g., vehicle service centers, municipal collection sites, and municipal collection days), and about the hazards of improper disposal. Solicit the support of industry and government to implement these initiatives.
- Increase collection of used motor oil by providing incentives, such as a deposit or coupon program for motor oil that is returned to a collection facility.
- Facilitate access to used motor oil collection services by increasing the number of retail stores collecting used motor oil through industry sponsored partnerships.

15. The PAH concentration maximum refers to the following PAHs: benzo(a)pyrene, benzo(e)pyrene, benz(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(j)fluoranthene, and dibenz(a,h)anthracene. A maximum PAH concentration in tire extender oils of 1 mg/kg has also been established for benzo(a)pyrene.

- Increase the end-use market for used motor oil by promoting the use of re-refined motor oil through government and corporate procurement programs and outreach initiatives.
- Reduce consumption of motor oil through proper vehicle maintenance and changing oil only when needed. Consider the use of synthetic motor oil that has a longer change interval than traditional motor oil.

Nonroad engine exhaust: Nonroad emissions include exhaust released from any class of two- or four-stroke gasoline or diesel engine, such as recreational, lawn and garden, construction, industrial, commercial, logging, airport maintenance, and agricultural equipment. Two-stroke engines have been found to release more PAHs than four-stroke engines, likely due to the lack of valves or emissions controls.

Nonroad equipment also generates used motor oil, specifically equipment with a four-stroke engine, and tire wear. The quantity of PAHs released from these sources is unknown, and it is possible that these are noteworthy sources of PAHs.

We estimate that approximately 32,500 kg of PAHs are released per year from engines in nonroad equipment within the Watershed region, mostly from lawn and garden equipment. Commercial and construction equipment, such as pressure washers, generators, and cranes, also release a large fraction of PAHs in this source category. The relative contribution of PAHs from the different equipment types varies within the Watershed region. For example, in rural communities most nonroad PAH emissions are from agricultural equipment, whereas in urban centers construction and industrial equipment predominate.

Summary of Recommendations for Nonroad Exhaust

While several types of nonroad equipment utilize internal combustion engines, lawn care, construction, and commercial equipment are estimated to contribute the most PAHs from this source category. Furthermore, construction and commercial activity are highest in the counties directly surrounding the Harbor. Therefore, our P2 recommendations focus on these two categories.

Lawn Care Equipment

- Promote the use of best available equipment through voluntary or legislative measures. For example, consider the implementation of

a voluntary changeout program that would facilitate the exchange of older, less efficient equipment for newer, more efficient engines, such as those utilizing a catalytic converter or an electric motor.

- Reduce the need for lawn equipment by providing incentives for low-emissions landscape design and maintenance such as a program that recognizes landscapers who reduce their emissions through design (e.g., by growing land cover that does not need to be mowed) and maintenance (e.g., by using no- or low-emission equipment). Homeowners should also be informed about emissions associated with yard care and about alternative landscape designs that demand little or no equipment for maintenance.

Construction and Commercial Equipment

- Consider the implementation of statewide emissions control programs that retrofit older equipment with pollution reduction technologies.
- Educate equipment owners on the importance of proper equipment maintenance and how to maintain equipment properly to ensure maximum fuel combustion performance. Consider addressing the following maintenance issues: restricted air filters, improper engine timing, malfunctioning fuel injectors, defective air fuel controllers, and poor fuel quality.
- Reduce unnecessary idling of equipment by extending idling restrictions to nonroad engines and through the promotion of idling reduction technologies such as automatic shut-off devices.

C.4 Petroleum Spills

The PAH content of crude oil can vary dramatically depending on the source rock (i.e., marine- or terrestrial-derived organic deposits) and the thermal environment prevailing during oil formation. Crudes with the highest PAH content tend to come from marine deposits that are found in the Middle East.

It is difficult to quantify the total amount of oil released in a given year. While larger spills are well documented, smaller spills often go unreported. It is estimated that approximately eight million gallons of crude oil (or approximately 14 tons of PAHs) are released to waters nationwide every year.

It is estimated that on average 70 kg of PAHs per year are released in the Harbor by petroleum spills.

This estimate is based on data in the U.S. Coast Guard Marine Information Safety and Law Enforcement (MISLE) database, and is likely incomplete, given that only “closed” cases are reported. Of the spills reported, most have been due to equipment failure. In addition to the spills reported in the MISLE database, several recent, relatively large petroleum spills in the Arthur Kill, Passaic River, and Rahway River, may have contributed another 192 kg of PAHs to the Harbor.

Very large hydrocarbon spills may become partially trapped in soil or sediment, and slowly release PAHs to the Harbor over many years. Quantifying the annual inputs of PAHs from individual historically contaminated sites was not possible. However, PAH inputs from this source are likely to be an important contributor of PAHs to the Harbor, given the number of contaminated sites in the region and the magnitude of contamination at certain individual sites.

Summary of Recommendations for Petroleum Spills

The available data indicate that most spills are the result of equipment failure. Therefore, our recommendations focus on improving preventive maintenance at the facility level.

- Consider strengthening the equipment failure and maintenance sections of the Spill Prevention Control and Countermeasures Plan federally required for owners or operators of facilities who drill, produce, gather, store, process, refine, transfer, distribute, use, or consume oil and oil products.
- Increase employee spill prevention knowledge through training and continuing education efforts.
- Support an expeditious and thorough cleanup of the historical petroleum spill along Newtown Creek (a tributary of the East River) that continues to seep into the creek.

Approach Used to Calculate Releases and Loadings and Cautions on Estimate Use

For most sources, emission estimates were calculated by applying emission factors for each source and me-

dium of release to area-specific activity levels. The emission factors that we used allow for a quantitative, compound-specific measure of PAH releases per unit of activity (i.e., mg pyrene released per kg hardwood burned or vehicle mile traveled). We emphasized using emission factors from peer-reviewed publications whenever possible. In some cases, when emission factors were not available, emission values reported to the U.S. EPA’s Toxics Release Inventory (TRI) were used.¹⁶ For example, emission factors for petroleum refineries were not available; therefore, emissions from refining facilities in the Watershed reported to the TRI are presented in this report.

Activity (i.e., kg wood burned or vehicle miles traveled per county per year) was quantified from regional activity data whenever possible. However, when necessary, we extrapolated from state or national activity data based on Watershed county population or a reported correlated activity expected to represent desired activity. For example, train track miles for New York and New Jersey were available; therefore, the miles of train track in the Watershed was extrapolated based on reported locomotive PM₁₀ emissions. Although not all counties are 100% in the Watershed, emissions calculated for diffuse/nonpoint sources were typically made for all counties that are entirely or partially in the Watershed.

The relevant region for releases to water and land is the New York/New Jersey Harbor Watershed; however, with regard to atmospheric sources, emissions both from inside and outside the Watershed (New York and New Jersey only) are estimated. Experts consulted concluded that considering atmospheric sources within the entire states of New York and New Jersey would likely capture most of the atmospheric emissions capable of reaching the Harbor.¹⁷

PAH releases are reported in kilograms per year. Release estimates represent order of magnitude estimates of releases for a given year and are not necessarily representative of the most recent year. All ton units are reported as metric tons (1000 kg).

Although efforts were made to use the best available data when calculating PAH releases and loadings, caution should be used when viewing these estimates. Three of the primary uncertainties with this report are 1) omission of potentially major sources of PAHs

16. The TRI is a publicly available U.S. EPA database that contains information on toxic chemical releases and other waste management activities reported annually by certain covered industry groups as well as federal facilities. Reporting to the TRI is triggered by how much of the chemical is manufactured, processed, or otherwise used during the year.

17. On August 4, 2005 we held a consultative meeting with experts on the issue of long-range transport of PAHs and other toxics. For a detailed discussion on this meeting, see page 40 of the report *Pollution Prevention and Management Strategies for Dioxins in the New York/New Jersey Harbor*, New York Academy of Sciences (2006).

to the Harbor; 2) lack of robustness of emission factors and activity estimates; and 3) limitations inherent in the simplifying assumptions included in the fate and transport modeling. Specific sources known or suspected to emit PAHs in the region but not included in this report include historically contaminated sites, brake dust, and outdoor boilers (see Section 3.6 SOURCES FOR WHICH EMISSION FACTORS ARE NOT AVAILABLE). PAH emission factors are available for many sources; however, they do not always characterize emissions of all 16 EPA priority PAHs and/or have associated uncertainties that transcend and impact calculated emissions. For example, the quantity of PAHs released in a combustion process is a function of oxygen concentration, temperature, and characteristics of the materials being combusted. Neither this degree of detail in emission factors, nor the corresponding detail in activity factors for each type of combustion conditions in a given location in a given year, are generally available. The limitations of the fate and transport modeling are discussed in APPENDIX C. But in spite of all these limitations, this report provides an assessment of PAH sources to the Harbor that is truly unprecedented in scope, and serves as a model for better understanding the key sources and transmission vectors of nonpoint source persistent and bioaccumulative toxins in modern, urban settings.

