The Role of Resuspended Soil in Lead Flows in the California South Coast Air Basin

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The inputs and outputs of airborne lead in the South Coast Air Basin of California (SOCAB) are quantified according to standard mass balance calculations. Results for 2001 show that approximately 49 000 kg of lead exit the Basin each year, but traditional sources contribute only about 6500 kg of lead each year. We resolve this discrepancy through a simple computer model that quantifies the resuspension of lead-containing particles. Our results suggest that these lead particles were deposited during the years of leaded gasoline use and that resuspension is responsible for generating an additional 54 000 kg of airborne lead each year. This agrees roughly with estimated outputs. Thus, we conclude that resuspension, although an insignificant source of airborne lead during the era of leaded fuel, became a principal source in the SOCAB as lead emissions from vehicles declined. The results of the resuspension model further suggest that soil lead levels will remain elevated for many decades, in which case resuspension will remain a major source well into the future.

Introduction

In the South Coast Air Basin (SOCAB) of California, which includes Los Angeles and suburbs, the first mass balance for lead was performed by Huntzicker et al. (*1*). At the time of that study in 1972, leaded gasoline was universally used and was responsible for virtually all of the airborne lead emitted.

An update of this work was performed by Lankey et al. (*2*) based on data from 1989. Although leaded gasoline use was reduced significantly from 1972 values, the Lankey et al. mass balance showed that gasoline combustion was responsible for approximately 50% of the overall lead emissions. They also considered the contribution of road dust, which is known to have an elevated lead mass fraction. Resuspension contributed approximately 40% of the overall lead emissions according to their estimates.

Additionally Ehrman et al. (*3*) in their receptor modeling in the SOCAB reported that dust and soil particles contributed a considerable amount to the PM$_{2.5}$ lead, although the actual quantity varied substantially. Young et al. (*4*) resuspended various southern California soils in the laboratory. Their results also indicate that resuspended lead particles contribute significantly to airborne concentrations.

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To consider how much lead is available for resuspension, we looked at estimates of gasoline consumption since 1970 (*5*) in conjunction with the lead content of gasoline (*6*), fraction of lead emitted from a vehicle (*6*), and deposition rates onto surfaces of the Basin (*7*−*9*). This indicates that more than 20000 mt (metric tons) of lead was deposited during the decades of leaded gasoline use. Elevated lead concentrations in the soil reflect this.

The average concentration of natural lead in crustal rock is reported as 12.5 ppm, which may be considered a baseline value (*10*). Page and Ganje (*11*) report soil lead levels in the SOCAB in 1919−1933 as 16 ± 0.5 ppm, close to the baseline. Concentrations in the SOCAB between 1967 and 1970 averaged 79 ± 23 ppm (*11*−*13*). Young et al. (*4*) report soil lead in Pasadena averaging 77 ± 19 ppm in 2001, showing that soil concentrations have remained high despite the shift to unleaded gasoline.

To determine if these high soil lead levels are contributing to the airborne concentrations, we use a mass balance model where sources of lead other than soil are known. First, the emissions of lead in 2001, our modeling year, are considered in each of several source categories. The sinks of the emitted lead are then analyzed. A simple mass balance model is developed next, using source and sink data. Finally, we use a computer model to assemble a time series for airborne and soil lead, and we use this information to estimate the contribution of soil resuspension to the overall mass balance.

Emissions

The main sources of lead in the SOCAB (see Figure 1) are assumed to be stationary point sources, on-road vehicles, aircraft, and contaminated dust and soil. We consider each of these categories for 2001, the most recent year for which we have relatively complete data. Although not warranted by the accuracy of the data, values are given to three significant figures for bookkeeping purposes. In addition, more significant figures are sometimes used in the original calculations to avoid rounding errors.

Emissions − Stationary Point Sources. Three sources of information on particulate lead emissions from SOCAB point sources are considered. The California Air Resources Board (CARB) reports emissions of 5410 kg/year as shown in Figure 2 (*14*). Manufacturing dominates the emissions; facilities in this category include lead-acid battery plants, smelters, lead alloy production facilities, and other industries.

