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# Tire-Wear Particles as a Source of Zinc to the Environment

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Tire-tread material has a zinc (Zn) content of about 1 wt %. The quantity of tread material lost to road surfaces by abrasion has not been well characterized. Two approaches were used to assess the magnitude of this nonpoint source of Zn in the U.S. for the period 1936−1999. In the first approach, tread-wear rates from the automotive engineering literature were used in conjunction with vehicle distance-driven data from the U.S. Department of Transportation to determine Zn releases. A second approach calculated this source term from the volume of tread lost during lifetime tire wear. These analyses showed that the quantity of Zn released by tire wear in the mid-1990s was of the same magnitude as that released from waste incineration. For 1999, the quantity of Zn released by tire wear in the U.S. is estimated to be 10 000−11 000 metric tons. A specific case study focused on Zn sources and sinks in an urban−suburban watershed (Lake Anne) in the Washington, DC, metropolitan area for a time period of the late 1990s. The atmospheric flux of total Zn (wet deposition) to the watershed was 2 $\mu$g/cm$^2$/yr. The flux of Zn to the watershed estimated from tire wear was 42 $\mu$g/cm$^2$/yr. The measured accumulation rate of total Zn in age-dated sediment cores from Lake Anne was 27 $\mu$g/cm$^2$/yr. These data suggest that tire-wear Zn inputs to urban−suburban watersheds can be significantly greater than atmospheric inputs, although the watershed appears to retain appreciable quantities of vehicular Zn inputs.

## Introduction

Zinc (Zn) in the environment has both natural and anthropogenic sources. The fate and transport of Zn in soil, freshwater, and marine systems and the essential metabolic roles and toxicologic effects of Zn in biological systems have been reviewed by Nriagu and Davidson (*1*). The U.S. Environmental Protection Agency (EPA) has set the drinking water standard for Zn at 5 mg/L based on aesthetic (i.e., taste) criteria (*2*). At elevated concentrations, Zn has been shown to cause a range of reproductive, developmental, behavioral, and toxic responses in a variety of aquatic organisms (*3*−*7*). Maximum permissible concentrations of Zn in water, based upon toxicity to aquatic organisms, have been set by EPA at 120 $\mu$g/L as dissolved Zn (*8*).

The largest anthropogenic sources of Zn to the atmosphere are activities related to metal production; second-tier sources to the atmosphere include waste incineration, fossil fuel consumption, phosphate fertilizer, and cement production (Table 1). Zn sources to air and water related to transportation activities include deicing salts (*9*, *10*), combustion exhaust (*11*, *12*), galvanized parts and railings (*13*, *14*), fuel and oil (*11*, *15*), brake linings (*11*, *15*, *16*), and rubber tires (*9*, *12*, *17*). Tire-wear particles have been recognized for several decades as a source of Zn to the environment (*9*, *11*, *17*−*22*). Smolders and Degryse (*23*) recently reported on the fate of Zn from soil-applied tire debris. They showed that approximately 10−40% of the Zn in the <100 $\mu$m fraction of ground tire material was released to the labile pool during a 1-year weathering period.

Callender and Rice (*24*) examined sediment cores taken from reservoirs in the Chattahoochee River Basin in Georgia and Florida and showed that the anthropogenic Zn concentration correlated well with traffic density. They suggested that tire-wear particles in highway runoff might be an important Zn source in urban−suburban areas. Investigators looking at lower South San Francisco Bay have estimated that approximately 60% of the total load of Zn derives from tire-wear particles (*25*). Rothmann (*26*) indicated that the majority of the Zn in municipal wastewater-treatment plant sludges derives from tire-wear particles washed into storm-water sewers.

Although the potential health and environmental impacts associated with such traffic-related releases of Zn have received little public attention in the U.S., there has been considerable dialogue on the subject in western Europe (*27*−*29*). Arguments as to the importance of the problem have generally hinged on the method of estimating the source term. These debates have prompted current investigations into reducing Zn concentrations in tire rubber compounds (*30*−*32*). A 1994 study (Ahlbom and Duus, as cited in ref *33*) concluded that approximately 10% of the total particulate Zn load in Swedish cities came from tire wear. Tire wear in Sweden, from about 4 million motor vehicles in the late 1990s, was estimated to contribute about 150 tons of Zn per year to the environment (*16*). (With its approximately 200 million motor vehicles at this same time, one might expect, by extrapolation, release of about 7500 tons of Zn annually in the U.S. from tire wear.) A 1996 assessment in Great Britain used differences between new and used tire weights and estimated Zn release from tire wear to be approximately 1435 $g_{Zn}$/km/yr (*34*). An ca. 1980s German study of urban roadways estimated Zn release from tire abrasion to range from 14 $g_{Zn}$/km/yr on residential streets to 810 $g_{Zn}$/km/yr on high-speed motorways (*22*). Studies for the period 1995−2000 (*35*−*37*) estimated the mass of tire-wear particles generated on German roads to be approximately 60 000 to 65 000 tons/yr. (Note: In this paper, "tons" refers to metric tons.)

For the year 1970, Dannis (*17*) estimated that 720 000 tons of tire tread were worn off tires in the U.S.—roughly one-quarter, by weight, of the new rubber produced domestically in that year (*38*). Pierson and Brachaczek (*11*) estimated 600 000 tons of tire wear in the U.S. during 1973, and Cardina (*18*) estimated 320 000 to 640 000 tons for 1974. The purpose of this paper is to go beyond such single year estimates and to produce a multidecadal (1936−1999) estimate of Zn release to the environment from tire wear particles for the United States. We begin by presenting field evidence for associations of anthropogenic Zn in sediment cores with vehicular usage in a variety of urban−suburban settings in the U.S. We then present two independent approaches to estimate Zn release from tire wear to bracket the size of this source term.