TECHNICAL REPORT

INTRODUCTION

The fifth class of contaminants chosen to be investigated by The Harbor Consortium was polycyclic aromatic hydrocarbons, a class of contaminant with both pyrogenic and petrogenic sources. PAHs elicit concern for several reasons: they are directly toxic to marine animals; they are harmful to humans; and PAH metabolites are potent animal and human carcinogens. Recent trends indicate an increase in PAH contamination in U.S. waterways in or near urban areas [10] [11]. In the New York/New Jersey Harbor region specifically, high concentration of PAHs in aquatic sediments have been documented [12] [13]. In addition, elevated concentrations of several PAHs have been detected in aquatic organisms found in the Watershed, including blue crab, perch, oysters, and mussels [14] [15].

The objective of this report is to identify the primary sources of PAH releases within the New York/New Jersey Harbor estuary, understand how these releases might reach the Harbor, and make recommenda-

tions aimed at stemming PAH loadings to the Harbor. An industrial ecology approach was used to identify sources and to estimate total emissions of PAHs from all known sources throughout the Harbor region. Subsequently, a semiquantitative analysis of the fate and transport of PAHs from the primary sources to the Harbor was conducted and a comparison with the PAH mass balance was made as a means of constraining our estimated values. It should be noted that this report does not attempt to estimate PAH availability to biota or environmental impact associated with each of the individual sources.

In the following sections we attempt to provide a comprehensive discussion on PAHs and their properties, emission sources (both national and regional), major sources of PAH releases within the Harbor watershed, and the fate and transport characteristics within the Harbor complex used to estimate loadings of PAHs to the Harbor.

1. GENERAL BACKGROUND ON PAHS

1.1. Physical and Chemical Characteristics

Polycyclic (or polynuclear) aromatic hydrocarbons, or PAHs, are a class of compounds characterized by two or more fused aromatic rings composed of carbon and hydrogen [16]. PAHs are found naturally in petroleum deposits and are the product of the incomplete combustion of organic matter. Elevated PAH concentrations can be found in urban air, in soil and groundwater adjacent to certain industrial operations, and, especially, in sediments underlying industrialized ports and waterways. While hundreds of different PAHs exist, 16 compounds were designated as “priority pollutants” by the 1977 Clean Water Act because of their toxicity and status as known or possible human carcinogens [17].¹⁸

The PAHs selected for investigation in this report, along with their chemical formulas and molecular weights, are listed in TABLE 1.1. These 17 compounds

include the 16 U.S. EPA-designated priority PAHs as well as perylene.¹⁹ They represent a wide range of molecular weights, which are closely related to the physical properties of the compounds. These PAHs (except naphthalene and acenaphthene) were measured in all three of the datasets used to construct the mass balance (see APPENDIX B). Naphthalene is frequently the most abundant PAH in the dissolved phase, and it is designated by the U.S. EPA as a possible human carcinogen [19].

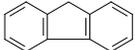
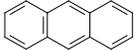
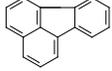
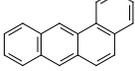
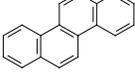
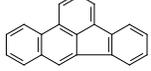
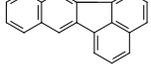
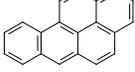
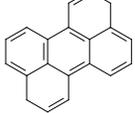
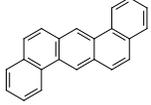
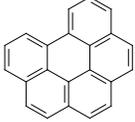
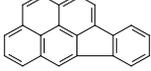
The physical-chemical properties of PAHs cause them to behave in the environment like other hydrophobic organic contaminants, including polychlorinated biphenyls (PCBs) and dioxins [16]. PAHs can bioaccumulate in fatty tissues, and one compound, benzo[a]pyrene, is on the U.S. EPA’s list of Priority Persistent and Bioaccumulative Toxins (PBT) [20].²⁰ These types of organic contaminants tend to accumu-

18. The 16 U.S. EPA priority PAHs are naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, dibenz[a,h]anthracene, benzo[g,h,i]perylene, and indeno[1,2,3-CD]perylene.

19. Oxygenated and methylated PAHs have the potential to be more toxic than their parent PAHs [18]. It is our assumption that presence of the parent PAHs addressed in this report indicates the presence of these metabolites.

20. Persistent, bioaccumulative, and toxic (PBT) chemicals do not break down readily in the environment, are not easily metabolized, may accumulate in human or ecological food chains through consumption or uptake, and may be hazardous to human health or the environment.

Table 1.1. Chemical formula, molecular weight (MW), and structure for selected PAHs

| PAH | Chemical formula | Molecular weight (g mol ⁻¹) | Structure |
|------------------------|---------------------------------|--|---|
| Naphthalene | C ₁₀ H ₈ | 128 |  |
| Acenaphthylene | C ₁₂ H ₈ | 152 |  |
| Acenaphthene | C ₁₂ H ₈ | 152 |  |
| Fluorene | C ₁₃ H ₁₀ | 166 |  |
| Phenanthrene | C ₁₄ H ₁₀ | 178 |  |
| Anthracene | C ₁₄ H ₁₀ | 178 |  |
| Fluoranthene | C ₁₆ H ₁₀ | 202 |  |
| Pyrene | C ₁₆ H ₁₀ | 202 |  |
| Benz[a]anthracene | C ₁₈ H ₁₂ | 228 |  |
| Chrysene | C ₁₈ H ₁₂ | 228 |  |
| Benzo[b]fluoranthene | C ₂₀ H ₁₂ | 252 |  |
| Benzo[k]fluoranthene | C ₂₀ H ₁₂ | 252 |  |
| Benzo[a]pyrene | C ₂₀ H ₁₂ | 252 |  |
| Perylene | C ₂₀ H ₁₂ | 252 |  |
| Dibenz[a,h]anthracene | C ₂₂ H ₁₄ | 278 |  |
| Benzo[g,h,i]perylene | C ₂₂ H ₁₂ | 276 |  |
| Indeno[1,2,3-CD]pyrene | C ₂₂ H ₁₂ | 276 |  |

Source: Mackay et al. [16].

late preferentially in hydrophobic compartments of the environment, including organic matter-rich soil and sediment, suspended particles in air and water, and the lipid fractions of biota, owing to the high PAH absorption capacity of these phases. As a result, very little of the total hydrophobic pollutants in the environment are found freely dissolved in water, and one might expect adverse effects to the ecosystem to arise primarily from contact with PAH-contaminated particles.

Hydrophobicity can be measured by determining the distribution of a given compound between octanol and water phases. This parameter is called the octanol-water partition coefficient (K_{ow}) and is often reported as $\log K_{ow}$. The higher the K_{ow} is, the greater the affinity for hydrophobic phases. Many PAHs also have very low vapor pressure, and only the lightest (e.g., naphthalene) are likely to be found in substantial quantities as free PAH in the atmosphere. Based on characteristic differences in physical and chemical properties, the first eight compounds are sometimes referred to as the “low molecular weight” PAHs, while

the last eight (plus perylene) are “high molecular weight” PAHs. The aqueous solubility, octanol-water partition coefficient, and vapor pressure for the 16 priority PAHs and perylene are given in TABLE 1.2.

In general, PAHs become more hydrophobic and less volatile with increasing molecular weight (FIG. 1.1).

1.2. Toxicology and Human Health Effects

PAHs can cause a variety of adverse human health effects, including liver, kidney, and hematologic effects; cataracts; and cancer [19]. Several noncarcinogenic reference doses (RfDs) have been established by the U.S. EPA for chronic exposure to various PAHs via oral and inhalation routes (TABLE 1.3). An RfD is the concentration to which humans (including sensitive subgroups) can be exposed on a daily basis over a lifetime without expecting adverse health effects [21]. The RfD for a given compound and exposure route is calculated from the “no observed adverse effects level” (NOAEL)²¹ from a human or animal study, according to EQUATION 1:

