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A Photographic Method for Estimating Wear of Coal Tar Sealcoat from Parking Lots

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Coal-tar-based sealcoat has been recognized as an important source of PAHs to the environment through wear and transport via stormwater runoff. Sealcoat removal rates have not been measured or even estimated in the literature due to the complex array of physical and chemical process involved. A photographic study was conducted that incorporates all sources of wear using 10 coal tar-sealed parking lots in Austin, Texas, with sealcoat age ranging from 0 to 5 years. Randomly located photographs from each parking lot were analyzed digitally to quantify black sealed areas vs lighter colored unsealed areas at the pixel level. The results indicate that coal tar sealcoat wears off of the driving areas of parking lots at a rate of approximately 4.7% per year, and from the parking areas of the lots at a rate of approximately 1.4% per year. The overall annual loss of sealcoat was calculated at 2.4%. This results in an annual delivery to the environment of 0.51 g of PAHs per m^2 of coal tar-sealed parking lot. These values provide a more robust and much higher estimate of loading of PAHs from coal tar sealcoated parking lots when compared to other available measures.

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a large group of organic compounds that are primarily formed during the partial combustion of carbon sources and are ubiquitous in the environment. Concentrations of PAHs have been increasing over the last 20−40 years in sediments in urban receiving water bodies throughout the United States (*1*). Until recently, urban sources of PAHs have been linked to vehicle use and atmospheric deposition (*2, 3*), primarily from coal and wood combustion. However, research by the City of Austin and the U.S. Geological Survey (USGS) has identified parking lot sealcoat, and particularly coal tar-based sealcoat, as a major and previously unrecognized source of PAH contamination (*4, 5*). Coal tar sealcoats are used in an emulsion form containing up to 35% refined coal tar, which is on the order of 50% PAHs by mass (*6*). The City of Austin analyzed a variety of coal tar sealcoat products ($n = 12$) whose dry weight average concentration using the Sediment Quality Guideline sum of PAHs (ΣPAH (*7*)), is 60 000 mg/kg or about 6% PAHs, see the Supporting Information (SI) for a table of these analyses). Coal-tar sealcoat is sold for application to parking lots in the U.S. at a rate of approximately 320 million liters per year (*8*). Recent biological studies indicated that abraded coal tar sealcoat was toxic to aquatic life (*9, 10*), and is implicated with localized PAH hotspots degrading stream communities (*11*). In 2006 the municipality of Austin, Texas, and Dane County, Wisconsin, passed bans against the use of coal tar sealcoat in their jurisdictions and the District of Columbia (Washington DC) passed a similar ban at the end of 2008.

Although the potency and ubiquitous nature of coal tar sealcoat as a source of PAHs to the environment has been demonstrated, understanding the rate and modes by which sealcoat-derived PAHs are released from pavement is not well understood. Such an understanding is essential in quantifying inputs of PAHs to urban water bodies, soils, and air relative to other PAH sources. The New York Academy of Sciences (NYAS) recently estimated contributions of PAHs to the New York/New Jersey Harbor, and although they concluded that coal tar sealcoat was a major contributor, they were not able to appropriately constrain loading from this source, indicating a very wide range, from 11 to 46% of the total PAH load to the harbor (*12*). Even that range underestimates true uncertainty, as it extrapolates single-event runoff yields measured by Mahler et al. (*4*), to annual yields per sealcoated area for the watershed, with no associated uncertainty. Most of the uncertainty in the NYAS estimate is attributed to a wide range in their estimate of annual application area, not wear rate.

Van Metre et al. (*13*) recently reported PAH concentrations in pavement dust from nine United States cities that are consistent with the Mahler et al. (*4*) results from Austin, suggesting a link between coal-tar sealcoat use and urban lake contamination by PAHs. They also showed evidence of off-parking-lot PAH transport by modes other than runoff, including, in one city, soil samples and street-dust samples from near coal-tar sealed pavement that were higher in PAHs than samples from near unsealed pavement. These authors also found higher PAH concentrations in dust from unsealed parking lots in cities where coal tar sealcoat dominates use than in dust from unsealed parking lots in cities where asphalt sealcoat dominates use (*13*), providing further evidence of widespread urban PAH contamination where coal-tar sealcoats are in use. These results indicate that coal-tar sealcoat is a PAH source with regional and perhaps national implications and that modes other than water runoff are important in the distribution of PAHs from coal tar sealcoats.