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**TABLE I. Total U.S. Emissions (Tons) of Zinc (Zn) to the Atmosphere**

| source category | reference | year | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | 1960 | 1965 | 1970 | 1975 | 1980 | 1985 | 1990 | 1995 |
| cement production | (*87*), (*88*) | 617 | 716 | 729 | 667 | 735 | 754 | 752 | 846 |
| fertilizer production | (*87*), (*89*) | 1000 | 1000 | 1054 | 1329 | 1632 | 1525 | 1390 | 1365 |
| copper mining and production | (*87*), (*90*) | 1035 | 1163 | 1200 | 983 | 915 | 795 | 1185 | 1448 |
| iron and steel | (*87*), (*91*) | 1628 | 2160 | 2236 | 1952 | 1682 | 1223 | 1342 | 1374 |
| fossil fuel combustion | (*92*), (*93*) | 1532 | 1719 | 2141 | 1878 | 1916 | 1984 | 1658 | 1298 |
| waste incineration | (*92*), (*94*) | 6367 | 7280 | 5920 | 5006 | 3232 | 5659 | 7298 | 7941 |
| zinc mining and production | (*87*), (*95*) | 101 500 | 126 280 | 111 440 | 55 580 | 47 600 | 36 540 | 36 820 | 32 480 |
| total | | 113 679 | 140 318 | 124 720 | 67 395 | 57 712 | 48 480 | 50 445 | 46 752 |

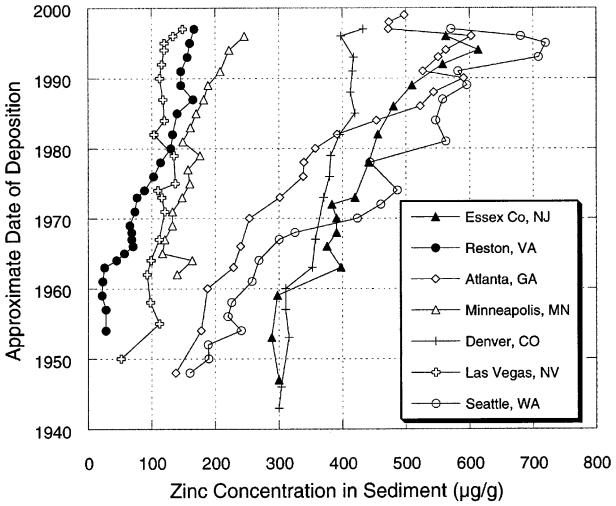


**FIGURE 1. Total sedimentary Zn versus approximate age of deposition in sediment cores from urban−suburban environmental settings.**

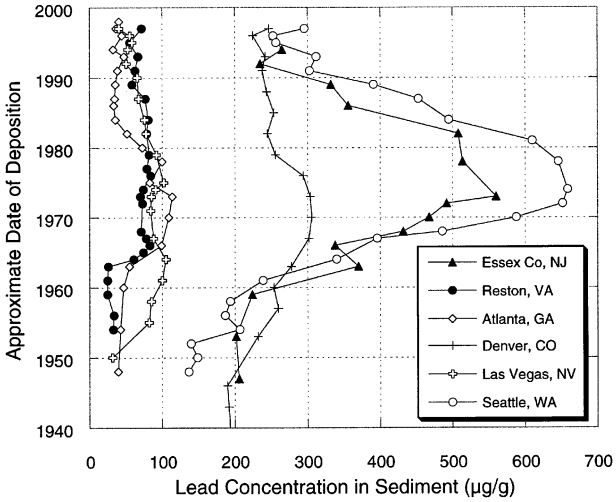


**FIGURE 2. Total sedimentary Pb versus approximate age of deposition in sediment cores from urban−suburban environmental settings.**

## Background

**Evidence for the Role of Tire Wear in Zn Contamination.** The distribution of total sedimentary Zn versus approximate age of deposition in sediment cores from urban−suburban environmental settings is shown in Figure 1. These data are published in Callender (*39*). Although there is quite a range in peak Zn concentrations (120−720 $\mu$g/g) and baseline (bottom of core) Zn concentrations (20−300 $\mu$g/g), it is clear that the total sedimentary Zn concentrations have increased with time over the past 50 years, coinciding approximately with increased urbanization in these watersheds. Data pertaining to the coring sites in these watersheds can be found at the U.S. Geological Survey (USGS) National Water-Quality Assessment Program, Reconstructed Trends National Synthesis Study Web page (http://tx.usgs.gov/coring; accessed December 1, 2003). A description of the sampling methods, analytical methods, and age dating can be found in Callender and Rice (*24*).

It is interesting to compare the total sedimentary Zn concentrations (Figure 1) to total sedimentary lead (Pb) concentrations in the urban−suburban cores (Figure 2). Both Zn and Pb concentrations increase for a time, suggesting a single source for both elements (*39*). However, the Pb concentrations peak in the 1970s when leaded-gasoline usage was at its maximum (*40*). Thereafter, leaded-gasoline usage declined rapidly into the 1980s, and this rapid decline is reflected in the total sedimentary Pb distributions at the top of the cores. On the other hand, total Zn concentrations increase to the 1990s.

Although the Pb and Zn distributions in sediment cores from urban−suburban environmental settings may be related to automobile use, the specific causes are quite different. The use and subsequent ban of leaded gasoline is responsible for the sedimentary Pb profiles, but some other traffic-related source appears to be influencing the sedimentary Zn profiles that indicate an ever-increasing release of Zn to the urban−suburban environment.

A more direct link between anthropogenic Zn and traffic density may also be made. We assumed that anthropogenic Zn concentration was the variable most related to traffic density. Furthermore, we assumed that the accumulation of anthropogenic Zn in sediment cores from lakes and reservoirs located in urban−suburban watersheds traversed by major roadways was a definitive measure of vehicle use. While the anthropogenic Zn concentration may be an adequate measure of its accumulation in sediment, the accumulation rate of anthropogenic Zn is the most direct measure of this metal's transport to and storage in the sediment column. Figure 3 is a plot of anthropogenic Zn accumulation rate versus average annual daily traffic for a number of watersheds across the U.S.

The anthropogenic Zn concentration is calculated as the difference between peak total Zn concentration at the top of the core (in the 1990s) and the baseline total Zn concentration at the bottom of the core (usually in the 1950s; Figure 1). The anthropogenic Zn accumulation rate in sediment cores is calculated by multiplying the anthropogenic Zn concentration ($\mu$g Zn/g sediment) times the mass sedimentation rate (g sediment/cm$^2$/yr) for each coring site. The mass sedimentation rate is calculated by summing the accumulated mass of sediment in the core to the depth of the $^{137}$Cs maximum (dated at 1964) and dividing this mass (g sediment/cm$^2$) by the number of years between the date when the core was taken and 1964.