The South Coast Air Quality Management District (SCAQMD) reports air emissions data for lead through their Annual Emissions Report. The emissions for the SOCAB total 2080 kg/year for fiscal year 2000−2001 and 2020 kg/year for fiscal year 2001−2002 (*15*). Although the area of the SOCAB according to the CARB and the SCAQMD is significantly larger than that shown by Huntzicker et al. (*1*), the area in Figure 1 nevertheless encompasses about 90% of the point source emissions of lead by mass (*14*, *15*) and about 80% of the population in the region (*16*).

The U.S. EPA Toxic Release Inventory (TRI) database meanwhile reports discharges of lead and lead compounds by county. The total for Los Angeles, Orange, San Bernardino, and Riverside counties is 6010 kg in 2001 (*17*). Note that the heavily populated portions of these four counties lie within the boundaries of the SOCAB, but large sparsely populated areas lie in the surrounding desert and mountain areas.

Because of the greater detail in the CARB database, we use their analysis here rather than the other available data (*18*). However, because quantitative uncertainties are not available for any of the individual point source emissions,
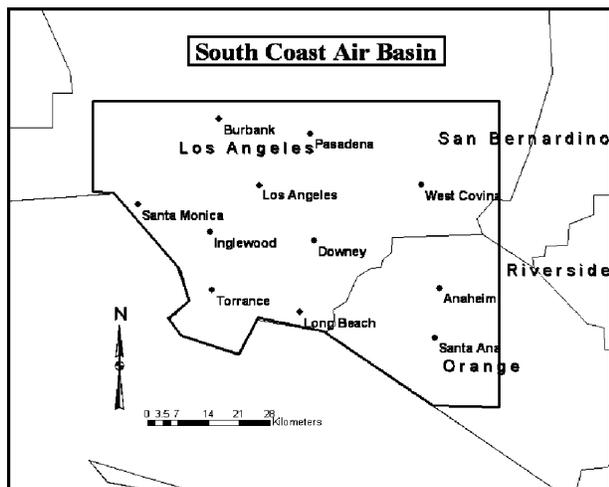
FIGURE 1. Map of the portion of the South Coast Air Basin used in this study taken from Huntzicker et al. (1).
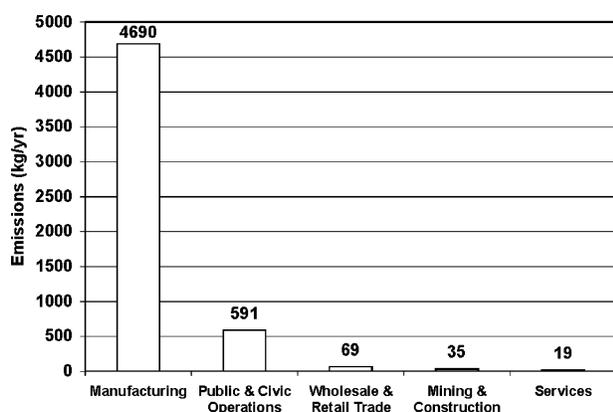

FIGURE 2. 2001 SOCAB point source emissions of lead by facility type (14). Data are from 2001.

we assign an overall uncertainty of ±50% to the CARB total, although the true uncertainty may be even greater.

Emissions − Motor Vehicles. Lead is no longer added to automotive fuel, but traces of lead are nevertheless present in gasoline. These trace amounts are significant in the SOCAB because of the large quantity of motor vehicle traffic.

Overall gasoline and diesel sales for the state of California in 2001 were 56.0 and 9.95 billion liters, respectively (5). We assume 36.5% of these values apply to SOCAB based on the fraction of California's population there (16). The average lead contents of gasoline and diesel fuels were 0.00005 and 0.0003 g/L, respectively, in 1996, the last year for which we have data (6). The uncertainty in lead content of diesel fuel is about 67% (6), and the uncertainty in lead content of gasoline is estimated to be 100% based on measurements of lead in unleaded fuel conducted in earlier years by the California Air Resources Board (2). The uncertainty is largely due to natural variation in the lead content. About 60% of the lead is deposited in the engine and exhaust system of vehicles; the remainder is emitted (6). Therefore, overall motor vehicle lead emissions in 2001 were about 848 ± 505 kg.

There is also evidence that lead weights used to balance wheels (19) and the attrition of brake pads (20) may contribute to lead in road dust and thus to airborne lead. These sources are neglected in the current analysis.