Field observations indicate that coal tar sealcoat applied to parking lots abrades and is removed from the lot on a time scale of months to years. The sealcoat wears unevenly with higher rates of loss in higher traffic areas where car tires abrade the sealcoat (Figure 1). Parking areas with little wear appear darker, and worn drive areas, where the aggregate shows through, appear lighter. This is because of the black color of coal tar sealcoat, which remains black over long periods of time (*14*), whereas, asphalt pavement binder oxidizes as it weathers, turning from black to gray (*15*), and exposed aggregate is generally light gray to light brown.

Recommended industry reapplication rates of every 2−5 years provide an initial estimate of sealcoat wear. Mahler et al. (*4*) performed an artificial wash-off study that estimated the yield of PAHs washing off parking lots for a single rain event based on measured concentrations and mass of particles from known areas. Although the study was not designed to determine annual wear rates, this value can be extrapolated using various assumptions (e.g., 44 rain events per year for Austin, 1.47 kg of sealcoat per m^2 applied per ASTM standard D3320), resulting in approximately 0.2% of

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FIGURE 1. Aerial photograph of a sealed parking lot, showing light gray wear pattern in drive area compared to darker park areas.

PAHs in sealcoat leaving the lot in stormwater runoff per year. The loading rates estimated by the NYAS used similar assumptions but with more rain events that resulted in a sealcoat wear rate of approximately 1% per year. This approach probably underestimates the actual annual removal rate, as the Mahler et al. (*4*) study used a gentle spray of distilled water (equivalent to 2.5 mm of rain with low rainfall energy), which is probably inadequate to represent the range of natural precipitation events. Further, the study sites were relatively small (50 m^2), gutters and other areas where abraded sealcoat would collect were avoided, and only one simulated event was conducted for each lot during a relatively rainy antecedent period, limiting the time for particles to accumulate.

When applied, the sealcoat forms a coating 0.25−1.0 mm thick on the parking lot surface at standard application rates (ASTM D3423-84, ASTM D3320-00). As the sealcoat wears and abrades, primarily by the weight and friction of car tires, the particles become mobile (Figure 2). There are three likely modes of transport removing these particles from the parking lot and delivering them to the environment: waterborne (runoff), Aeolian (wind), and mechanical (tracking by car tires, snow plows, etc.). Additionally, some PAHs escape through volatilization (*16*). To our knowledge, there are no studies which have attempted to integrate the effects of these dynamic processes in exporting sealcoat, and its PAH load, to the environment. We developed a novel photographic method to quantify the area covered by visible sealcoat on parking lots and used area covered, time since sealcoat application, and application rate to estimate long-term wear rate. The approach was applied to parking and drive areas and along a gradient of sealcoat ages from 0 to 5 years old. The amount of sealcoat determined to have been removed over a known period of time from 10 parking lots was used to estimate the annual wear rate. This wear rate was used to estimate PAH loading rates to the environment. Additionally, individual parking lot sealcoat loss was compared to estimated traffic volume to determine how parking lot use might affect sealcoat wear.

Materials and Methods

Site Selection and Sampling Plan. Ten coal-tar sealed parking lots in the Austin area where date of sealcoat application was known were selected for study. Parking lots were chosen to provide a distribution of sealcoat ages up to the industry-recommended maximum reapplication period of 5 years (Table 1a). The lots were sealed by four different contractors with three different coal tar-based sealcoat products and all lots were commercial, serving strip malls, high turnover restaurants, or large churches. The sealcoat products used were all national brand names that essentially use the same base ingredients based on ASTM standards D 3320, D 5727, and D 6946-03: RT-12 refined coal tar, colloidal clay, and water (see SI Table S2). All study lots had similar topography, being relatively flat, but slope was not a variable that was measured or used for control.