The traffic density is represented by the average annual daily traffic (AADT, vehicles/day) for roadways that traverse

**TABLE 2. Measured Zn Content of Tire-Tread Materials**

| (Zn) (wt %) | samples | description | analytical method[i] | sample preparation[k] | reference | year |
|---|---|---|---|---|---|---|
| 0.04[a] | 1 | used tread particles | XRF | | (*12*) | 1991 |
| 0.06 | 1 | coarse ground tire | AAS | $HNO_3$ to pH 2, sonicate for 32 h, filter | (*71*) | 1998 |
| 0.20 | 1 | coarse ground tire | AAS | $HNO_3$ to pH 2, sonicate for 32 h, filter | (*71*) | 1998 |
| 0.40[b] | 10 | treads of discarded tires | AAS | 500 °C/3 h; $HNO_3$/90 min/100 °C | (*72*) | 1996 |
| 0.69 | 1 | "old" Goodyear A-78-13 | NA | | (*19*) | 1979 |
| 0.70 | 1 | radial steel belted tire | NA | | (*19*) | 1979 |
| 0.7 | 2 | tread "matrix" | EP | | (*96*) | 1964 |
| 0.72 | 1 | fine ground tire | AAS | $HNO_3$ to pH 2, sonicate for 32 h, filter | (*71*) | 1998 |
| 0.74 | 1 | ultrafine ground tire | AAS | $HNO_3$ to pH 2, sonicate for 32 h, filter | (*71*) | 1998 |
| 0.75 | 1 | belted tubeless tire | NA | | (*19*) | 1979 |
| 0.75 | 1 | Goodyear A-78-13 | NA | | (*19*) | 1979 |
| 0.80 | 4 | rerubbering powder[f] | AAS | 500 °C/4 h; 0.01M $HNO_3$ | (*33*) | 1999 |
| 0.84[c] | >4 | used car, truck, bus tires | NA | | (*10*) | 1986 |
| 0.84 | 1 | used car tread, outer 0.5" | AAS | 500 °C/4 h; 0.01M $HNO_3$ | (*33*) | 1999 |
| 0.85 | 1 | unused tire rubber | EDTA titration[j] | 550 °C/90 min; dissolve in HCl | (*97*) | 1972 |
| 0.87 | 1 | fine ground tire | AAS | $HNO_3$ to pH 2, sonicate for 32 h, filter | (*71*) | 1998 |
| 0.88 | 1 | fine ground tire | AAS | $HNO_3$ to pH 2, sonicate for 32 h, filter | (*71*) | 1998 |
| 0.96 | 12 | tire-wear particles | EDTA titration | | (*97*) | 1972 |
| 1.0 | >2 | automobile treads | NA | | (*11*) | 1974 |
| 1.0 | 11 | treads of discarded tires | NA | | (*15*) | 1974 |
| 1.01 | 1 | ASTM standard | NA | | (*11*) | 1974 |
| 1.02 | >1 | tire rubber | AAS | 550 °C; HCl + HF | (*13*) | 1999 |
| 1.06 | 1 | ultrafine ground tire | AAS | $HNO_3$ to pH 2, sonicate for 32 h, filter | (*71*) | 1998 |
| 1.26[d] | 1 | ground used tire rubber[g] | ICP−OES | microwave digestion | this study | 2001 |
| 1.33 | 1 | coarse ground tire | AAS | $HNO_3$ to pH 2, sonicate for 32 h, filter | (*71*) | 1998 |
| 1.35 | 1 | coarse ground tire | AAS | $HNO_3$ to pH 2, sonicate for 32 h, filter | (*71*) | 1998 |
| 1.40[e] | 1 | ground used tire rubber[h] | ICP−OES | microwave digestion | this study | 2001 |
| 1.46 | 1 | truck tread, outer 0.5" | AAS | 500 °C/4 h; 0.01 M $HNO_3$ | (*33*) | 1999 |
| 1.53 | 1 | scrap tires | not given | | (*98*) | 1986 |
| 1.55 | 1 | ground tire rubber | ICP−OES | digested in $HNO_3$ | (*77*) | 1996 |

[a] Hildemann et al. express doubt about the representativeness of this result. [b] Mean of 10 samples, five replicates each (range = 0.25−0.60 wt %). [c] Value given; range given = 0.65−1.1 wt %. [d] Mean of five replicates (range = 1.10−1.32 wt %). [e] Mean of seven replicates (range = 1.26−1.48 wt %). [f] Powder is derived from used tires. [g] Source: commercial tire recycling plant, Baltimore, MD. Cadmium content: 5.84 ppm. [h] Source: USDA-ARS Cotton Ginning Lab, Stoneville, MS. Cadmium content: 1.3 ppm. [i] Key to abbreviations: XRF = X-ray fluorescence; AAS = atomic absorption spectrometry; EP = electron microprobe; NA = neutron activation; ICP−OES = inductively coupled plasma optical emission spectrophotometry. [j] ASTM D297: Section 35, Referee Ash Test Method, and Section 47, Zinc Oxide. Note that total zinc is calculated as ZnO. [k] Where this column is blank, no sample preparation method is given. Methods such as XRF, NA, and EP are performed on the whole sample. The first temperature indicates ashing, with time where given. Where no concentration is given for subsequent acid treatment, full-strength acids were used. The time and temperature of acid treatment is noted unless the author states that the ashed residue was dissolved. The microwave digestion method is EPA 3052, in which samples are totally digested in concentrated $HNO_3$ and possibly HCl for 15 min using microwave heating. Handreck is unclear on the methods of sample preparation and analysis; the methods given here are used for other, similar, samples.