Emissions − Aircraft. Aircraft operating with piston engines consume avgas, which contains between 0.1 and 1.0 g of tetraethyllead additives per liter (21). Nearly all piston-engine aircraft are small planes categorized as general aviation (GA)

TABLE 1. Average Time Spent in Each Mode of a GA Flight (27)

mode	average time-in-mode (min)
takeoff	0.3
climbout	5.0
approach	6.0
taxi/idle	16.0

aircraft. These planes tend to fly locally, keeping emissions close to takeoff and landing sites. GA aircraft also include nonpiston engines that use jet fuel as explained below. In 2002, the only year for which we have data, 3.56 million GA landing-takeoff (LTO) cycles occurred at 28 SOCAB airports (22). We assume that the number of LTOs in 2001 was the same.

We use the Emissions and Dispersion Modeling System (EDMS) of the Federal Aviation Administration (FAA) to estimate lead emissions from SOCAB aircraft. Of the approximately 200 different models of GA aircraft, the EDMS software provides data for only the three most common models: Cessna 172, Piper PA28, and Cessna 150. These three models are all piston engine planes and constitute 31.5% of the GA fleet (23). According to the FAA (24), 32.7% of the GA fleet burns avgas, while the rest burns jet fuel, which does not contain lead additives. We assume that both of these percentages apply to 2001. The EDMS gives emissions of sulfur oxides for the three models listed above, and these emissions are directly proportional to the original sulfur content of the fuel. Although emissions vary depending on the flight mode, we assume that lead and sulfur oxides emissions should be in approximately constant proportions throughout all modes. This is reasonable because emissions of both pollutants are highest when the engine power is greatest, during takeoff and climbout (25). The proportion of sulfur to lead mass in the most common type of GA fuel, 100LL avgas, is 0.739 (21). According to the EDMS, the three aforementioned aircraft models emitted 902 kg of sulfur dioxide into the airspace above the SOCAB in 2001 (26).

To determine the amount of lead emissions that is released below the mixing height, we use the average time-in-mode as listed in Table 1.

We calculate an average flight time of 48.9 min from the FAA estimate of 4.53 million hours of GA flight time in the western U.S. spanning 5.56 million flights (24). Therefore, an average LTO cycle is 64.9 min (48.9 min in flight, 16.0 min in taxi/idle) of which approximately 27.3 min or 42.1% is spent below the mixing height.

To find overall emissions from aviation, we use the 42.1% value and assume the emission rate of the three most common GA models applies to all GA aircraft using avgas. Thus, the overall lead emissions from aviation into the SOCAB are 267 kg/year. These aircraft emit an additional 367 kg/year of lead above the mixing height. We estimate the uncertainty in this calculation from the data derived by the International Civil Aviation Organization that are used by the EDMS. The relative standard deviation of pollutant emissions ranged from a minimum of 3.7% for nitrogen oxides to a maximum of 17.5% for hydrocarbons. Because data on sulfur dioxide emissions were not listed, we assume the maximum relative standard deviation and thus apply 17.5% uncertainty to our calculation (28).

The final emissions from on-road gasoline, on-road diesel, and aviation in the SOCAB are given in Figure 3. We assume the following additional sources contribute insignificant quantities of lead, because their fuel use is so much smaller than that of automobiles and trucks: racing cars, off-road vehicles, construction equipment, marine vessels, and locomotives.
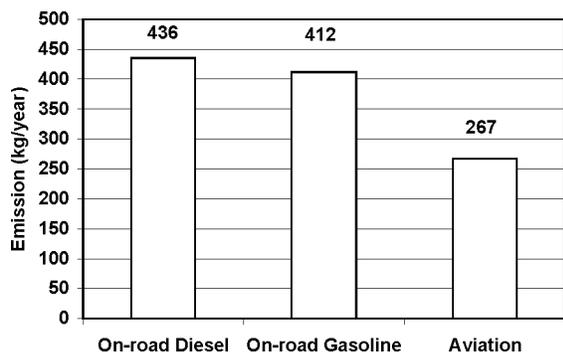
FIGURE 3. Mobile source emissions of lead in the SOCAB for 2001. Uncertainties are estimated to be 100% for on-road gasoline, 67% for on-road diesel, and 17.5% for aviation emissions.