Each lot was divided into two areas: parking-space area (park), and drive-isle area (drive). An a priori 200-photo data set was selected to statistically represent wear patterns on the 10 study lots, distributed on the basis of the parking lot area, with a minimum of 3 and a maximum of 20 data points for each of the two area types, i.e., the smallest lot got three photos each for the drive and park areas and the largest lot got 20 each for drive and park areas. Maps of each study parking lot were generated with 50 randomly distributed and numbered dots that were used to identify target photo locations. Beginning with the lowest numbered dot on the map and progressing sequentially, the requisite number of photos was taken per lot. If a location was unavailable because of a parked car or other obstruction, or unsuitable because of obvious recent repair, paint, or oil spots, the next random dot location was used, until the stipulated number of photos was taken for drive and park areas. Among drive areas, there was no a priori distinction between straight and turn areas. Locations were randomly distributed in all areas so among the study lots, heavier wear areas and lighter wear areas should be appropriately represented (See SI Figure S1 for an example study lot showing park and drive areas and random photo locations and SI Figure S2 for examples of photos with different sealcoat coverage).

Four asphalt parking lots were also selected and photographed as reference samples using the same procedure as above, two with newly applied sealcoat and two that had never been sealed and were at least 5 years old (Table 1b). These control lots were used to calibrate photographs from the sealed lots to the appropriate "black" of sealcoat and "gray" of unsealed asphalt after sufficient aging had occurred. One of the reference parking lots was sealed with coal-tar-based sealcoat and the other with asphalt-based sealcoat due to the limited availability of new coal tar sealed lots.

Photography. All photos were taken with a Nikon D50 digital SLR camera. The camera was mounted on a tripod positioned with the back of the camera 1.46 m above the ground and the focal length of the lens set at 32 mm, giving a sampling frame of 1.05 × 0.69 m (0.725 m^2). For a consistent exposure, a 13.8 × 10 cm piece of 18% gray card (Kodak) was placed in the center of the frame and the camera was set to "Program" mode with the on-board light meter set to "spot" mode so that the gray card would have the same density in all images regardless of the ambient light level (Figure 3). All photos were 3008 × 2000 pixels (1.5 megabytes). To obtain even, flat lighting, the camera and subject were shaded by a large piece of nonreflective black mat board. All photos were taken from August 22 to September 21, 2006, and between 9:00 a.m. and 2:00 p.m.

Analysis. Images were cropped and converted to grayscale tiff format and analyzed with Scion Image (a repackaged version of NIH Image available at www.scioncorp.com) using the "density slicing" function, a one-dimensional classification technique that divides a grayscale image into classes based on brightness values. Density slicing was used to identify the black coal tar sealcoat in photographs of parking lot surfaces (Figure 3). This simple analysis separates dark or black pixels from lighter gray pixels into classes that can then be quantified by area and exported to spreadsheet and statistical software for further analysis. Scion Image was configured to select all pixels darker than the neutral gray card (the median in the gray scale) used in all photos (lower left-hand corner in Figure 3).

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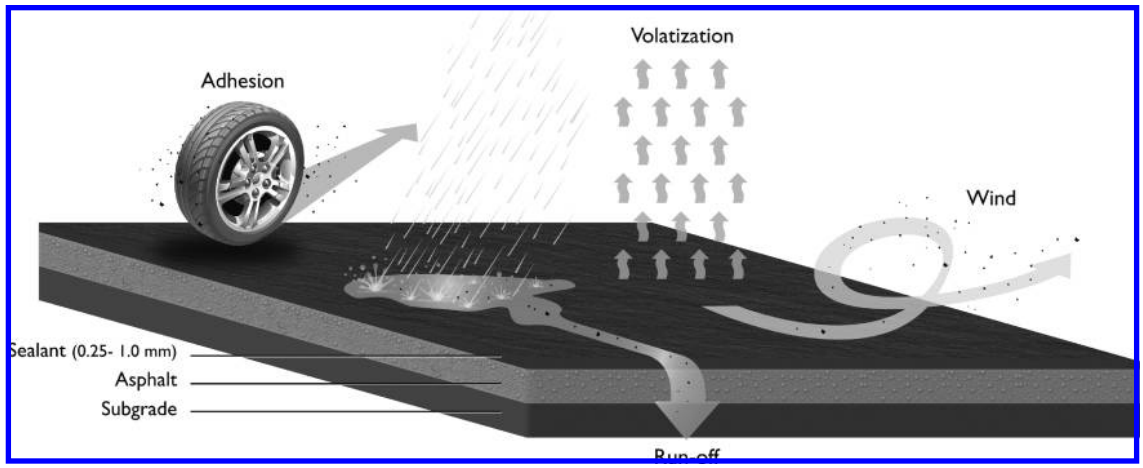