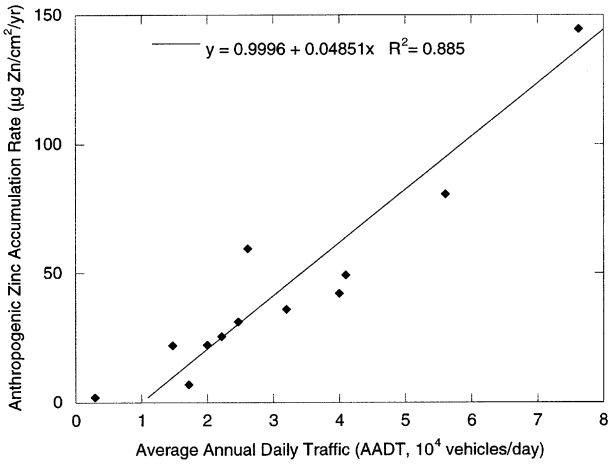


**FIGURE 3. Average annual daily traffic (AADT; vehicles/day) versus anthropogenic Zn accumulation rate ($\mu$g Zn/cm$^2$/yr). The anthropogenic Zn accumulation rate is the total Zn in the 1990s sediment minus the total Zn in the 1950s baseline sediment ($\mu$g Zn/g sediment), multiplied by the mean mass sedimentation rate (g of sediment per cm$^2$ of lake surface deposited annually).**

adjacent to the watershed coring site. AADT data for all but two of the sites shown in Figure 3 come from various state departments of transportation, and the data for the two other sites come from Shrank and Lomax (*41*).

The data plotted in Figure 3 come from 11 urban−suburban watersheds scattered throughout the Eastern U.S., the Midwest and Southwest, and the Western U.S. The datum close to the origin of the plot is from a rural watershed in Northwestern New Jersey. It is obvious that these data represent a strong relationship between anthropogenic Zn accumulation rate and average annual daily traffic ($R^2$=0.88). A similarly strong positive relationship was found ($R^2$=0.77) between anthropogenic Zn and traffic density in the Chattahoochee−Flint River Basin, Georgia (*24*) suggesting that there may be a causal relationship between anthropogenic Zn and traffic density.

**Zn in Tire Rubber Manufacturing.** Zn is added to tire-tread rubber mostly as zinc oxide (ZnO), and in lesser quantities as a variety of organozinc compounds, to facilitate vulcanization of the rubber. Analyses of the Zn content of rubber tires reported in the literature range from 0.04 to 1.55 wt % Zn, but both the mean and the majority of results can be described as roughly 1 wt % Zn. We analyzed two samples of ground tire material and found 1.26 wt % Zn (USDA sample) and 1.40 wt % Zn (Baltimore sample) (Table 2). No clear trend in Zn content of tire rubber as a function of year of sampling (~ year of manufacture) emerges from Table 2; the assumption of a 1% value for the entire time span seems reasonable. General information in the literature suggests that tire-tread formulations have historically included about 2.5% Zn (*42*−*48*). Although values higher than 1% Zn seem

**TABLE 3. Compilation of Tire-Wear Rates from Literature**

| wear rate ($g_{tread}$/km per tire) | comments | reference | year |
|---|---|---|---|
| 0.0012 | geometry estimate from assumed rate of 1 cm/32 000 km | (*19*) | 1979 |
| 0.006−0.019 | given as an estimate of mild wear | (*11*) | 1974 |
| 0.010 | given as typical wear rate for mild driving conditions | (*64*) | 1980 |
| 0.012 | measured by weight loss | (*51*) | 1985 |
| 0.017 | "light vehicles," geometry estimate; assumed rate: 68 mg/ vehicle km | (*13*) | 1999 |
| 0.018−0.174 | smooth treads on a range of surfaces; measured as depth of loss | (*53*) | 1942 |
| 0.023 | "gentle" driving | (*52*) | 1996 |
| 0.024 | given for cruise driving | (*17*) | 1974 |
| 0.026 | calculated value for 1 cm of wear per 64 360 km (40 000 miles) | (*25*) | 1992 |
| 0.03 | assumed value | (*62*) | 1979 |
| 0.03 | direct measurement by mass loss during simulated driving conditions | (*99*) | 1971 |
| 0.034 | "heavy vehicles" (>3.5 t); assumed: twice the rate for "light vehicles" | (*13*) | 1999 |
| 0.038−0.075 | direct measurement | (*11*) | 1974 |
| 0.041 | natural-rubber tires | (*100*) | 1944 |
| 0.042 | normal driving | (*52*) | 1996 |
| 0.054−0.072 | direct measurement by mass loss during simulated driving conditions | (*97*) | 1972 |
| 0.06−0.186 | estimated for urban conditions | (*11*) | 1974 |
| 0.07 | field data: tread depth measurements of 17 cars in Cincinnati, OH | (*99*) | 1971 |
| 0.073 | "hard" driving | (*52*) | 1996 |
| 0.09 | derived from assumed total tire weight loss (3.5 kg/ 40 000 km) | (*17*) | 1974 |
| 0.093 | given as "average U.S. wear rate" | (*11*) | 1974 |
| 0.49 | given for fast cornering | (*17*) | 1974 |
| 4.35−24.9 | given as typical wear rate for severe conditions | (*11*) | 1974 |

plausible, we have no supporting analytical data. In the absence of such data, we have opted to use the more conservative 1% Zn value throughout this study.

## Estimates of Zn Release

Estimates were made of the amount of Zn released from automobile tires due to tread wear to the road surface and environs in the U.S. from 1936 to 1999. Two approaches were used employing independent sources of data: (1) a wear-rate method, utilizing tread-wear rates from the literature with estimates of distances driven by various classes of vehicles, and (2) a geometric method, using annual U.S. tire production data (as a surrogate for number of tires disposed) with geometric estimates of the amount of tread gone at the time of disposal. A tread Zn content of 1% was used (Table 2) with both approaches.

**Wear-Rate Approach.** Annual distances driven (*49*) and the methods used to arrive at these estimates (*50*) were obtained for four classes of vehicles: passenger vehicles, buses, light trucks, and combination (i.e., tractor-trailer) trucks. Numbers of tires for each vehicle type were assumed: 4 tires for passenger vehicles, 6 for buses and light trucks, and 18 for combination trucks. (In 1966, the classification of pick-up trucks, vans, and sport utility vehicles was changed from light trucks to passenger vehicles (*50*). The pre-1966 inclusion of such four-wheel vehicles in our six-wheel category leads to some overestimates multiplying by six instead of four tires. We were unable to estimate the number of such vehicles within this group to correct the data. However, as these vehicles were a small percentage of the total distance traveled in either category, this should not affect the outcome of the calculations substantially.)