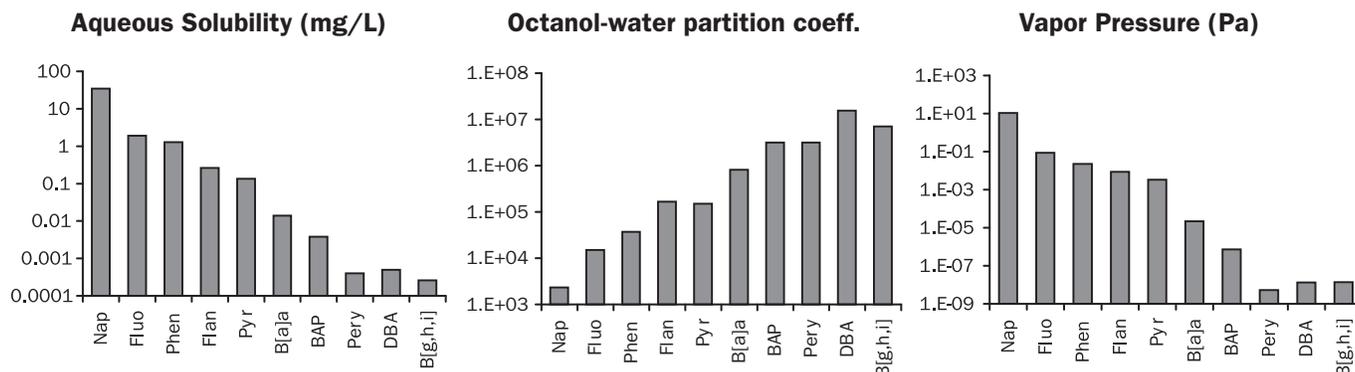
Table 1.2. Physical-chemical properties of selected PAHs including aqueous solubility, log octanol-water partition coefficient, and vapor pressure

| | PAH | Aqueous solubility (mg L ⁻¹) | Log K_{ow} | Vapor pressure (Pa) |
|------------------------|-----------------------|---|--------------|------------------------|
| Low molecular weight | Naphthalene | 34.4 | 3.37 | 10.8 |
| | Acenaphthylene | 3.93 | 4.07 | 0.89 |
| | Acenaphthene | 3.88 | 4.03 | 0.30 |
| | Fluorene | 1.90 | 4.18 | 0.09 |
| | Phenanthrene | 1.29 | 4.57 | 2.27E-02 |
| | Anthracene | 0.073 | 4.54 | 3.60E-03 |
| | Fluoranthene | 0.26 | 5.22 | 8.61E-03 |
| | Pyrene | 0.14 | 5.18 | 3.39E-03 |
| High molecular weight | Benz[a]anthracene | 0.014 | 5.91 | 2.20E-05 |
| | Chrysene | 0.0020 | 5.91 | 5.70E-07 |
| | Benzo[b]fluoranthene | 0.0015 | 6.50 | 5.00E-07 |
| | Benzo[k]fluoranthene | 0.00081 | 6.84 | 5.20E-08 |
| | Benzo[a]pyrene | 0.00380 | 6.50 | 7.32E-07 |
| | Perylene | 0.00040 | 6.50 | 5.31E-09 |
| | Dibenz[a,h]anthracene | 0.00050 | 7.19 | 1.33E-08 |
| | Benzo[g,h,i]perylene | 0.00026 | 6.85 | 1.39E-08 |
| Indeno[1,2,3-CD]pyrene | 0.00019 | 7.66 | NA | |

Log K_{ow} = log octanol-water partition coefficient
Source: Mackay et al. [16].

21. “No observed adverse effects level” (NOAEL) is the highest tested dose of a substance that has been reported to have no harmful (adverse) health effects to humans or animals. In the case of NOAEL, the adverse health effect is known prior to testing.

Figure 1.1. Aqueous solubility, octanol–water partition coefficient, and vapor pressure for selected PAHs



The vertical line separates the low molecular weight PAHs from the high molecular weight PAHs. (From Mackay et al. [16].) Naphthalene (nap), fluorine (fluo), phenanthrene (phen), pyrene (pyr), benz[a]anthracene (b[a]a), benzo[a]pyrene (BAP), perylene (pery), dibenz[a,h]anthracene (DBA), and benzo[g,h,i]perylene (B[ghi]).

Equation 1. The RfD for a given compound and exposure route is calculated from the “no observed adverse effects level” (NOAEL) from a human or animal study

$$RfD = \frac{NOAEL}{UF \cdot MF}$$

UF and MF represent, respectively, the associated uncertainty factors and modifying factors for the study.

In each case, confidence in the parameter is low (or “low-medium” in the case of the naphthalene inhalation reference dose). No RfD values are available for exposure to acenaphthylene, phenanthrene, or any of the high molecular weight PAHs.

Seven priority PAHs including BAP have been designated level B2, for “probable human carcinogen,” and naphthalene is designated level C, for “possible human carcinogen.” Other priority PAHs are designated cancer classification D (not classifiable as to human carcinogenicity; TABLE 1.4) [19].²² Risk from exposure to carcino-

genic compounds is defined as excess cancer risk, or the number of additional cancers that would result from a lifetime of exposure to that contaminant [23]. Excess cancer risk is computed by multiplying the total quantity of contaminant to which an individual would be exposed over a lifetime, given various exposure scenario-specific exposure factors, by a measure of the carcinogenic potency of the compound, called the cancer slope factor (CsF). Data to define a CsF are sufficient for only one PAH, benzo[a]pyrene (BAP) [19]. Excess cancer risk for exposure to other PAHs is computed by multiplying the total exposed dose by the toxic equivalency factor (TEF, also sometimes termed potency equivalency factor or PEF) for that compound, and then applying the CsF for BAP to the resulting BAP-equivalent dose. TEF values reported by Nisbet and LaGoy [24] and provisional guidance to the U.S. EPA [25] (TABLE 1.4) continue to be used and cited widely [26] [27], and are consistent with other PAH toxic equivalency studies [28]. According to both the U.S. EPA and the Nisbet and LaGoy values, the most carcinogenic PAHs besides BAP are dibenz[a,h]anthracene, indeno[1,2,3-cd]pyrene, benzo[b]fluoran-

22. The following information defining cancer classes is available on the U.S. EPA web site, <http://www.epa.gov/iris/>. The cancer class system was used from 1986 to 1996. However, until the system can be replaced with a better approach, U.S. EPA still reports cancer classifications in the Integrated Risk Information System (IRIS) database for all PAHs:

GROUP A—HUMAN CARCINOGEN: used only when there is sufficient evidence from epidemiologic studies to support a causal association between exposure to the agents and cancer.

GROUP B—PROBABLE HUMAN CARCINOGEN: includes agents for which the weight of evidence of human carcinogenicity based on epidemiologic studies is limited and also includes agents for which the weight of evidence of carcinogenicity based on animal studies is sufficient. The group is divided into two subgroups. Group B1 is reserved for agents for which there is limited evidence of carcinogenicity from epidemiologic studies. Group B2 is used for agents for which there is sufficient evidence from animal studies and for which there is inadequate evidence or no data from epidemiologic studies.

GROUP C—POSSIBLE HUMAN CARCINOGEN: used for agents with limited evidence of carcinogenicity in animals in the absence of human data.

GROUP D—NOT CLASSIFIABLE AS TO HUMAN CARCINOGENICITY: generally used for agents with inadequate human and animal evidence of carcinogenicity or for which no data are available.

GROUP E—EVIDENCE OF NONCARCINOGENICITY FOR HUMANS: used for agents that show no evidence for carcinogenicity in at least two adequate animal tests in different species or in both adequate epidemiologic and animal studies [22].

thene, and benz[a]anthracene. Both sources consider dibenz[a,h]anthracene to be at least as carcinogenic as BAP; Nisbet and LaGoy [24] consider dibenz[a,h]anthracene to be five times more carcinogenic than BAP, but data are still insufficient to compute a cancer slope factor for this compound [19].

While direct human exposure to contaminated sediment is unlikely to cause adverse health effects (for ex-

ample, from exposure of recreational swimmers and boaters to contaminated sediment [27]), the human and ecological toxicity of PAHs in sediment triggers restrictions on available options for dredged sediment disposal and thereby constitutes a high financial burden to the region. In addition, some human exposure risks may not be negligible, such as health risks to area fishermen who ignore posted warnings and consume

Table 1.3. All available reference doses for chronic noncarcinogenic adverse health effects from exposure to PAHs via oral and inhalation routes

| PAH | Critical effect (oral) | RfD ^a (mg/kg-day) | Critical effect (inhalation) | RfC ^b (mg/m ³) | Other effects |
|--------------|---|------------------------------|--|---------------------------------------|--|
| Naphthalene | Decreased mean terminal body weight (male rats) | 0.02 | Respiratory hyperplasia and olfactory metaplasia | 0.003 | Hemolytic anemia (oral, inhalation); cataract formation (inhalation) |
| Acenaphthene | Hepatotoxicity | 0.06 | NA | | |
| Fluorene | Decreased RBC & hemoglobin | 0.04 | NA | | |
| Anthracene | No observed effects | 0.3 | NA | | |
| Fluoranthene | Kidney, liver, and blood effects | 0.04 | NA | | |
| Pyrene | Kidney effects | 0.03 | NA | | |

NA = not assessed

Source: U.S. EPA [19].

a For oral ingestion, RfD= noncarcinogenic endpoint reference dose, defined as the concentration to which humans (including sensitive sub-groups) can be exposed on a daily basis over a lifetime without expecting adverse health effects.

b For inhalation, RfC= carcinogenic endpoint reference dose.