FIGURE 2. Generalized representation of the export of abraded sealcoat from an asphalt parking lot. Sealcoat is abraded into mobile particles, primarily by car tires, which are exported from the parking lot by water runoff, wind and by adhesion to moving objects (tires, feet, etc). Lighter PAHs also volatize off of lots as sealcoats cure.

TABLE 1. Age of Sealcoat Application, Parking Lot Areas by Use, And Number of Photos Taken Per Lot Per Area Use for Test (a) and Reference (b) Parking Lots

(a) test parking lots	age of sealcoat (years)	park area (m^2)	drive area (m^2)	no. park photos	no. drive photos
1	0.5	652	857	3	3
2	0.9	3590	2980	5	4
3	1.0	5299	7804	8	11
4	2.0	1230	2915	3	3
5	2.0	15578	16184	21[a]	19
6	2.2	5830	8752	8	13
7	3.7	4067	5714	6	8
8	4.1	10018	13136	14	20
9	4.1	3936	4122	6	6
10	5.4	7854	9800	6	7

(b) reference parking lots	Age of sealcoat (years)	Park Area (m^2)	Drive area (m^2)	# Park photos	# Drive photos
unsealed 1	n/a	2278	1635	3	3
unsealed 2	n/a	781	500	3	3
newly sealed 1 (asphalt)	0.1	239	599	3	3
newly sealed 2 (coal tar)	0.3	2822	3573	4	4

[a] Due to a field error, an extra park area photo was taken on lot no. 5, instead of the drive area photo, which would have provided the max of 20 photos each for this large lot.

A linear calibration curve was generated using the reference photographs from two unsealed parking lots (0% coverage of sealcoat) and two newly sealed parking lots (100% coverage). Drive areas and park areas were combined on these lots to simplify analysis and since there was no visible difference between them. There was some variability between the mean number of dark pixels on the two unsealed lots (80 000 vs 170 000 dark pixels), but very little difference between the dark pixel counts at the two newly sealed lots (433 488 vs 433 375 dark pixels). The equation of the calibration curve (no. of pixels/10000 = 0.308 × % sealcoat coverage + 12.6, $R^2 = 0.96$) was used to standardize all test lots to the same relative distribution of known sealcoat coverage versus raw pixel scores (Figure 4).

As an independent "common-sense" check, a series of quartiles of approximately 0, 25, 50, and 75% sealcoat coverage were selected by three observers from a coal tar sealed, but heavily worn, parking lot that exhibited a range of sealcoat coverage. The images were analyzed as detailed above, placed along the calibration line and generally had excellent agreement, except for the 0% sealcoat area, which had higher pixel counts than predicted by the calibration eq (245 692 actual vs 125 000 predicted dark pixels). This indicates that this method may not be accurate in very low levels of sealcoat coverage.

Estimation of Traffic Volume on Parking Lots. The number of times a parking lot was driven across was estimated according to the Trip Generation Handbook (*17*), which gives mathematical formulas for determining the number of trips a business will generate on the basis of business size and type. To enable comparison of lots, the total traffic volume of each lot was normalized by dividing by drive area to obtain the lot unit exposure to traffic: traffic volume = (daily estimate of trips × number of days since sealed)/(area of drive area of lot).