A range of tire-wear rates (0.006, 0.012, 0.05, and 0.09 $g_{tread}$/km) was selected from the literature (Table 3). These values provided a reasonable range to bracket our estimates because the median value for the wear rates in Table 3 is 0.04 g/km, and most of the values fall below 0.1 $g_{tread}$/km. In 1974, Pierson and Brachaczek (*11*) defined 0.006−0.019 $g_{tread}$/km as "mild wear" and 0.09 $g_{tread}$/km as an "average U.S. wear rate". A 1984 Texas roadway study by Pierce (*51*) using weight loss measurements showed an average wear rate of 0.012 $g_{tread}$/km. In a 1995 study using tire-testing machines, Sakai (*52*) measured tire wear under "gentle", "normal", and "hard" driving conditions and found wear rates of 0.023, 0.042, and 0.073 $g_{tread}$/km, respectively. We chose 0.05 $g_{tread}$/km wear rate as our best available value for average driving conditions. Interestingly, the weight of the tread (down to the 0.16 cm depth limit) calculated for the LT-235/75/R15 General Tire Ameri 550AS tire (3320 g), divided by the typical tire lifetime for passenger car tires (64 000−129 000 km or 40 000−80 000 miles), gives a wear rate of 0.025−0.05 $g_{tread}$/km, which agrees well with the range selected for consideration in the wear-rate approach. For each class of vehicle and year, the distance driven (km) was multiplied by the number of tires per vehicle and the tread-wear rate ($g_{tread}$/km). This value was then multiplied by the estimated Zn content of the tread material (1 $g_{Zn}$/100 $g_{tread}$) and summed for all vehicle classes, yielding $g_{Zn}$/yr released due to tire wear. It was assumed that the wear rate remained constant from 1935 through 1999. Although this assumption was made, we realize that tire life has increased greatly during this period and that the advent and widespread use of radial tires in the 1970s had a dramatic effect on tire performance. However, it is of interest to note that wear rates determined for pre-World War II tires (*53*) are not that much different from later rates given in Table 3.

**Geometric Zn Approach.** A tire schematic drawing used in the geometric approach is shown in Figure 4. Examples of the tread volume, weight, and Zn content calculated by this method for various sizes and styles of passenger and light truck tires are given in Table 4. From the tread footprint for a LT-235/75/R15 General Tire Ameri 550AS tire, the area occupied by solid tread (as opposed to void space in grooves) was measured by weighing a cut-out photocopied image. Tread wear down to the 0.16 cm (1/16 in.) tread-depth indicators (or "wear bars") was assumed (*54*). The calculated volume of tread lost ($cm^3$) was multiplied by the density of tire tread (1.18 g/$cm^3$) (*17*, *19*, *45*, *55*) and the estimated Zn content (1 $g_{Zn}$/100 $g_{tread}$) to obtain the mass of Zn in the tread of a tire. Calculations were based on the most common tire size and model in today's passenger car market, which was a passenger (P) or light truck (LT) 235/75/R15 tire with a 73-cm rim diameter (*38*). This yielded a value of 33 $g_{Zn}$ per tire in the tread. This value was then multiplied by the number of tires produced, giving the mass of Zn in tire tread produced in the U.S. each year.

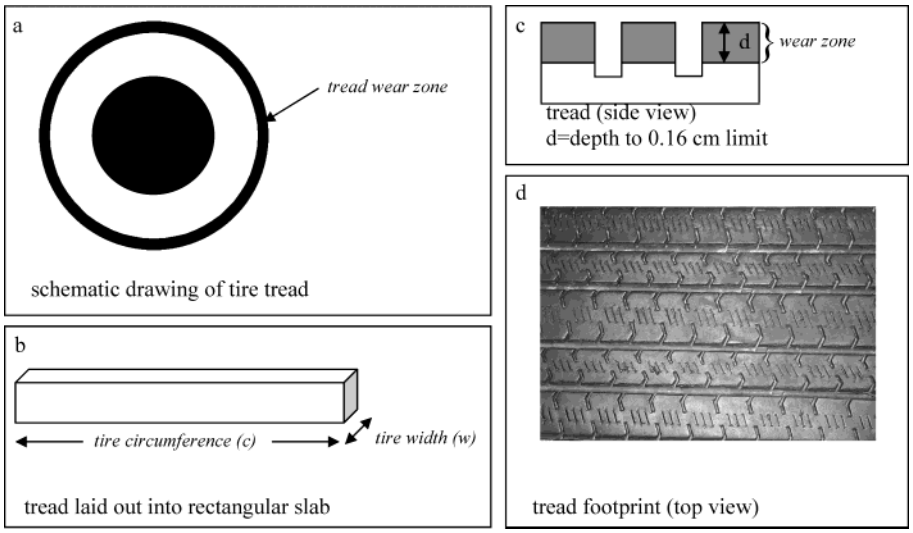


**FIGURE 4. Terms used in the geometric approach.**

**TABLE 4. Calculated Tread Volumes, Weights, and Zn Contents for Several Tires**

| manufacturer, brand | model | type | tire diameter (cm) | tread volume ($cm^3$) | tread weight (g) | Zn (g) |
|---|---|---|---|---|---|---|
| General Tire Ameri*550AS | Lt-225/75R16 | passenger | 74.4 | 3034 | 3580 | 35.8 |
| | Lt-235/75R15[a] | passenger | 72.1 | 2816 | 3323 | 33.2 |
| | Lt-235/75R15 | passenger | 71.1 | 2668 | 3149 | 31.5 |
| | Lt-235/75R15 | passenger | 77.5 | 3570 | 4212 | 42.1 |
| General Tire Ameri*GS60 | P-215/70R15 | passenger | 68.6 | 2122 | 2503 | 25.0 |
| General Tire XP 2000 H4 | P-215/60R14 | passenger | 61.5 | 1873 | 2210 | 22.1 |
| | P-205/60R15 | passenger | 62.7 | 1786 | 2107 | 21.1 |
| General Tire Grabber MT | 33 x 12.50R15 | small truck/SUV | 83.3 | 4398 | 5190 | 51.9 |
| Firestone Wilderness | H/T-P235/75R15 | small truck/SUV | 73.4 | 2292 | 2704 | 27.0 |
| Michelin | 9R22.5 | truck | 97 | 5093 | 6009 | 60.1 |

[a] This is currently the most popular model of passenger car tire in the U.S., according to ref *38*.