Lead Sinks − Flow out of the Basin. As in Huntzicker et al. (*1*) and Lankey et al. (*2*), we consider the Basin to be a continuously stirred tank reactor, in which all air pollutants are fully mixed immediately upon release. In reality, mixing is not complete and the emissions are nonuniform, being greater in central Los Angeles than in suburbs east of the city. However, winds are mainly from the west, transporting emissions from central LA over the eastern suburbs. This compensates somewhat for the lower emissions east of the city and tends to produce relatively uniform airborne concentrations. To test this hypothesis, we graphed airborne lead concentrations at all available sites for 2001 against distance from the Pacific coast. No statistically significant correlation between concentration and distance was observed.

We calculate the flow rate of lead out of the Basin, q_{Pb} (kg/day), following Huntzicker et al. (*1*):

$$q_{Pb} = q_{CO}\overline{[Pb/CO]} \tag{1}$$

where q_{CO} is the flow of carbon monoxide out of the Basin, and $\overline{[Pb/CO]}$ is the spatially and temporally averaged ratio of the airborne concentrations of lead and carbon monoxide. These concentrations were recorded individually at sites throughout the Basin. Huntzicker et al. (*1*) used carbon monoxide as a tracer for lead because both species were emitted primarily from motor vehicles. They also assumed that the ratio of airborne concentrations $\overline{[Pb/CO]}$ was constant with elevation up to the mixing height. Although CO in 2001 was still emitted mainly from motor vehicles, Figures 2 and 3 show that most of the lead in that year came from other sources. The assumption of a constant $\overline{[Pb/CO]}$ ratio with height is also likely to be more tenuous in 2001 than earlier, due to different mixing rates from different sources. Because some of the measured lead is likely from resuspended soil, the expected larger particle sizes may confine a greater fraction of the lead closer to the ground and thus not all of the particles are available to be advected out of the Basin. Nevertheless, eq 1 can be used as an approximation if we scale the flow rate of CO out of the Basin to the volumetric flow rate of air. Carbon monoxide is a conserved species. Therefore, we assume the emission rate of CO is equal to the rate of advection of CO leaving the Basin.

The volumetric flow rate of air, *Q*, can be calculated by dividing the annual emissions of carbon monoxide (g/day) by the airborne concentration of carbon monoxide (g/m³). Using data from Huntzicker et al. for 1972 (*1*) and from Lankey et al. for 1989 (*2*), volumetric flow rates for the SOCAB are 2.2×10^{12} and $3.0 \times 10^{12} \pm 1.4 \times 10^{12}$ m³/day, respectively. We assume carbon monoxide emissions for 2001 are equal to the average of 2000 and 2002 emissions, or 4.52 million

kg/day (*29*). The average carbon monoxide concentration in 2001 was 0.00114 g/m³ (*30*). Thus, emissions and concentration data for 2001 suggest a flow rate of $4.0 \times 10^{12} \pm 1.0 \times 10^{12}$ m³/day, which agrees roughly with the 1972 and 1989 values.

Combined data from CARB (*30, 31*) and EPA (*32, 33*) give an average lead to carbon monoxide ratio $\overline{[Pb/CO]}$ of $2.31 \times 10^{-5} \pm 9.55 \times 10^{-6}$. Thus, we deduce a lead flow rate out of the Basin of $38\,100 \pm 15\,800$ kg/year.

Lead Sinks − Deposition. The deposition flux to a horizontal surface is given by:

$$F = V_d \times C_{air} \tag{2}$$

where V_d is the dry deposition velocity in m/s, C_{air} is the airborne concentration of lead in μg/m³, and *F* is the downward flux in μg/m²s. Using an average airborne lead concentration of 0.0310 μg/m³ (*31−33*) and an approximate deposition velocity of 0.0026 ± 0.0013 m/s over all particle sizes (*7−9*), we calculate a downward flux of $11\,300 \pm 5630$ kg/year. The mean V_d was calculated by Friedlander et al. (*8*) as the average for southern California over the submicrometer size range, meterological conditions, and surface types. Although we do not know the overall size range of lead particles in the SOCAB, this estimate of V_d is justified because data from Singh et al. (*34*) show that between 70% and 85% of all southern California lead particles by mass are submicrometer with current sources.