Results

A total of 174 data points were analyzed to calculate the amount of sealcoat coverage on the 10 study parking lots. There was a wide range of sealcoat coverage on lots of each age, with most lots having some spots with 100% sealcoat coverage, primarily in park areas, and a minimum sealcoat coverage of 51%, with the lower % coverage primarily in drive areas on lots sealed two or more years ago (Figure 5). The mean sealcoat loss per year for all lots (average sealcoat loss per lot/age of each lot) was 3.1% per year (standard deviation 2.8%) and the average rate of sealcoat wear from drive areas was significantly greater than from park areas by the Wilcoxon matched pairs test (4.7% per year vs 1.4% per year respectively, $p < 0.05$).

A linear relation between sealcoat wear and unit exposure to traffic volume was significant and explained 54% of the variation in observed wear rates (Figure 6A, $p = 0.009$, $R^2 = 0.54$). The relationship of traffic to wear was observed in both drive ($p = 0.01$, $R^2 = 0.50$) and park areas ($p = 0.02$, $R^2 = 0.44$) when analyzed separately. Generally, with each unit increase of traffic volume/m^2, there was a 0.01% increase in sealcoat loss.

Total sealcoat loss (park and drive areas combined) was significantly explained by time in a linear regression ($p = 0.03$, $R^2 = 0.39$), with an annual loss rate (slope) of 2.4% per year (Figure 6B). This relationship predicts that a 3-year-old lot would have 7.2% sealcoat loss and a 5-year-old lot, 12% loss. The oldest study lot, which had sealcoat that was 5.3 years old had an average sealcoat loss of 22%, seven % higher than predicted by the regression model but within the 95% confidence limits of the regression equation. Two of the lots,

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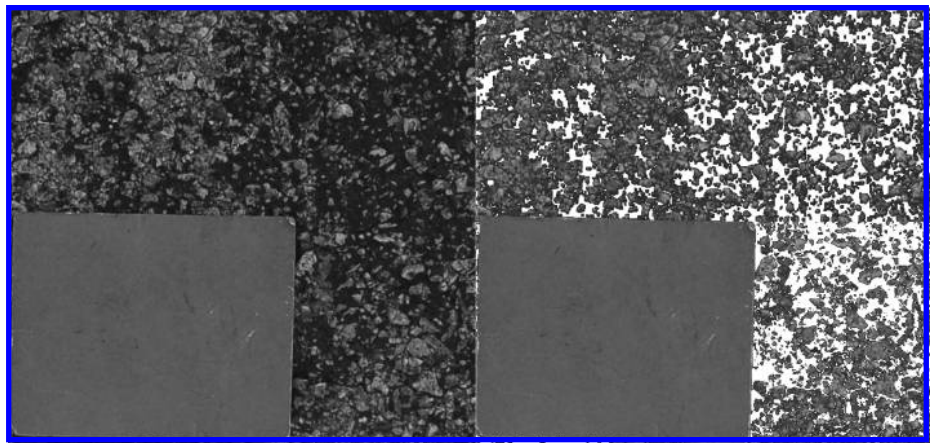

FIGURE 3. Example of density slicing technique, separating sealcoat from unsealed aggregate. The photo on the left shows an unaltered gray scale image where black represents sealcoat, while the photo on the right shows the separation of sealcoat from background aggregate, using a false white color. The gray square on the bottom left is a neutral gray card used to standardize image processing.