Table 1.4. Cancer class and relative carcinogenic potency expressed as toxicity equivalency factors (TEF) for selected PAHs

| PAH | Cancer class | TEF (U.S. EPA) | TEF (Nisbet & LaGoy) |
|------------------------|--------------|----------------|----------------------|
| Naphthalene | C | | |
| Acenaphthylene | D | | |
| Acenaphthene | N/A | | |
| Fluorene | D | | |
| Phenanthrene | D | | 0.001 |
| Anthracene | D | | 0.01 |
| Fluoranthene | D | | 0.001 |
| Pyrene | D | | 0.001 |
| Benz(a)anthracene | B2 | 0.1 | 0.1 |
| Chrysene | B2 | 0.001 | 0.01 |
| Benzo(b)fluoranthene | B2 | 0.1 | 0.1 |
| Benzo(k)fluoranthene | B2 | 0.01 | 0.1 |
| Benzo(a)pyrene | B2 | 1 (index) | 1 (index) |
| Perylene | | | |
| Dibenz(a,h)anthracene | B2 | 1 | 5 |
| Benzo(g,h,i)perylene | D | | 0.01 |
| Indeno(1,2,3-CD)pyrene | B2 | 0.1 | 0.1 |

TEF= toxic equivalency factor

Sources: Nisbet and LaGoy [24]; U.S. EPA. [25]

fish caught in contaminated areas (Shor, unpublished data). While these involuntary risks are likely to be far lower than various voluntary PAH exposure risks (e.g., cigarette smoking, indoor pollution from fireplaces, consuming grilled meats [29]), the purpose of this document is to quantify the major sources of PAH pollution to the Harbor. This document provides the necessary background information for the Harbor Consortium to deliberate on recommendations to curb loadings of PAHs to the Harbor. The ensuing policy recommendations are expected to be consistent with the regional goals of ensuring the economic and ecological vitality of the Harbor.

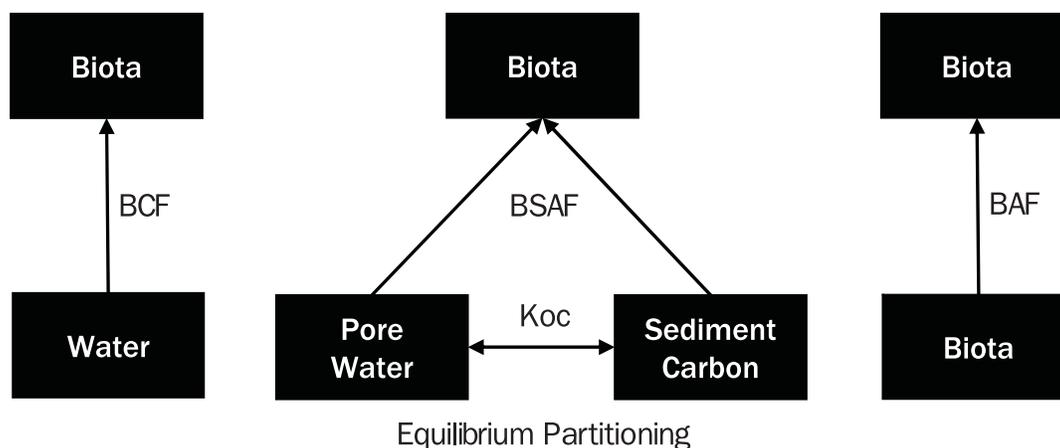
1.3. Ecological Impacts

PAHs are commonly detected in U.S. surface waters [30], in sediments underlying U.S. ports, and in other industrialized waterways [31] [32]. PAH contamination is known to adversely affect the viability of benthic species and the diversity of aquatic, estuarine, and marine ecosystems [33, 34]. Given the importance of estuaries in the life cycle of many marine species, including commercially important fish populations [35-39], the adverse impact to the NY/NJ Harbor caused by PAH contamination extends far beyond its geographic boundaries to include various species and humans.

As with other hydrophobic organic contaminants, PAHs can become concentrated in the lipid-rich tissues of animals and accumulate along the food chain [16]. Three different bioaccumulation parameters are

defined according to the route of exposure: 1) bioconcentration factors (BCF) are for exposure from water; 2) biota-sediment accumulation factors (BSAF) are from exposure to sediment; and 3) biomagnification factors (BMF) are for exposure through trophic levels [40] (FIG. 1.2). Species that live in sediment are exposed to PAHs by contact with and ingestion of PAH-contaminated sediment and pore waters, and ingestion of PAH-contaminated biota. Species that live in the water column are exposed to PAHs by direct exposure to PAH-contaminated water, as well as via trophic transfer. In general, biomagnification is not as pronounced for PAHs as it is for some other persistent and bioaccumulative compounds (e.g., PCBs, dioxins), because many animals have the ability to metabolize and eliminate PAHs from their bodies [41]. In some instances, trophic dilution is observed in predatory organisms as a result of metabolic transformation within organisms and their prey species [42]. However, many studies measure only the parent compound and not PAH metabolites. For example, one study with polychaete worms found some cases where parent compounds represented less than 10% of the total burden of parent compound plus metabolites [43]. Since the mechanism of benzo[a]pyrene (BAP) carcinogenicity to humans begins with sequential oxidation to BAP-7,8-oxide, BAP-7,8-diol, and, ultimately, BAP-diol-epoxide [44], ideally, the concentration of metabolites would also be measured to estimate their potential impact more accurately.

Figure 1.2. Diagram showing different ways biota concentrations are estimated from other types of data, including water, pore water, and biota concentrations via a bioconcentration factor (BCF), a biota-sediment accumulation factor (BSAF), and a biomagnification factor (BMF), respectively



Modified from DiToro et al. [45].

BSAF [dimensionless] is computed by dividing the lipid-normalized PAH concentration in biota [mg PAH/kg dry weigh] by the organic carbon content-normalized sediment concentration [mg PAH/kg dry weight], as below:

Equation 2. Biota-sediment Accumulation Factor

$$BSAF = \frac{\left(\frac{C_{biota}}{\% \text{ lipid}} \right)}{\left(\frac{C_{sed}}{\% \text{ org C}} \right)}$$

For well-defined systems in thermodynamic equilibrium, ecologically protective sediment quality criteria can be computed readily via equilibrium partitioning theory from readily measured parameters, including sediment organic carbon content and toxic effects thresholds for target species [45]. For example, DiToro et al. [46] [47] use equilibrium partitioning theory to generate “final chronic value” (FCV) sediment quality guidelines. An important feature of their narcotic toxicity approach is harmful effects from PAHs, which are assumed to be additive, so the FCV given for individual PAHs (acenaphthene, phenanthrene, pyrene, and fluoranthene) are nearly equal to each other and to the FCV for total PAHs [47].

Equilibrium partitioning is a unified and useful approach, but deviations from equilibrium behavior have often been noted in measured sediment-water-biota partitioning. Factors that lead to nonideal partitioning include selective feeding, biotransformation, and bioturbation by biota [40]; various sediment-related factors such as reduced bioavailability with contaminant aging [48] and differing organic matter properties [49] [50]; and factors related to variability in the PAH-binding ability of dissolved organic carbon (DOC) [51]. In reality, measured BSAF values vary broadly, even within the same region. For example, Baumard et al. [52] reported a range of BAF for mussels (*Mytilus* sp.) in the Baltic Sea of 0.02–52, depending on sampling date, PAH concentration, carbon content of sediment, and lipid content of the organism. Another study reported BSAFs for eight locations in Hoffman Marsh (San Francisco Bay) for several species of benthic invertebrates, including Asian clam (*Potamocorbula amurensis*), Japanese littleneck clam (*Tapes japonica*), and

polychaetes [50]. Total PAH concentrations in sediment at the study sites varied from 10 to 150 mg/kg; however, BSAF values ranged nearly three orders of magnitude (0.0069–5.4), with reduced PAH bioavailability in sediments with a high soot content (from urban runoff) as a determining factor. Given the variability in BSAF values, bioavailability assays [53] [40] or biomarker approaches [54, 55] are probably necessary to generate criteria that account for site-specific differences in contaminant availability. The U.S. EPA has evaluated the status and needs of bioaccumulation information for sediment quality assessment, and lists improving test methods, developing more BSAF values, and better understanding of bioavailability, food chain multipliers, and mixture effects as high priority research areas [56].