The results of these calculations indicate that the amount of airborne lead removed from the Basin in 2001 exceeds the amount of lead emitted into the Basin by more than an order of magnitude. The discrepancy suggests that there are significant sources of airborne lead beyond the aforementioned point and mobile sources. We hypothesize that resuspension of previously deposited lead is responsible for the disagreement.

Mass Balance Model

We begin with typical mass balance equations for both air and soil lead concentrations:

$$\frac{dC_{air}}{dt} = \frac{E}{V} + \frac{\Lambda C_{soil}A}{V} - \frac{V_d C_{air}A}{V} - \frac{QC_{air}}{V} \tag{3}$$

$$\frac{dC_{soil}}{dt} = V_d C_{air} - \Lambda C_{soil} \tag{4}$$

where C_{soil} is the surface soil concentration in kg/m², *E* is the rate of emissions from point and mobile sources in kg/s, *V* is the volume of the SOCAB in m³, Λ is the resuspension rate defined as the fraction of lead resuspended per second, *t* is time in seconds, and *A* is the surface area of the Basin taken here to be 4430 km² as shown in Figure 1. Although some urbanization has occurred surrounding these boundaries, particularly east of this area, we use the area shown in Figure 1 because it includes the significant point sources in the SCAQMD, CARB, and TRI emission databases and also includes nearly all of the heavily traveled roadways during the time of leaded gasoline. Consistent with the airflow calculations above, we assume that the Basin can be modeled as a continuously stirred tank reactor. This is compatible with known wind patterns, which are onshore during the day and offshore (easterly) at night over 24-h cycles. Because we are interested in lead in any chemical form, we ignore chemical reactions. Furthermore, we assume the flow of lead into the Basin by winds from upwind regions is negligible. We use a mean annual mixing height of 500 m for southern California as published by the EPA (*35*) and as implied in the calculations of Lankey et al. (*2*).

These coupled differential equations (eqs 3 and 4) have analytical solutions. We use the resulting decoupled equations to predict airborne and soil lead concentrations for 1970−2001. Historic emission estimates for vehicles are taken from gasoline consumption for the state of California, and we assume that the consumption in the SOCAB is proportional to the population there (*5*, *16*). The fraction of unleaded gasoline consumed in each year is taken from several sources (*36−40*), while the lead content of each type of fuel is taken from EPA (*6*). Aviation emissions are not included, because they are small relative to automobile and truck emissions for much of this time period, and historic data for aircraft are not readily available. Historic emissions from point sources are available for 1972 (*1*), 1976 (*41*), 1988−2001 (*17*), 1989 (*2*), and 1996−1997 (*42*). Point source emissions for years when data are not available are interpolated from existing data.

We define 1970 as $t = 0$ and use the initial airborne lead concentration as 3.6 μg/m^3 (*36*). The initial soil lead concentration is 79 $\pm$ 23 ppm, taken from Page and Ganje (*11*), Page et al. (*12*), and Miller et al. (*13*) as given in the Introduction. Using a soil density for southern California of about 1400 kg/m^3 (*43*) based on soil types in the SOCAB (*44*), the average initial soil lead concentration corresponds to 0.0054 $\pm$ 0.0016 kg of lead per square meter of surface soil. This assumes that the data of Page and Ganje, Page et al., and Miller et al. apply to the top 5 cm of soil, although the depths of their measurements range between 2.5 and 7.5 cm. Garland gives 5 cm as the mean depth of soil contamination originating at the surface (*45*, *46*), and Sehmel lists both 5 and 1 cm as appropriate depths to sample such contamination (*47*, *48*). Young et al. (*4*) state that resuspension generally affects soil within 10 cm of the surface. In the absence of a significant mechanical disturbance, resuspension below this depth is unusual.

A large source of uncertainty exists in estimating the resuspension term. We quantify this term through the resuspension rate Λ defined as:

$$\Lambda(s^{-1}) = \frac{R(\mu g/m^2 \, s)}{C_{soil}(\mu g/m^2)} \qquad (5)$$

where R is the resuspension flux upward from the surface. Λ is limited by its dependence on the depth to which C_{soil} is measured. However, it is useful for our purposes because it can provide a suitable prediction of the amount of material resuspended across regions with varying levels of soil contamination (*49*).