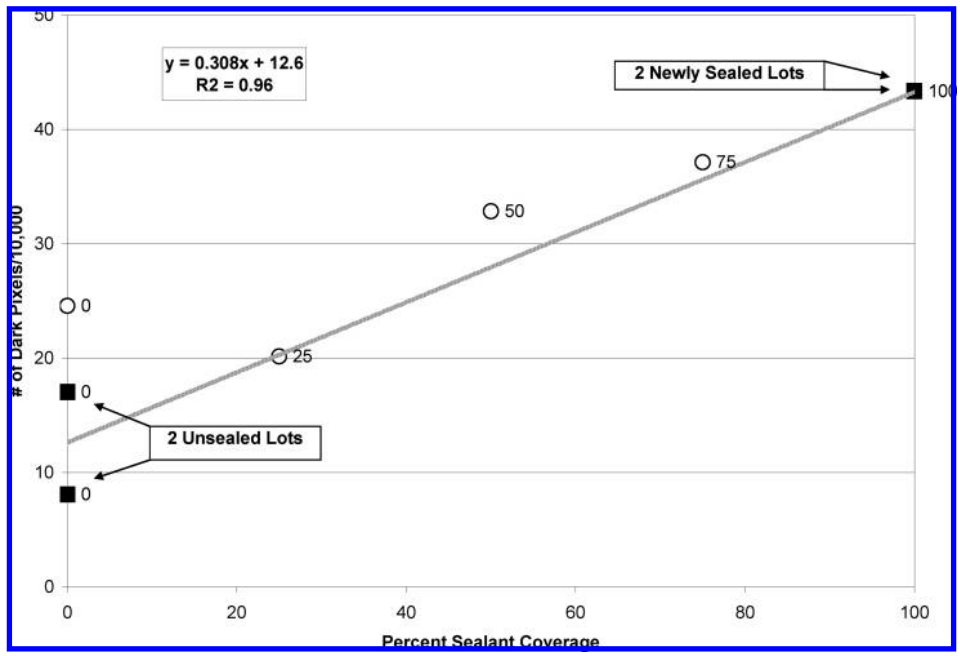

FIGURE 4. Two point calibration curve (two unsealed lots, two newly sealed lots) used to convert dark pixel values in photos to standardized % sealcoat on 10 study lots (R^2 = 0.96, p = 0.02). Open circles represent quartile check standards used to verify accuracy of the calibration curve. The two newly sealed lot data points are coincident on the graph.

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with sealcoat applications just over four years old, were outside confidence limits with sealcoat losses of 4.3 and 5.1%, less than the predicted loss of about 10%. The measured mean annual sealcoat loss (both park and drive areas) of 3.1% was higher than the 2.4% rate predicted by the regression model, but close in scale and the regression slope (2.4%) is within the 95% confidence interval of the mean (3.1% ± 1.7%).

PAH yield from sealcoated parking lots was estimated using the sealcoat wear rate regression model and the mean concentration of ΣPAH in scraping samples of sealcoat from coal-tar sealed parking lots in Austin, TX (*18*) and Milwaukee, WI (*19*). Scraping samples are dried, weathered sealcoat scraped from sealed pavement using a stainless steel spatula. Sampling and chemical analysis methods are presented in refs *18* and *19*. The mean concentration of ΣPAH from four scraping samples from Milwaukee is 14 500 mg/kg (SD 4200 mg/kg) and the mean from six parking lots in Austin is 14 500 mg/kg SD 5900 mg/kg). These lots are of variable and, in many cases unknown, age. The comparison of scraping samples to the mean concentration of ΣPAH in 12 coal tar sealcoat off-the-shelf products of 60,000 mg/kg (SI Table S1) suggests loss of approximately 75% of PAHs in sealcoat by volatilization (e.g., 60 000 mg/kg down to 14 500 mg/kg in aged sealcoat).

We can estimate the yield of PAH from sealed lots by water, wind, and mechanical pathways using the recommended application rates, the 14 500 mg/kg mean value of the dried scraping samples, and the sealcoat wear rate:

(1.47 kg of coal tar sealcoat applied per m^2 per ASTM 3423) X (14,500 mg/kg PAHs in dried sealcoat) X (the wear rate of 2.4% per year) = 0.51 $g/m^2/yr$.

This annual estimated ΣPAH yield is over 500 times greater than the single event runoff yield estimated by Mahler et al. (*4*) of about 0.0009 g/m^2, the mean from 6 parking lot wash-off yields, and 5 times greater than the annual yield estimated by the NYAS (*12*) of 0.11 $g/m^2/yr$.