Given the difficulty in establishing universal sediment concentration limits, several agencies have established sediment criteria guidelines to identify potential impacts of sediment contamination on coastal resources and habitats. The National Oceanic and Atmospheric Association (NOAA) developed a set of sediment screening concentrations for inorganic and organic contaminants. NOAA reports a sediment screening concentration for total PAHs in freshwater of 12,000 ppm dry weight for upper effects level and in marine waters of 44,792 ppm dry weight for median effects range.²³ The New York State Department of Environmental Conservation, Division of Fish and Wildlife and the Division of Marine Resources, has also established sediment criteria for several PAHs (TABLE 1.5) [57]. These criteria do not necessarily represent the final concentrations that must be achieved through sediment remediation and comprehensive sediment testing. Risk management is necessary to establish when remediation is needed.

23. For more information, visit the NOAA web site at http://response.restoration.noaa.gov/book_shelf/122_squirt_cards.pdf.

Table 1.5. New York State Department of Environmental Conservation PAH sediment criteria

| PAH | Human health | | Benthic aquatic life, acute toxicity | | Benthic aquatic life, chronic toxicity ^a | |
|------------------------|---------------------|-----|---|-----|--|----------|
| | FW | SW | FW | SW | FW | SW |
| | µg/g Organic carbon | | | | | |
| Acenaphthene | | | | | 140(E) | 140(E) |
| Benzo(a)pyrene | 1.3 | 0.7 | | | | |
| Fluoranthene | | | | | 1020 (E) | 1340 (E) |
| Phenanthrene | | | | | 120 (E) | 160 (E) |
| Anthracene | | | 986 | | 107 | |
| Benz(a)anthracene | 1.3 | 0.7 | 94 | | 12 | |
| Fluorene | | | 73 | 348 | 8 | 38 |
| Benzo(b)fluoranthene | 1.3 | 0.7 | | | | |
| Benzo(k)fluoranthene | 1.3 | 0.7 | | | | |
| Chrysene | 1.3 | 0.7 | | | | |
| Indeno(1,2,3-cd)pyrene | 1.3 | 0.7 | | | | |
| Naphthalene | | | 258 | 328 | 30 | 38 |
| Pyrene | | | 8775 | | 961 | |

FW= freshwater; SW= saltwater

Source: New York State Department of Environmental Conservation [57].

Criteria marked (E) extracted from U.S. EPA [58]

2. PAH EMISSIONS SOURCES

In years past, literature reports have identified certain industries, including energy and metal production, as the main culprits releasing large quantities of PAHs to the atmosphere. However, modern atmospheric emissions controls have dramatically reduced the emission of PAHs from these processes.²⁴ Similarly, catastrophic releases of petroleum to land and water have become less common nationwide as a result of improved engineering controls, although the cumulative impact of smaller petroleum spills is still of concern.

Today, a general consensus of literature reports two major national and global sources of PAHs in the environment: 1) incomplete combustion of organic matter, especially common, nonpoint activities that utilize modern emissions controls (e.g., cars) or less common activities with no emissions control (e.g., tire fires); and 2) releases of petroleum, including oil spills and illegal dumping. Major sources include forest fires, motor vehicle emissions, open burning, domestic fireplaces, and spills and dumping of petroleum products. In the sections that follow, what is known about releases of PAHs from natural and anthropogenic sources in the Watershed to air, water, and land will be quantified.

2.1. General Anthropogenic Sources

Anthropogenic emissions of PAHs to the environment are predominantly incidental, including combustion byproducts from motor vehicles and various industrial processes. Unlike many other environmental contaminants of concern in the NY/NJ Harbor, very few PAHs are intentionally manufactured. The only PAH that is produced industrially in large quantities and has direct commercial uses is the bicyclic compound naphthalene [59]. The total annual consumption of naphthalene in the United States in 2000 was approximately 109,000 metric tons [60]. Major uses for naphthalene include mothballs, paint thinner, and solvent for metal surface prep [61]. Naphthalene is also used as a chemical intermediary in pharmaceutical and photographic industries, and, to a limited extent, in the production of soaps, pigments and dyes, insecticides, fungicides, plastics, and processing of certain foods [62]. In addition, naphthalene is used for the production of phthalic anhydride, an intermediate for polyvinyl chloride (PVC) plasticizers [59].

Other PAHs, including acenaphthene, anthracene, phenanthrene, fluorene, and pyrene, have some limited industrial uses. Acenaphthene is used for production of naphthalic anhydride, an intermediate for pigments [59]. Anthracene is used as a chemical intermediary for dyes, as a dilutant for wood preservatives [62], and as a scintillant for the detection of high-energy radiation [59]. Fluorene is used for the production of fluorenone, a mild oxidizing agent [59]. Phenanthrene's primary use is for production of phenanthrenequinone, an intermediate for pesticides, and for diphenic acid, an intermediate for resins, while pyrene's main use is to make dyes [59]. Annual national production data for PAHs other than naphthalene are not available, but emissions from manufactured PAHs are not expected to be a major emission source locally. As described below, 1999 U.S. EPA's National Emissions Inventory (NEI) reported that national annual atmospheric emissions of total PAHs (where the total is the sum of the 16 priority PAH compounds) from all chemical and pharmaceutical sources combined were less than 1000 kg/yr, a small fraction of the greater than 5000 metric tons/yr released from all atmospheric sources.

2.1.1 General Sources of PAHs Released to the Atmosphere

Data from the U.S. EPA National Emissions Inventory (NEI) suggest that mobile sources are an important source of atmospheric PAH emissions (approximately 90% of total atmospheric emissions in 1999, excluding forest fires [63]; TABLE 2.1).²⁵

Although the NEI database is a very convenient repository for national chemical emissions, these data

Table 2.1. U.S. annual atmospheric emissions of total PAHs (defined as the U.S. EPA 16 priority PAHs) for all sources except forest fires

| PAH source | PAH (tons/yr) | Percent total PAHs |
|-------------------------------|---------------|--------------------|
| Motor vehicles | 4474 | 90 |
| Nonpoint sources ^a | 489 | 10 |
| Point sources ^b | 13 | <1 |
| Total PAHs | 4977 | 100 |

Source: U.S. EPA National Emissions Inventory, 1999 [63].

^a For examples of nonpoint sources, see TABLE 2.3.

^b For examples of point sources, see TABLE 2.4.

24. See Section 4.5.4. Incineration for a description of typical emissions control devices utilized by industry today.

25. Forest fires are excluded because the typical incidence of forest fires nationwide is not similar to the incidence in the New York City metropolitan area. Further, if transboundary sources of PAHs including western U.S. forest fires were a dominant source of PAHs on the East Coast, then we would expect to find uniform PAH concentrations in urban and rural areas, which plainly is not the case.

should be used with some caution. The data are voluntarily reported by various regional authorities, and in some cases are not reported consistently across all states or regions. Data verification or monitoring is not expected to be part of this effort. However, as a general accounting of the major sources of PAH emissions to the atmosphere, it is a useful resource.

These NEI data indicate that, nationwide, mobile sources release nearly 90% of total atmospheric emissions (again, excluding forest fires) of total PAHs. When these sources are broken down further by category, it is apparent that on-road traffic of cars and trucks is responsible for more than 99% of mobile source emissions of total PAHs to the atmosphere (TABLE 2.2). According to the NEI data, national emissions of total PAHs from motorcycles is only 0.7% of emissions from all mobile sources, and national emissions of total PAHs from all other nonroad sources, including aircraft, trains, all-terrain vehicles, recreational marine vessels, tractors, and lawnmowers, is only 0.1% of the total.

Table 2.2. U.S. annual atmospheric emissions of total PAHs (defined as the U.S. EPA 16 priority PAHs) from all mobile sources according to NEI data

| Mobile source | PAHs (tons/yr) | Percent total PAHs |
|---------------------|----------------|--------------------|
| Light-duty vehicles | 1966 | 44 |
| Light-duty trucks | 1411 | 32 |
| Heavy-duty vehicles | 1065 | 24 |
| Motorcycles | 29 | <1 |
| Nonroad | 3 | <1 |
| Mobile source total | 4474 | 100 |

Source: U.S. EPA National Emissions Inventory, 1999 [63].

A breakdown by fuel type shows that 95% of the total mobile emissions of total PAHs nationwide comes from gasoline-powered vehicles and the remaining 5% comes from diesel [63].²⁶

The second largest (<10%) category for national atmospheric emissions of total PAHs according to the NEI is from nonpoint sources. The three largest major sources within this category are consumer and commercial product use, open burning, and fireplaces (TABLE 2.3). These three sources constitute 80% of the nonpoint source total. Other sources include gasoline distribution and cement manufacturing.

Table 2.3. U.S. annual atmospheric emissions of PAHs (defined as the U.S. EPA 16 priority PAHs) from all nonpoint sources

| Nonpoint source | PAHs (tons/yr) | Percent total PAHs |
|--|----------------|--------------------|
| Consumer and commercial product use ^a | 133 | 27 |
| Open burning | 130 | 27 |
| Fireplaces | 126 | 26 |
| Gasoline distribution | 53 | 11 |
| Concrete manufacturing | 39 | 8 |
| Other | 9 | 2 |
| Nonpoint source total | 489 | 100 |

Source: U.S. EPA National Emissions Inventory, 1999 [63].