Long-term resuspension rates can vary by several orders of magnitude depending on conditions such as windspeed, soil moisture content, soil particle size, presence of saltating particles, magnitude of particle adhesion forces, meteorological conditions, and vegetation cover. In addition to wind, traffic-induced resuspension of road dust may be a significant contributor to the overall resuspension rate, but it is difficult to quantify. We focus on quantifying Λ based on wind only but with the assumption that road dust resuspension is incorporated into the overall value. Typical long-term values of Λ fall between 10^{-11} and 10^{-7} s^{-1} (*48*, *49*).

We expect the long-term resuspension rate to be at the lower end of this scale, given the relatively low average wind speed of 2.9 m/s in the SOCAB (*50*). Jensen (*51*) predicts resuspension rates of 1×10^{-10} to 5×10^{-10} s^{-1} for wind speeds around 3.0 m/s. Sehmel (*48*) meanwhile provides data on erosion in California that corresponds to Λ in the range of 6×10^{-11} to 3×10^{-10} s^{-1}. He further graphs resuspension rates found experimentally. With wind speeds on the order of 2.9 m/s, Λ is approximately in the $10^{-12}-10^{-10}$ s^{-1} range depending roughly on particle size. Results of Garland (*46*)
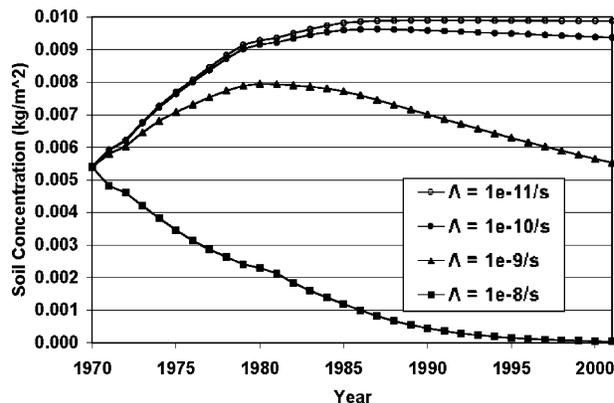
FIGURE 4. Modeled soil lead concentration with time. Points are graphed at midyear values.
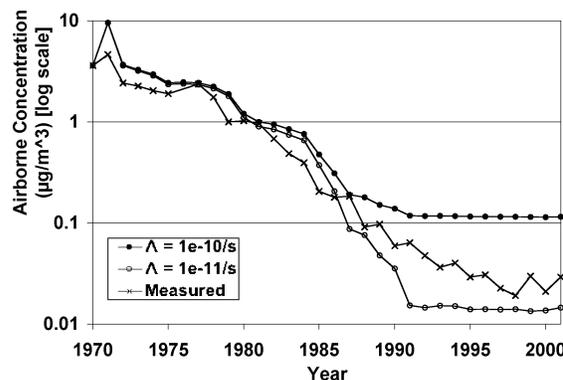
FIGURE 5. The modeled results of airborne lead concentration with time. Points are graphed at midyear values. Measured concentrations are also shown (*32*, *33*, *36*, *42*).

indicate that after long exposures, resuspension rates are about 10^{-10} s^{-1} even for wind speeds as great as 11 and 14 m/s.

Figure 4 shows the modeled change in soil lead concentration over time for four different values of Λ. Altering the deposition velocity by $\pm$50% changes the predicted concentrations for the three upper curves in 1980 by $<\pm$21% and in 2000 by $<\pm$28%. Figure 5 illustrates the modeled airborne concentration over time. The same alterations in the deposition velocity change the predicted concentrations in 1980 by $<\pm$18% and in 2000 by $<\pm$7%.

Attempts to run the model prior to 1970 suggest that either the limited available input data or the assumption of uniform emissions in the SOCAB is not accurate. Here, we begin our analysis with data from 1970. Despite the significant uncertainty in the emissions inventories after 1970, doubling the emission rate affects the model predictions by much less than a factor of 2.

Results and Discussion

Using resuspension rates of 10^{-10} and 10^{-11} s^{-1} in the model provides the best match to the measured airborne concentrations, as reported by EPA and CARB (*32*, *33*, *37*, *43*). The concentrations are only approximate as the data were obtained with high-volume samplers; measured concentrations are somewhat dependent on the wind speed and the orientation of the equipment with respect to wind direction (*52*).

