Use of Mass Balance Bounding Estimates and Sensitivity Analysis to Prioritize PAH Inputs in Urban Systems

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ABSTRACT

PAH transport via the atmosphere and runoff inputs related to high density population and traffic in urban areas have been extensively studied for decades. In recent years, several studies focused on the use of specific PAH-containing materials, particularly pavement sealer products, as possible sources of sediment PAHs. The big-picture studies are advantageous for documenting patterns of PAH distribution actually established in the environment, but typically provide limited information about specific source inputs in a given location. The product-based studies can serve to identify locally relevant sources, but typically have limited resolution to evaluate relative inputs from numerous sources in larger systems. And, the types of controlled monitoring programs that can provide empirical information on the connections between specific sources and environmental load in an urban area are large, expensive and time-consuming. As a study design tool to help optimize monitoring programs, we developed bounding estimates for inputs that affect relative mass loading from local traffic, pavement sealer and residential wood combustion sources of PAHs. Using U.S. Census Bureau data for 42 metropolitan areas, average rainfall patterns and relationships between parking lot densities and population densities to get a probabilistic assessment of the relative PAH contributions from the various sources considered in the model. The output shows that residential wood combustion is the dominant source of PAHs in most metropolitan areas and that pavement sealer products contribute on average less than 5% of PAH mass inputs. On a more local scale, critical factors likely include traffic density, rainfall washout patterns and the size of sealed parking lots relative to streams receiving uncontrolled runoff.

INTRODUCTION

- Well known sources of PAHs in urban areas include utility and industrial combustors, automobiles and heating systems, particularly wood combustion.
- In recent years, pavement sealer products containing refined tar, derived from coal, have been suggested as another significant source.
- Monitoring and source apportionment are complicated by the complexities of multiple types and routes of source input.
- To help design field studies with relevant scales of resolution and put expected relative source contributions in context, we developed input estimates covering pavement sealer, vehicle and residential wood burning from published literature and census information.
- Probabilistic approach incorporated information from 42 metropolitan areas covering all regions of the U.S.

METHODS

- **PAH mass loading from CT-Sealed parking lots** (mg/m^2-rain event) estimated from data reported in four studies (Horwatich et al., 2005; Mahler et al., 2005; Selbig, 2009; Watts et al., 2010); corrected for loading from unsealed lots.
- **PAH mass loading from Asphalt-Sealed parking lots** (mg/m^2-rain event) estimated from data reported by Mahler et al. (2005); corrected for loading from unsealed lots.
- **PAH mass loading from vehicles** includes vehicle exhaust, tire wear and motor oil.
  - PAH emissions from vehicle exhaust – fleetwide emissions summarized in USEPA (1998) and 15 to 35 daily VMT/person
  - PAH emissions from tire wear – PAH content in HA oil (KEMI, 2003) and tire wear estimate (Councell et al., 2004).
  - PAH released in used motor oil – PAH content in used oil and drip estimate of 2 ml per 1000 km (CalEPA, 2006).

- **PAH mass loading from residential wood combustion** -- numbers of households using wood as heating fuel (U.S Census Bureau American Housing Survey [AHS]) ; PAH emission factors for fireplaces and wood stoves (MARAMA ,2006).
  - AHS data used for distribution of residential wood combustion as a function of population and climate zone.

Monte Carlo Simulation run with 100,000 iterations using Crystal Ball Software.

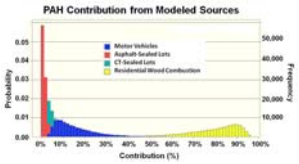
- **Randomly select a climate zone**.
- **Randomly select a land area** (Crystal Ball Fit to geographical definitions of metropolitan areas covered by U.S Census Bureau American Housing Survey (AHS).
- **Select a population density** (pop/mi^2) based on selected land area using Crystal Ball fits to AHS geographical definitions divided into four size ranges.
- **Estimate land area covered by parking lots** as a percentage of total area based on best least squares fit to data presented by Davis et al. (2010)
- **Estimate proportion of sealed lots** -- Simulation assumes 15% to 40% of all lots are sealed and random split between CT and asphalt emulsion sealer.

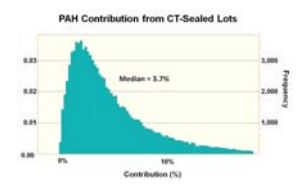
RESULTS

- Several studies and 68 measured values available to characterize loading via runoff from parking lots with tar-based sealers (CT-sealed) - median = 0.9 mg/m^2 per rain event.
- Median contribution from CT-sealer less than 4% of urban input, less than 1/3rd of input from vehicle exhaust, tire wear and motor oil.
- Roadway and pavement sources overall typically amount to less than 1/4th of PAH input.
- Airborne contributions (i.e., wood burning) expected to be majority source.

PAH Emission Factors Used in the Simulation

Source	Emission Factor	References/Notes
Vehicle Exhaust	0.17 mg/VMT	U.S. EPA (1998); does not include off-road vehicles
Tire Wear	0.056 mg/VMT	KEMI Report (2003); assumes 1% PAH in HA Oil
Motor Oil	2300 mg/L oil	Average of 13 measurements over 3 studies
Residential Wood Combustion	300 mg/kg wood	MARAMA (2006); weighted average of wood stoves and fireplaces
CT-Sealed Parking Lots	0.91 mg/m²-event	Median estimate (n=68); corrected for unsealed lots
Asphalt Emulsion-Sealed Parking Lots	0.19 mg/m²-event	Median estimate (n=3); corrected for unsealed lots





PAH SOURCE	PERCENTILE ESTIMATES	
	50th %ile	90th %ile
Residential Wood Combustion	82	92
Vehicle Emissions	13	29
CT-Sealed Parking Lots	3.7	11
Asphalt-Sealed Parking Lots	0.77	2.4

CONCLUSIONS

- Mass balance model and probabilistic results consistent with empirical study on NY/NJ Harbor (Valle et al., 2007)* and historical literature – airborne, especially wood burning, a dominant source.
- Loading contributions from CT-sealer small overall and small relative to vehicular inputs.
- Given relatively high PAH concentrations in CT-sealer, contributions from parking lots are expected to be highly localized compared to other sources.
- Overall PAH reduction following restrictions/bans on CT-sealer expected to be small and difficult to discern.
- Monitoring programs will require high resolution, highly localized designs to find effects from reducing CT-sealer use.

*Mass balance model does not include potential loadings from creosote which was determined to be a major PAH source in the NY/NJ Harbor watershed.

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