We assumed that the tires were disposed of when tread wear reaches the wear bars. It is of interest to note that in a U.S. tire disposal pile recently examined by Hartwell and co-workers, approximately 25% of the tires were not worn down to the wear bars, and approximately 25% were worn bald with no tread left (*56*). We further assumed that the number of tires disposed of at this level of tread loss was equal to the number of tires produced to replace them. This assumption is supported by comparing the number of tires produced for domestic use [tires produced in the U.S., adjusted for imports and exports] with the number of tires disposed of in the U.S. The disposed tire estimates were available for 1990−2000 (not all years) and were only slightly (5−15%) lower than produced/imported-tires data reported by the Rubber Manufacturers Association (RMA) (*38*). Thus the assumption appears justified. In addition, we assumed a constant Zn content in tire tread and a constant tire-tread density over the time period considered. Furthermore, the parameters for the current most common tire were used for all calculations. Thus, the results are also based on the assumption that popular tire size and tread area have not changed substantially since 1936. Although there are numerous tire sizes and a range of dimensions among tire brands and models, especially among truck tires, we chose a single popular size as indicated by the RMA (*38*) for the purposes of this study.

## Results and Discussion

The results of both methods for estimating tire-wear-related Zn released in the U.S. from 1936 to 1999 are shown in Figure 5. The wear-rate method estimates that 10 000 tons of Zn were released in 1999, using the selected wear rate of 0.05 $g_{tread}$/km. For this wear rate, total Zn released during the period from 1936 to 1999 is 285 000 tons; for the other wear rates considered (0.006−0.09 $g_{tread}$/km), the values for this period range between 30 400 and 513 000 tons. The geometric Zn method estimates 11 100 tons of Zn released in 1999 and 375 000 tons of Zn between 1936 and 1999. Given the different data sources and assumptions made before beginning these

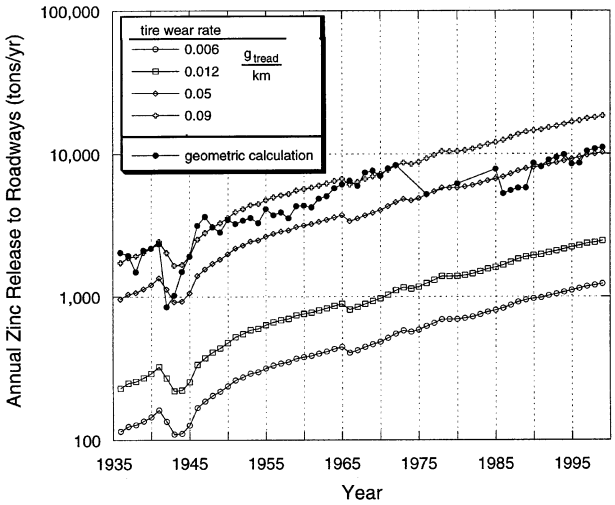


**FIGURE 5. Comparison of wear-rate and geometric approaches for estimating Zn release from tire-wear particles to roads in the U.S. (for some time periods, data were only available in 5-year increments).**

**TABLE 5. Comparison of U.S. Atmospheric Emissions Data to Estimates of Tire-Wear-Particle Zn Release for 1995**

| source category | Zn emission (metric tons) | % atmospheric emissions |
|---|---|---|
| total U.S. atmospheric emissions | 46 752 | 100 |
| nonferrous (Zn and Cu) production | 33 928 | 73 |
| wear-rate estimate (0.09 g/km) | 16 612 | 36 |
| wear-rate estimate (0.05 g/km) | 9 229 | 20 |
| geometry/tread estimate | 8 464 | 18 |
| waste incineration | 7 941 | 17 |
| fertilizer production | 1 365 | 2.9 |
| iron and steel production | 1 374 | 2.9 |
| fossil fuel combustion | 1 298 | 2.8 |
| wear-rate estimate (0.006 g/km) | 1 107 | 2.4 |
| cement production | 846 | 1.8 |

[a] See Table 1 for notes and references regarding atmospheric emissions data.

calculations, the wear rate and geometric Zn estimates agree fairly well, both for individual years and for the sum of the period between 1936 and 1999.

**The Relative Impact of Zn from Tire-Wear Particles.** In Table 5, the estimates for U.S. tire-wear release of Zn for 1995 are compared with the U.S. atmospheric emission sources listed in Table 1. The mean particle size of tire wear particles generated under driving conditions would appear to be about 10−20 $\mu$m (*17*, *99*). While the initial release of tire-wear particles is to the air, it is clear that these particles will settle from the air column at distances related to their aerodynamic diameters. The comparison in Table 5 is between the total releases of tire wear particles (all particle sizes) and a well characterized inventory of other atmospheric sources. Tire-wear particles released approximately 8500 tons of Zn in 1995, according to the geometric/tread Zn estimate. For a wear rate of 0.05 $g_{tread}$/km, the estimate is 9200 tons of Zn. Estimated release values of Zn from tire tread (using the wear-rate value of 0.05 $g_{tread}$/km) exceeded all other U.S. atmospheric emissions sources except nonferrous metal production. The 0.05 $g_{tread}$/km wear-rate estimate equals roughly 20% of the total U.S. atmospheric emissions of Zn for 1995. This is comparable to emissions from waste incineration and about seven times that from fossil fuel combustion.