^a Includes powders, polishes, soaps, hair care products, coal tar and asphalt coatings, and pesticides. For a full list of activities included under this and other source categories see the U.S. EPA National Emissions Inventory, <http://www.epa.gov/ttn/chief/net/index.html>.

The third main emission category of the NEI is point sources (<1%). These sources are various industrial facilities. The major contributors to PAH emissions fall into the categories of transportation equipment; fabricated metal products; and electric, gas, and sanitary services (TABLE 2.4). As can be seen in the following sections, national trends do not necessarily hold true for regional trends.

Emission trends for total PAHs can be quite different from emissions trends for individual compounds.

Table 2.4. U.S. annual atmospheric emissions of PAHs (defined as the U.S. EPA 16 priority PAHs) from all point sources

| Point source | PAHs (tons/yr) | Percent total PAHs |
|---|----------------|--------------------|
| Transportation equipment ^{a,b} | 9.1 | 69 |
| Fabricated metal products | 1.3 | 10 |
| Electric, gas, and sanitary services | 1.1 | 9 |
| Chemicals and pharmaceuticals | 0.7 | 5 |
| Stone, clay, and glass products | 0.6 | 5 |
| Other | 0.3 | 2 |
| Point source total | 13.1 | 100 |

Source: U.S. EPA National Emissions Inventory, 1999 [63].

^a Includes manufacturing and repairing vehicle, marine, and air equipment.

For a full list contact the U.S. EPA Clearing House for Inventories and Emissions Factors.

^b These emissions are the result of reporting by one motor company in Kentucky.

26. Vehicle emissions reported by the NEI are calculated using the emission fractions described in Section 3.3.1. Vehicle Exhaust and may not accurately reflect emissions of the current vehicle fleet.

For example, in many of these categories, naphthalene accounts for more than 50% of the total PAH emissions. When the NEI data are examined for the emissions signature of another compound, a totally different set of major sources is found. For example, let us consider emissions of the five-ring PAH benzo[a]pyrene (BAP). This compound receives a lot of attention because of its relatively high carcinogenicity and because it is included in the EPA's list of Priority Persistent and Bioaccumulative Toxins (PBT). BAP emissions are dominated by several nonpoint sources, not by mobile sources (as is the case for total PAHs) [64]. Major emissions sources nationwide for BAP are open burning, fireplaces, and a few industrial processes that include metal production, petroleum refining, and incineration (FIG. 2.1).

2.1.2. General Sources of PAHs Released to Land

While a substantial amount of PAHs reach land surfaces via dry and wet deposition of atmospheric particles, other emission sources release PAHs directly to land. Major sources of PAH emissions to land are spills and dumping, especially improper disposal of used motor oil; mobile sources, including leaking oil from cars, tire wear, and brake dust; and use of PAH-containing products such as creosote wood preservatives and coal tar-based asphalt sealants. A catalog of national sources of PAH emissions to land was unavailable.

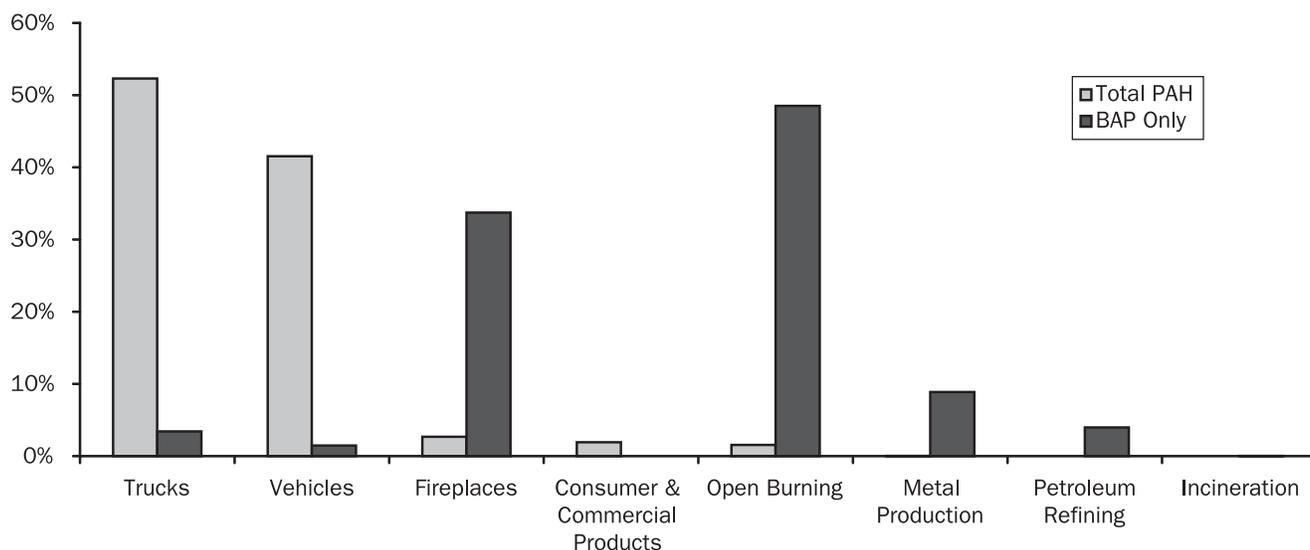
2.1.3. General Sources of PAHs Released to Water

PAHs may be released to water by several routes, including from oil spills onto water or land, stormwater runoff, wastewater treatment effluent, and creosote-treated wood use [59]. Very few data are available on typical PAH loadings in wastewater from specific industrial processes. However, some reports have indicated that important national sources of PAH releases to water are oil spills, dumping of used motor oil, stormwater runoff, and release of PAHs from products such as creosote and coal tar sealant.

2.2. General Natural Sources

PAHs have been present in the environment since long before anthropogenic activity. Naturally occurring forest fires, volcanic activity, and fossil fuel deposits are all examples of sources of PAHs in the environment.

Figure 2.1. Comparison of the breakdown by category of total PAH emissions to the breakdown by category for BAP emissions, according to the U.S. EPA 1999 National Emissions Inventory [63] [64]



3. MAJOR REGIONAL SOURCES OF PAHS

We have identified over 30 different sources of PAHs in the Watershed; however, 11 major sources each contribute more than 2% to the total PAHs released to their primary media of release (see SUMMARY OF FINDINGS). These sources include residential wood combustion, creosote-treated wood, surfaces sealed with refined coal tar-based sealants, vehicle exhaust, tire wear, leaking motor oil, improper disposal of used motor oil, and petroleum spills. Contaminated sites are also considered a major source of PAHs. While we have not estimated releases from these sites, given their pervasiveness in the region and proximity to the Harbor, they are potentially a significant source of PAHs.

The following sections of this report provide technical detail on the major sources of PAHs in the Watershed, including PAH release estimates, relevant policy, and pollution prevention recommendations.

3.1. Residential Fuel Combustion

3.1.1 Wood Combustion (Wood Stoves and Fireplaces)

Wood Stoves and Fireplaces: National Trends and Emission Factors

Nationwide there are approximately 37 million residential wood combustion devices, of which 72% are estimated to be fireplaces, 25% wood stoves, and the remaining 2% are appliances such as pellet stoves and outdoor wood boilers (OWBs) [65]. Wood stoves and fireplaces release PAHs through the incomplete combustion of wood. While a suite of PAHs are released during this process, wood combustion has been identified by several sources (including the NEI and the Great Lakes Binational Toxic Strategy) as a dominant source of benzo(a)pyrene, one of the more toxic PAHs [66, 67].

Emissions factors for PAHs from domestic wood combustion devices are highly variable and depend on many factors, including type of fuel (e.g., spe-

cies of wood, and the aging process), wood moisture content, and design of the combustion device. The U.S. EPA provides emission factors for conventional, noncatalytic,²⁷ and catalytic²⁸ wood stoves (TABLE A.1) [68]. However, the report cautions that the available data used to generate the emission factors were sparse and/or had a high degree of variability, and should therefore be used with caution. Of the three wood stove types, emission factors for conventional stoves are the highest, likely because they lack emissions control devices.

Currently, regulations require all wood stoves manufactured after 1992 to pass U.S. EPA emissions certification, Phase II classification, in which noncatalytic and catalytic stoves have a smoke emission limit of 7.5 and 4.1 grams of smoke per hour, respectively [69]. Woodstoves have a relatively long life span, and it is estimated that only 11% of woodstoves currently in use are U.S. EPA certified [65].