Soil lead concentrations over time and throughout the geographic area of the Basin are shown in Figure 6, based on the individual data points of Page and Ganje (*11*), Page et al. (*12*), Miller et al. (*13*), and Young et al. (*4*). The outlier value of 250 ppm from Miller et al. (*13*) applies to a site near
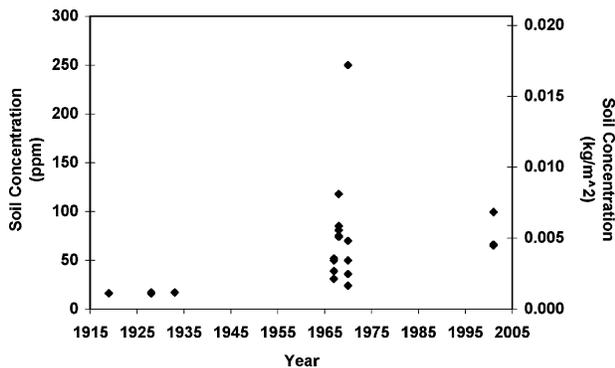
FIGURE 6. Measured soil lead concentrations in southern California over time and corresponding surface soil concentrations (soil depth = 5 cm) (4, 11−13). Three data points are available for 2001, although one (~66 ppm) is a double point.

TABLE 2. Sources and Sinks of Airborne Lead in the South Coast Air Basin in 2001

input		output	
source	kg/year	sink	kg/year
stationary	5410 ± 2700	air flow	38 100 ± 15 800
mobile	1120 ± 506	deposition	11 300 ± 5630
resuspension	~54 000		
total	~61 000	total	49 400 ± 16 800

a "traveled street". Only those data points that represent urban background levels in these studies are shown. The three values of Young et al. (*4*) from 2001 are within experimental error of the concentrations measured in 1967−1970, showing insignificant loss of soil lead over 30 years despite the decrease in lead emissions in the SOCAB. This is consistent with model results when resuspension rates of 10^{-9}, 10^{-10}, and $10^{-11} s^{-1}$ are used as inputs: the modeled 2001 soil concentrations are 0.0055, 0.0094, and 0.0099 kg/m^2, respectively.

With $\Lambda = 10^{-8} s^{-1}$, we find the model results in disagreement with observations. Not only are predicted 2001 soil concentrations much lower than measured, they also drop immediately from the initial value of 0.0054 kg/m^2 (79 ppm), indicating that even the initial measured concentration is inconsistent with such high resuspension rates. We would not expect a decrease in soil lead until after the phase-out of lead in gasoline had begun in 1975. The 1967−1970 soil lead concentrations of 79 ± 23 ppm (*11–13*) as compared to the 1919−1933 measurements of 16 ± 0.5 ppm (*11*) in the SOCAB further suggest an upward slope in soil concentrations over time (Figure 6).

Values of C_{air} for $\Lambda = 10^{-9} s^{-1}$, not shown in Figure 5, level off at ~1 μg/m^3, far in excess of observed airborne concentrations, suggesting that smaller resuspension rates are likely. In light of this observation as well as reasonable agreement between modeled and measured airborne concentrations, we propose a long-term resuspension rate for the SOCAB in the range of $10^{-10}-10^{-11} s^{-1}$. These rates are consistent with predictions and measurements elsewhere (*46, 48, 51*).

Using a resuspension rate of $5 \times 10^{-11} s^{-1}$, which shows good agreement with measured C_{air}, resuspension contributes an additional 54 000 kg/year of airborne lead in 2001 as shown in Table 2. Despite the large uncertainty in the value for Λ, the amount of lead remaining in the SOCAB soil decreases only very slowly with time, virtually independent of the resuspension rate when Λ lies between 10^{-10} and $10^{-11} s^{-1}$.

Both modeled and measured results indicate that lead has a very long residence time in surface soil. Assuming that emissions remain at 2001 levels, the model predicts that it would take more than a century for soil concentrations to approach steady-state; the steady-state soil concentration with 2001 emission levels is 29 ppm. Current evidence thus suggests that soil contamination contributes most of the total airborne lead currently measured in the SOCAB and is likely to continue to do so for many years.

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