Discussion

The approach presented in this paper offers the advantage of measuring total sealcoat loss, independent of the transport

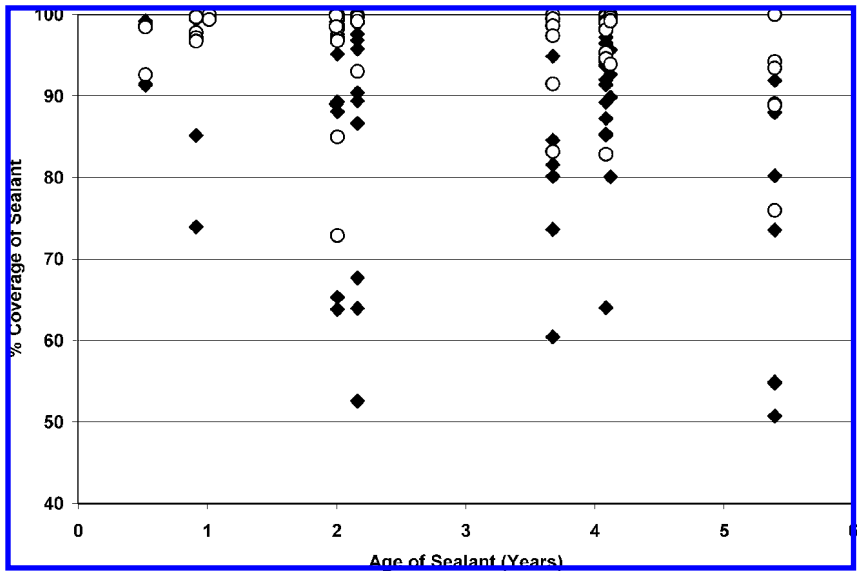

FIGURE 5. Percent sealcoat coverage on 10 study parking lots as a function of age in years. The vertical lines of data points (diamonds and circles), many of which are coincident on the plot, represent all data from each of the study lots. Solid diamonds are drive areas of the lots and open circles are park areas.

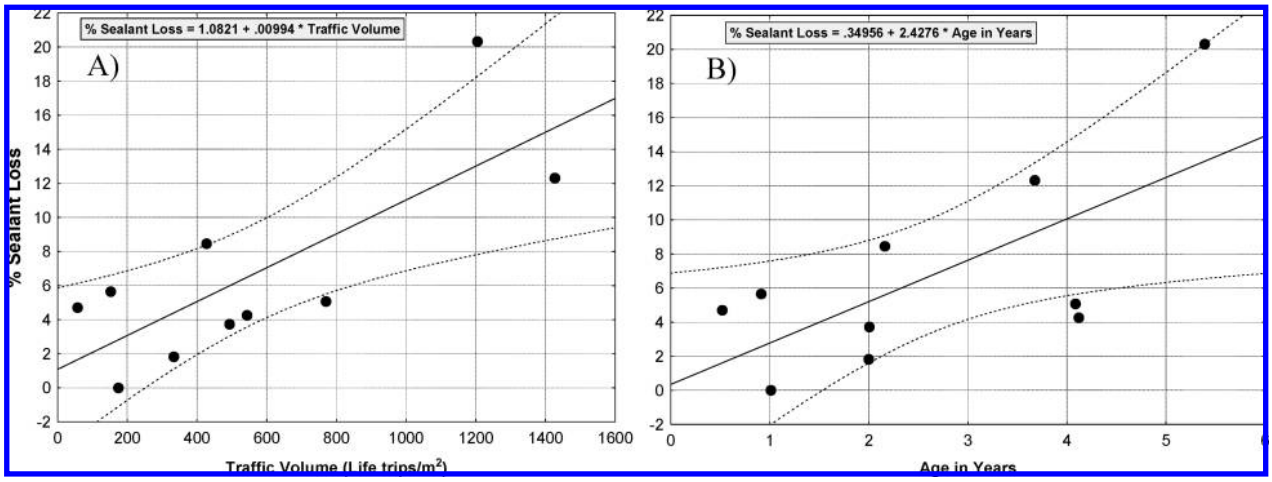

FIGURE 6. Relationship between sealcoat loss and A) traffic volume ($p = 0.009$, $R^2 = 0.54$) and B) age of sealcoat ($p = 0.03$, $R^2 = 0.39$) for 10 study lots. Dashed lines are 95% confidence limits.