A recent report provides PAH emissions factors for the domestic burning of seasoned hardwood in a fireplace (TABLE A.1) [70]. Unlike wood stoves, fireplace emission standards are regulated at the state and local level [71]. The U.S. EPA has, however, certified fireplace inserts that adhere to the same emission limits as woodstoves [72].²⁹

Little information is available on the release of PAHs from outdoor wood boilers (OWBs).³⁰ However, a study by the New York State Attorney General's Office states that on average wood boilers emit 0.97 grams of PAHs per hour and 71 grams of particulate matter per hour [73]. A recent report by the U.S. EPA compared PAH emissions from OWB and certified wood stoves (catalytic and noncatalytic) and found wood stove emissions to be 1.5 times greater. OWBs are not currently subject to any Federal regulations, likely because of their low popularity during the 1980s, when wood stove standards were initially established by the U.S. EPA. Recently, however, OWBs have increased in popularity [73]. In fact, U.S. OWB sales in 2005 doubled from the previous year, reaching approximately 67,500 units [74].

Pellet stoves are another device used to combust wood. Pellet stoves consume wood and biomass that have been compressed into pellets. While some pel-

27. Noncatalytic stoves increase the potential for complete combustion by utilizing large baffles to create a longer and hotter gas flow path, as well as by introducing preheated combustion air.

28. Catalytic stoves are equipped with a noble metal-coated combustor that ignites and burns the combustible components in the effluent.

29. Fireplace inserts are structured similar to free-standing woodstoves and are designed to fit into an existing fireplace opening. Fireplace inserts reduce the amount of heat that is typically lost up a chimney [72].

30. OWBs are freestanding combustion units resembling a shed with a low-standing chimney and are located outside of the home. The combustion exhaust is used to heat a water reservoir, which is then piped into the structure to be heated. A thermostat adjusts heat by controlling the amount of air that is supplied to the burn box. While restricting the air to the burn box results in lower temperatures, it also creates a lower quality combustion environment, generating PAHs.

let stoves are subject to the 1988 New Source Performance Standards, others are exempt, because of their high air-to-fuel ratio of 35:1[68]. Available emission factors are incomplete for pellet stoves.

Wood Stoves and Fireplaces: Regional Releases

The New York and New Jersey residential sector consumed over 4.5 million tons of cordwood in 2001 [75].³¹ Watershed consumption of cordwood was estimated by extrapolating from state data based on the percentage of homes in the Watershed that reported using wood as their primary heating source to the U.S. Census—40% and 44% in the New York and New Jersey Watershed, respectively (or 2 million tons of cordwood) [77]. While it is unknown what type of combustion unit the wood is burned in, it is assumed that more wood is consumed in wood stoves (72%), because wood stoves are used as a primary heating source, whereas fireplaces are typically used as a supplementary heating source or for aesthetic value [65]. It is not known how much wood is consumed in OWBs. However, OWB sales in New York have increased by almost 70% over the past five years, and it is likely that some of the wood consumed by the Watershed residential sector is combusted in these units [73].

It was unknown whether any of the emission factors for the catalytic and noncatalytic wood stoves are calculated based on emissions from U.S. EPA-certified units (stoves that are likely to have lower PAH emissions than conventional models). Therefore, it was as-

sumed that 11% of the wood consumed in wood stoves is combusted in a noncatalytic wood stove, the model with the overall lower PAH emission factor, and that the remaining 89% of the wood is consumed equally between the conventional and catalytic wood stoves.³²

Releases to the Atmosphere. Estimated PAH emissions from residential wood combustion were calculated by applying the emission factors presented in TABLE A.1 to the estimated wood consumption in the Watershed (TABLE 3.1). Over half of the estimated PAH emissions from this source come from the combustion of wood in conventional wood stoves, followed by those stoves with catalytic converters. It appears that more PAHs are released from combustion devices outside of the Watershed.

There is some uncertainty with this estimate. The emission factors are based on laboratory testing of wood combustion devices, not actual use. Stove maintenance, type of wood combusted, and operating procedures will impact the efficiency of the combustion device and, in turn, PAH emissions.

Measures to Reduce Releases of PAHs from Residential Wood Combustion

Currently, there are no residential wood burning guidelines in New York and New Jersey other than those established by the U.S. EPA [79] [80]. U.S. EPA-certified noncatalytic wood stoves have been demonstrated to emit less particulate matter, volatile organic compounds, and PAHs (85% less) than noncertified

Table 3.1. Estimated residential wood combustion activity and associated PAH emissions in New York and New Jersey^a

| | Estimated wood consumption (tons/yr) | PAH emissions (kg/yr) | | | |
|--------------------------|---|-----------------------|----------------|----------------|---------------|
| | | Fireplace | Conventional | Catalytic | Noncatalytic |
| Watershed | | | | | |
| New York | 1,634,400 | 16,800 | 170,600 | 96,200 | 23,100 |
| New Jersey | 184,000 | 1,900 | 19,200 | 10,800 | 2,600 |
| Total | 1,818,400 | 18,700 | 189,800 | 107,000 | 25,700 |
| Outside Watershed | | | | | |
| New York | 2,487,700 | 25,600 | 259,700 | 146,400 | 35,100 |
| New Jersey | 235,600 | 2,400 | 24,600 | 13,900 | 3,300 |
| Total | 2,723,300 | 28,000 | 284,300 | 160,300 | 38,400 |

Source: Total Wood Consumed by Residential Sector, http://www.eia.doe.gov/emeu/states/sep_use/total/pdf/use_nj.pdf.

^a PAH emission estimates are based on emission factors presented in TABLE A.1. For estimated PAH emissions by compound, see TABLE A.2.

31. Wood consumption data are an estimate of wood that is purchased and harvested by end users. Data were collected through surveys by the Energy Information Administration (EIA). It is assumed that the consumption of manufactured wood, such as wax/sawdust fire logs, is not represented in the data. Wax wood has been shown to emit fewer total PAHs than cordwood [76].

32. In a cooperative study between Environment Canada and the Hearth Products Association of Canada, U.S. EPA-certified stoves were found to emit, on average, 66% fewer PAHs than conventional models [78]. Based on emissions calculated using U.S. EPA emission factors, noncatalytic woodstoves were found to emit 86% less PAHs than conventional stoves.

conventional stoves [81].³³ As previously mentioned, all wood stoves manufactured after 1992 must meet the U.S. EPA Phase II guidelines; however, their penetration into the market has been slowed by the long life span of wood burning stoves. Some communities have combated this issue by sponsoring changeout programs that offer incentives or rebates when older stoves are exchanged for newer, more efficient models, or when fireplace inserts are purchased. For example, a community in Libby, Montana participated in the U.S. EPA's Woodstove Changeout Campaign, in which residents owning stoves that are older than eight years and who qualify for public assistance are eligible for a wood stove replacement [82].³⁴ A similar program was conducted in the Great Lakes region from February to April of 2001, and encompassed Illinois, Indiana, Iowa, Kansas, Michigan, Minnesota, Nebraska, North Dakota, New York, Ohio, South Dakota, Wisconsin, and the Georgian Bay region of Canada. This program facilitated the replacement of over 1200 older stoves with newer, less polluting stoves [83].

The U.S. EPA emission standards at this time do not reflect best available technology. Washington state, for example, has a limit of 4.5 g/hr for noncatalytic woodstove and 2.5 g/hr for catalytic stoves (almost half the U.S. EPA's limit) for all stoves sold in the state. Based on an evaluation of industry data that indicate approximately 75% of new EPA-certified woodstoves meet the more stringent Washington state standards, the U.S. EPA has chosen to focus on facilitating the exchange of older stoves, rather than implementing stricter emission standards [84].

WOOD HEATING BEST MANAGEMENT PRACTICES

Firewood

- Using seasoned wood (wood with less than 20% water) produces more heat and makes less smoke.
- Wood is ready to burn when it has cracks in the end grain, does not hiss or sizzle in the fire, is lighter to carry than when it was cut, and has darkened to brown or gray.
- Don't burn household garbage (especially plastics), treated wood, particle board, or saltwater driftwood.
- Never use gasoline or oil to start a fire.

Fireplace or Wood Stove

- When purchasing a wood stove make sure it is EPA certified, or when purchasing a fireplace buy an EPA certified insert.
- Before adding wood, rake coals to the front near the air inlet.
- When starting a fire, add seasoned wood, close door, and open the air control fully and/or crack the door to get the wood flaming quickly.
- Once the fire is going, close and latch the door and adjust the air inlet.
- Wood burns best when there are several logs burning at a time; less smoke means a more efficient fire.

Maintenance

- Make sure door closes properly (check hinges, door latch, and gaskets).
- Replace any cracked or damaged firebrick.
- Inspect the chimney regularly, cleaning as needed to avoid creosote buildup

Source: Summarized from *Burn it Smart! Wood Heating Guide to: Burn Less Wood, Make Less Smoke, Be More Comfortable*. Government of Canada.