**Release in Urban vs Rural Settings.** In 1999, U.S. urban roads comprised 3 050 000 km of lanes or about 23% of the total lane distance (13 158 000 km) (*57*). However, 60% of annual miles driven that year were on urban roadways (*58*). If Zn release is uniform over all roadways, then urban areas receive more Zn from tires due to the higher traffic volume and greater road density. Release in each setting (urban and rural) was estimated by multiplying traffic per lane kilometer by the total amount of Zn released from tire wear and dividing by the lane-km of road in that setting, yielding the mass of Zn released (Figure 6). It should be noted that the Federal Highway Administration (FHWA) only reported "road length" (e.g., 1 km of two-lane highway is 1 km of road length) prior to 1980. To convert this to lane-km (measure of length of all lanes; i.e., one km of two-lane highway is two lane-km), two lanes were allotted for each km of road length. The calculated lane-km values slightly underestimate (by less than 5%) the actual lane-km distance, as some roads contain more than two lanes. In addition, the wear rate for tires is higher in urban environments than in rural environments due to more frequent stopping and cornering; this is not taken into account in the estimates. Despite these limitations, the estimates clearly show that road length-normalized Zn release from tires in urban environments is significantly higher (by an order of magnitude) than that in rural environments. Baumann and Ismeier (*36*, *37*) have estimated that about

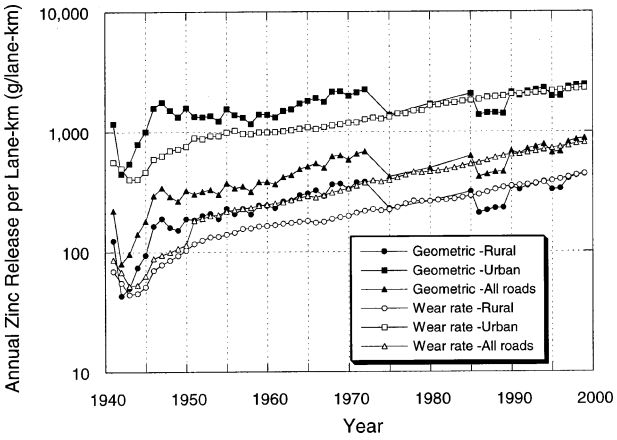


**FIGURE 6. Annual release of Zn per lane-km to rural and urban roadways in the U.S. estimated by the wear rate (0.05 $g_{tread}$/km) and geometric methods (for some time periods, data were only available in 5-year increments).**

1000−3000 g of Zn from tire wear are deposited annually per road-kilometer in Germany (note: this estimate used a 2% Zn content for the tread material). These loadings are in general agreement with contemporary U.S. estimates for all roads shown in Figure 6.

Lake Anne (Fairfax County, VA) is a reservoir constructed for stormwater control and recreation. It was filled in 1962 as part of a real estate development project. The watershed is relatively densely populated with condominiums and single-family homes. A geochemical study of Lake Anne (*59*) identified the major Zn inputs to the watershed as atmospheric precipitation directly to the lake surface and stream transport which includes atmospheric deposition to and transmission through the catchment, runoff from impervious surfaces, and weathering components. The major Zn outputs are fluvial export (one tributary drains Lake Anne) and sediment accumulation. Based on measurements of total Zn concentrations in water sampled regularly during 1998 at the inflow and outflow streams of the lake, we estimated the annual fluvial import to be 44 $\mu$g Zn/cm$^2$/yr and the annual fluvial export to be 13.2 $\mu$g Zn/cm$^2$/yr; these values are normalized to the area of the watershed.

We assume that the difference between the annual fluvial fluxes (30.8 $\mu$g Zn/cm$^2$/yr) combined with the total wet deposition directly to the lake surface (0.41 $\mu$g Zn/cm$^2$/yr) (*60*) is retained in the sediments of Lake Anne as the net annual accumulation of Zn (31.2 $\mu$g Zn/cm$^2$/yr). This value compares well to the measured annual estimated accumulation of Zn for the years 1990−1995 in the lake sediment (27.4 $\pm$ 1.2 $\mu$g Zn/cm$^2$/yr), as measured in three Lake Anne sediment cores (E. Callender, unpublished data). This flux is normalized to the depositional basin-surface area to facilitate direct comparisons with the watershed; the methodology for comparing watershed mass balances with lake sediment mass accumulation is described by Rice et al. (*61*). The estimated annual anthropogenic (total concentration minus background concentration) Zn accumulation measured in the sediment cores (22 $\pm$ 1 $\mu$g Zn/cm$^2$/yr) is about 70% of the net annual accumulation of Zn (based upon the mass balance above).

The input of anthropogenic tire-wear Zn to the Lake Anne watershed is calculated to be 42 $\mu$g/cm$^2$/yr. This value was arrived at by summing all roadway lengths (km) that border the watershed (urban roadways) and multiplying the total by the 1998 annual Zn release to urban roadways (Figure 6); all roadway lengths within the watershed (rural roadways) were summed, and the resultant total length multiplied by the 1998 annual Zn release to rural roadways (Figure 6). The total quantity of Zn released for 1998 was divided by the

watershed area, thus resulting in the above value for anthropogenic Zn release to the watershed. This value, 42 $\mu g/cm^2/yr$, is nearly double the anthropogenic Zn accumulation rate for Lake Anne sediments (22 $\mu g/cm^2/yr$). There may be many reasons for this discrepancy. We used the average tire wear rate (0.05 $g_{tread}/km$, Table 3) for the above calculations. Using the other wear rate values considered in Figure 5, the anthropogenic Zn release from rubber tire wear could easily vary between 5 and 75 $\mu g\ Zn/cm^2/yr$. Actual wear will vary with vehicle and tire type, driving conditions, road factors such as pavement texture, and environmental factors such as ambient temperature. For example, a test in central Texas in the mid-1980s with four brands of passenger car tires found that summer wear rates were 24−37% higher than winter rates (*51*).

When considering the fate and transport of tire-derived Zn, other very important factors are the leaching of Zn from tire-wear particles, the sorption of this dissolved Zn by surficial soils, and physical retention of tire-wear particles by vegetation and other components of the land surface. Undoubtedly these processes will retard the movement of Zn in the watershed toward Lake Anne and reduce the anthropogenic Zn accumulation rate in the sediment column. On the other hand, a significant area of the Lake Anne watershed is covered by parking lots of townhouse clusters and these impervious surfaces would promote relatively free flow of material to the lake. Finally, particulate Zn release by tire wear may not be the only source of anthropogenic Zn in the Lake Anne watershed. A recent assessment of Zn fluxes to the environment in Sweden (*16*) estimated corrosion of vehicle components and release of brake-wear debris as other vehicular sources of Zn; the corrosion source term has not been well characterized. The Zn contribution from brake wear has received more attention. However, the composition of brake pads and shoes has changed over time with the elimination of asbestos-bearing friction materials and a shift toward a variety of semimetallic, low metallic, and nonasbestos organic materials in recent years. A multidecadal assessment of this brake-wear source term is complex and beyond the scope of this report.