process by which the sealcoat is removed (e.g., water, wind, mechanical). This is the first attempt we are aware of to systematically quantify total sealcoat and PAH loss over time from coal tar sealed parking lots. One limitation of this method is that sealcoat loss is only assumed to occur when the underlying asphalt aggregate is exposed. It is a two-dimensional method that can not account for losses in sealcoat thickness up to the point where the aggregate is exposed. On the basis of the industry standards (http://mspave.com/index-3.html, http://www.neyra.com/jenapp.htm, http://www.starseal.com/starseal.htm), the average newly sealed commercial parking lot has two coats of sealcoat in park areas and three coats in drive areas. This should result in sealcoat thickness of at least 1 mm, most of which would need to be removed before aggregate was visible. Thus, sealcoat loss rates estimated by this methodology are likely to be conservative.

It is possible that the top part of the aggregate might have a thinner coat of sealcoat than the valleys between the aggregate, resulting in visible wear after only a small amount of sealcoat has worn off. However, because multiple coats of sealcoat are generally reapplied every 1−5 years, this likely occurs only on very poor sealcoat applications (substandard thickness) and only after the initial application. The majority of our study lots had multiple sealcoat applications over their lifetime, suggesting that there are multiple layers of underlying sealcoat permanently filling the valleys for subsequent sealcoat applications. In addition, in high traffic drive and turn areas of parking lots, where most wear occurred, aggregate tends to be "polished" on the surface, resulting in a very smooth, slippery surface (*20*). Due to this polishing and the older layers of sealcoat filling valleys, drive areas are likely the smoothest place in the parking lot and visible aggregate there, in our photographic method, reflects loss of most if not all sealcoat that had been applied there.

There was considerable variability in this data set, both in the amount of sealcoat coverage over time within individual parking lots and among our 10 study lots, which is not surprising considering the wide range of influences that affect both sealcoat application quality and subsequent wear rates. In general we saw very little wear in parking areas, significantly more wear in drive areas, and a good correlation between estimated traffic volume and sealcoat loss. These data support the hypothesis that abrasion from tires was the primary influence on sealcoat wear on our study lots and may be helpful in generating best management practices to address this environmental problem. The sealcoat wear estimates presented here can be assumed to represent the minimum

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yields from sealed parking lots in the Austin area, which has very minimal freeze/thaw cycling and no snow plowing. In temperate climates where these phenomena occur more frequently, sealcoat wear rate and reapplication frequency are likely higher.

If quantitative estimates of sealed parking areas in a drainage area can be made, the wear rate of 2.4% per year, or a yield of 0.51 $g/m^2/yr$ of ΣPAH from coal tar sealed parking lots, may be the most appropriate method for calculating watershed annual loads from sealcoat. For example, the sealcoat industry estimate of 320 million liters of coal tar sealcoat sold each year in the U.S., assuming a conservative application rate of 1 L/m^2 (ASTM 4866, ATM 3423), translates to 320 km^2 of sealed parking lots and driveways and national annual yields of PAHs to the environment on the order of 160 000 kg. In Austin, Mahler et al. (*4*) mapped sealed parking lots in the Williamson Creek watershed and calculated that about 1.9% of the watershed or 1.5 km^2 was covered by sealcoat. This would correspond to about 800 kg/yr of ΣPAH delivered to the environment from this one suburban watershed, if all sealcoat used was coal tar-based. In neither of these examples are the fluxes of volatized PAHs from the sealcoat included. Comparison of product analyses and weathered sealcoat scraping samples suggests that volatilization losses could be very large, possibly up to 75% of initial product PAH concentration.

The results of this study, when compared to other sealcoat PAH studies, indicate a relatively rapid wear rate for coal-tar sealcoat from parking lots and suggest that there are several major PAH pathways from sealed pavement to the environment. To date, the only quantitative study of sealcoat PAH wear was for single-event yields in runoff. Sealcoat wear rates measured here further indicate that transport by pathways other than runoff (wind blown dust, mechanical transport) also could be quite large, possibly contributing to more general urban contamination by PAHs (e.g., soil contamination near sealed pavement).

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Supporting Information Available

Tables S1 and S2. Figures S1 and S2. This material is available free of charge via the Internet at http://pubs.acs.org.

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