Some communities have directed their air pollution reduction efforts towards exchanging wood stoves for cleaner fuel-burning devices such as gas-powered stoves. For instance, in 2003 Santa Clara County, California started a switchout campaign in

33. This is consistent with our estimate in which noncatalytic stoves emit approximately 86% less total PAHs than do conventional stoves.

34. From 2005 through 2006, the U.S. EPA sponsored two to three changeout programs, targeting particulate matter, 2.5 nonattainment areas, and communities that have community-based air toxics programs. For more information visit <http://www.epa.gov/airprog/m/oar/woodstoves/changeout.html>.

which residents received a \$300 or \$500 rebate, depending on age and combustion device, for switching from an old wood burning device to a natural gas system.

In addition to promoting the use of more efficient combustion units, PAH emissions from wood combustion can be addressed through energy efficiency measures and reduced fuel combustion. Various government and nonprofit groups have established energy efficiency programs for residential construction. One example is the Energy Efficiency Construction Code of New York State, which requires minimum standards of energy efficiency in new residential and commercial buildings. Another example is New Jersey's Clean Energy Program, which offers incentives for the installation of energy efficient measures in homes, specifically those that are income eligible. The Federal government supports energy efficiency through the Energy Star program, in which homes that are verified to be at least 30% more energy efficient than homes built to the 1993 national Model Energy Code or 15% more efficient than the state energy code, whichever is more rigorous, are certified. In addition to government sponsored energy efficiency programs, The U.S. Green Building Council, a nonprofit organization that promotes environmentally responsible and profitable buildings, is in the pilot stage of developing a Leadership in Energy and Environmental Design (LEED) certification for homes.³⁵

Three approaches have been identified to reduce PAH emissions from residential wood combustion: reduce fuel consumption, improve combustion conditions and reduce releases of PAHs, and substitute combustion technology.

The following are pollution prevention recommendations to reduce release of PAHs from residential wood combustion activity in the Watershed:

- Reduce fuel consumption.
 - Educate retailers and consumers on how to choose a heating unit that is properly sized for the desired heating area. An oversized stove may need to be damped down regularly, creating a less ideal combustion environment.

- Educate designers and local government officials on sustainable/green design and passive solar design elements that will contribute to a reduction in heating demands, such as increased insulation, elimination of thermal bridges,³⁶ optimum value engineering,³⁷ and properly installed and sealed windows.
- Increase incentives for homes that are heated by non-PAH-releasing energy.
- Adopt energy efficiency measures as code, such as those recommended by Energy Star program.
- Reduce the quantity of PAHs released.
 - Establish an ongoing state or municipal program that promotes the use of fireplace inserts and the exchange of older, less efficient stoves for new, less polluting stoves, possibly modeled after the U.S. EPA's changeout program that has been implemented in several states.
 - Educate the community on the environmental impact of combusting contaminated materials in their wood burning units, such as treated wood and household waste, as well as good maintenance practices, such as regularly checking the condition of the baffle or catalyst; cleaning the catalyst; burning only seasoned wood; and removing excess ashes.
 - Promote the sale and use of wood stoves that utilize the best available control technologies, such as those sold in Washington State.
- Consider substituting wood combustion units with heating devices that combust cleaner burning fuel, possibly through incentives.
- Data gaps.
 - As previously indicated little data are available on the prevalence of OWB use and associated PAH emissions. Given the design of these units and the lack

35. The USGBC currently provides LEED certification for commercial projects.

36. An example of where a structural thermal bridge may occur is at the point where projecting beams and slabs pass through the building envelope between the cold and warm side of a building. This may allow the transfer of heat to the cold side of the structure.

37. Optimum Value Engineering (OVE) framing optimizes the amount of lumber used to frame homes, creating more space for the insulation of exterior walls. Examples of OVE strategies include utilizing two-stud corner framing with inexpensive drywall clips; increasing floor joist and rafter spacing to 24 inches; eliminating headers in nonloadbearing walls; increasing stud spacing from 16 inches to 24 inches; and using single top plates with in-line framing to transfer loads directly.

of regulation, it is possible that this is a significant source of PAHs. Emission factors and activity level data should be developed for this source.³⁸

3.2. Materials Containing PAHs

3.2.1. Creosote-Treated Wood Production and Use

Creosote-Treated Wood Production and Use: National Trends and Emission Factors

Creosote, a distillate of coal tar,³⁹ is an insecticide, fungicide, miticide and sporicide commonly used to waterproof and preserve wood. In the U.S., creosote-treated wood is used for railway ties (50%), utility poles (30%), and fence posts (14%), with a small fraction used in marine pilings (0.17%) [5].⁴⁰ Creosote has been designated as a probable human carcinogen by the U.S. EPA [89], and it is not available for consumer purchase.⁴¹ Furthermore, pressure treatment of wood with creosote is the only wood treatment method used and creosote can only be used by applicators that have completed a U.S. EPA approved training program [91].

In 2004, approximately 28 million cubic feet of creosote-treated wood were produced in the U.S. from 87 million gallons of creosote [5]. The manufacture of creosote-treated wood appears to be on a downward trend. The USDA reported that 97 million cubic feet of creosote-treated wood were produced in 1999, approximately 13% of all treated wood for that year, and a 27% reduction from 1983 [92]. In the 1950s, two other wood preservatives began to replace creosote in some applications: pentachlorophenol (PCP)⁴² and chromated copper ar-

senate (CCA), the most common preservative used to treat wood [93].^{43,44}

WOOD TREATMENT TECHNIQUES

There are generally three techniques for introducing preservatives into wood: pressure, hot-cold, and superficial. Pressure treatment, by which the vast majority of wood is treated and the only method used with creosote, involves placing wood and preservative in a chamber and forcing the preservative into the wood under extreme pressure, thus impregnating the wood with the preservative [85]. Hot-cold treatment, only used when treating red cedar with copper naphthenate or pentachlorophenol, involves placing the wood in a hot bath of preservative and then quickly flooding the tank with cold preservative, causing the heated air to contract and pull in the preservative. Superficial treatments (a method no longer permitted for creosote) include brushing, spraying, and dipping wood in the preservative [86]. The quantity of preservative used depends on the deterioration zone of the region in which the wood will be used (as defined by the American Wood-Preservers' Association). For example, some southern states fall within a severe zone, while northern states, including New York and New Jersey, fall within a less severe zone, requiring lower quantities of preservative to treat wood for an average product lifetime of 35-40 years [87].

38. A concern with OWBs has surfaced in the New York/New Jersey region as a result of increasing complaints from OWB neighbors. The New York State Attorney General's Office recently sent a petition to the U.S. EPA requesting that OWB standards of performance be promulgated under 42 U.S.C. § 7411(b)(1)(B). Several states have also signed the petition, including Connecticut, Maryland, Massachusetts, Michigan, Vermont, and New Jersey [79]. The North East States for Clean Air Future, with participation from the U.S. EPA Office of Air Quality and Standards, are composing model rules for state adoption and developing an incentivized voluntary program for outdoor wood boiler manufacturers. Issues to be addressed in the model rule are emission limitations, zoning, stack height, and operations and maintenance [80].

39. Coal tar is a byproduct of coal coking. In 1992, 0.7 million tons (1.5 billion lbs) of crude coal tar were produced [88].

40. Estimates assume treatment with the following volumes (cubic feet) of creosote per piece produced: railway tie, 3.54; utility pole, 60; fence post, 1.5. Marine piling estimates were provided in cubic feet [5].

41. EPA is currently reassessing creosote under its four-phase process as part of its ongoing re-registration program for older pesticides (see http://www.epa.gov/oppsrrd1/public_summaries.htm - 4phase for more information on the process). Federal law directs U.S. EPA to periodically reevaluate older pesticides to ensure that they continue to meet current safety standards. A decision is scheduled to be made by winter of 2007 [90].

42. Materials treated with PCP are associated with dioxin release. For more information see *Pollution Prevention and Management Strategies for Dioxins in the New York/New Jersey Harbor*, G. Muñoz et al., page 175.

43. A representative of the Creosote Council indicated that PCP and CCA were less expensive, and the replacement of creosote with these preservatives can be partially attributed to this economic factor [87]. Utilities also changed from creosote-treated wood to PCP-treated wood because of performance, environmental, and safety issues—wood treated with PCP is stronger, and creosote is known to seep from wood and rub off on utility pole climbers (which does not happen with PCP- or CCA-treated wood) [94].

44. Due to concerns with the chromium and arsenic found in this preservative, the U.S. EPA established a voluntary phase-out of CCA-treated wood used for residential construction by the end of 2003. Several alternatives to CCA have been developed, including ammoniacal copper quat (ACQ) and copper azole. ACQ is a water-based wood preservative that combines copper with an ammonium compound. Copper azole contains copper and boron. Copper can be harmful to aquatic systems.