The strong positive relationship between anthropogenic Zn accumulation rate of sediments in lakes and reservoirs impacted by major roadways and the average annual daily traffic for these roadways (Figure 3) supports vehicular releases as a likely source term. As noted above, this vehicular source cannot be unambiguously attributed to tire wear. Not withstanding the uncertainty of estimates of tire wear rates and the transport of released materials to a receiving water body, the above data show that significant amounts of Zn are released to the watershed in high-traffic areas such as the Lake Anne environs.

**Fate, Transport, and Bioavailability of Tire Wear Particles and Associated Zn.** To better access environmental dispersion of intact tire-wear particles, we need a better knowledge of their particle size distribution under actual driving conditions, their atmospheric and fluvial resuspension properties once deposited on the land surface, and their reactivity in the soil and aqueous environments. The majority of the tire-wear particles tend to settle close to the roadway (*17*, *62*, *63*). Cadle and Williams (*62*) estimated that if 80% of the generated material from one highway settled evenly over a 5-m-wide strip on either side of the road, a layer of rubber 0.5 mm deep would accumulate in a year (assuming no movement or degradation). In contrast, the amount of material found next to that highway (based on the styrene-butadiene rubber content of soil samples) represented only 5 months' accumulation. Once deposited on pavement and adjacent soil surfaces, tire-wear particles are subjected to physical removal by fluvial and atmospheric processes and to degradation by abiotic and microbial processes (*17*, *62*, *64*).

Elevated levels of Zn in aqueous solutions, pharmaceuticals, and biological fluids exposed to rubber have long been recognized in the biomedical community. Zn compounds used as vulcanization accelerators in the manufacture of rubber gloves (*65*) and latex urinary catheters (*66*) have been implicated in instances of toxic reactions in humans. Zinc dimethyl- and dibutyldithiocarbamate accelerators in latex rubber are thought to immobilize sperm in latex condoms (*67*). Samples of drugs and blood products in contact with rubber stoppers and syringe plungers have shown high concentrations of Zn (*68*, *69*). Thus, it appears that Zn in a variety of rubber products exists in a readily bioavailable form or forms.

Studies suggest that Zn in whole and ground-rubber tires may also be mobile and bioavailable. Zn in tire rubber occurs in at least two chemical forms with differing displacement behavior by cations in aqueous solution (*70*). Elevated Zn levels have been demonstrated in leaching experiments using tire rubber and simulated rainwater (*71*), synthetic acid rain (*72*), lake water (*73*, *74*), and synthetic seawater (*75*). As compared to recently applied soluble Zn salts, the fraction of the Zn in tire debris (exposed to 11 months of outdoor weathering in Belgium following soil application) that was labile in the pore water was 10−40% (*23*). Tissue of plants grown in soils amended with ground tire material showed elevated concentrations of Zn (*76*−*79*). Microorganisms inhabiting soil and water can probably influence the mobilization of Zn from tire rubber in two distinct ways. Microbes may degrade the rubber hydrocarbons and vulcanized materials (*80*−*83*), thereby removing the substrate binding the Zn. Alternately, metabolic products from microbial activity (e.g., pyrite oxidation) may enhance leaching of Zn indirectly by changing the local environs of tire-wear particles (e.g., by lowering pH).

In addition to tires, there are numerous other sources of Zn to highway runoff waters. These include brake-pad dust, oil, and deicing chemicals (*84*). The yearly tonnage of Zn consumed in the U.S. that goes to tires is relatively small (4.75% in 1999). Roughly half the Zn consumed in the U.S. goes into galvanization of iron and steel to prevent corrosion (50% in 1999). Of the remaining half, about 17% goes into Zn-based alloys, 13% into brass and bronze, and 20% into nonmetallurgical applications (e.g., rubber, paint pigments). Galvanized metal has numerous outdoor applications, such as highway guardrails, roofing materials, and automobile components, that can be sources of dissolved and particulate Zn to the watershed. For example, rainwater coming from the rusty, galvanized metal roof of a building on the Washington State coast was shown to have a dissolved Zn concentration of 11 900 $\mu g/L$ (*85*).

Road dust consists of local soils, pavement particles, vehicle exhaust-particles, rust from vehicles and highway structures, tire-wear particles, and deicing agents. Tire-wear particles make up about one-third of the vehicle-derived particulates in highway runoff. Because the fine road dust can be mobilized by wind and vehicle turbulence, atmospheric aerosols in the urban environment tend to be similar in composition to fine road dust (*84*). Watersheds receive tire-wear particles from this aerial fallout as well as overland runoff and point-source discharges of stormwater. Our results have attempted to quantify the input source term (i.e., at the road surface, at the time of generation) in the U.S. from the pre-World War II era to the present. We estimate that about 285 000 tons of Zn were released from tire wear in the U.S. between 1936 and 1999 and that about 10 000 tons of Zn were released in 1999 alone. With their higher density of roads, high volumes of vehicular travel, and higher rates of tire wear, the release of Zn from tire-wear particles is greatest

in urban watersheds. We estimate that urban tire-wear release of Zn ($g_{Zn}$/lane km) is roughly 10 times as great as in rural settings.

In future studies, it would be of interest to track Zn and organic signature compounds for tire-wear particles in sediment cores from lakes in watersheds with differing land uses. Some molecular markers such as styrene-butadiene rubber will track the tire-wear particles directly and act solely as particle phase tracers. Semivolatile tire pyrolysis products, such as benzothiazole, may also be useful as molecular tracers. Analysis of environmental samples for solvent-extractable benzothiazole will assay for compounds trapped within the tire-wear particles and compounds that have sorbed to a variety of road-dust particles (*86*). Zinc in sediment cores should be viewed as more like this latter case, in that tire-wear particles released to a roadway may give rise both to residual particles with entrapped Zn and to leaching and subsequent sorption of Zn on soil and anthropogenic particles.

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