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Very Fine and Ultrafine Metals and Ischemic Heart Disease in the California Central Valley 2: 1974–1991

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The southern part of Central Valley of California in winter has long had high PM₁₀ mass, which until about 1990 included sulfate, vanadium, and nickel from the burning of crude oil used to generate steam to enhance heavy petroleum recovery. In roughly 1990, natural gas became the major energy source used for steam generation. In 1989-1991, data were collected throughout California on the mortality from strokes and ischemic heart disease (IHD). Although no spatial variability was seen for strokes, the southern San Joaquin Valley was found to have IHD mortality rates roughly 60% greater than the rest of the valley. PM₁₀ was statistically identified as the major factor associated with the IHD mortality. However, when the rate of IHD was reexamined in the 2003–2007 period, a sharp reduction, about 30%, was seen in the relative rates for southern San Joaquin Valley as compared with the northern Sacramento Valley. We have measured very fine and ultrafine vanadium and nickel aerosols in a winter experiment in 2009, which shows an order of magnitude reduction in vanadium and nickel aerosols as compared with the pre-1990 data, which is a consequence of the switch from burning crude oil to natural gas to generate the steam. The inference of a causal relationship between the reduced vanadium and nickel and the improved IHD rate is supported by a growing body of laboratory and epidemiological work on the toxicity of vanadium and nickel, including from oceangoing ships burning crude or residual oil.

INTRODUCTION

The Central Valley of California is well known for exhibiting in much of its area serious violations of federal PM₁₀, PM_{2.5}, in winter, and ozone standards, in summer, matching or exceeding those seen for any commensurate area of the United States. Yet other parts of the valley share agriculturally based land uses and meteorology and do not exhibit serious violations of these standards. Although efforts continue to control particulate matter (PM) and ozone violations, this pollutant gradient offers an ideal situation to examine the effects of pollutants on health. Many of the confounding variables seen in comparison studies, such as meteorology, land use patterns, and cigarette smoking, are very similar throughout the Valley, and a wealth of data exists on the pollutants and their sources from district air monitoring and Air Resources Board (ARB) studies. The presence of detailed analysis of mortality from ischemic heart disease (IHD) and stroke during 1989-1991 reported by the Cardiovascular Disease Outreach and Epidemiology (CORE) Program (Kao et al. 1994) and the California Department of Health Services (CADHS 2010) provides a natural opportunity to compare the pollutant data and mortality data for the identical time ranges. While this article focuses on PM effects, the original report (Cahill et al. 1998) also examined the effects of ozone and carbon monoxide (CO) on IHD and stroke mortality: some of which are presented in this article for completeness. However, complexities in the spatial and temporal gradients of ozone require further interpretation, and thus, they are not examined in any detail in this article.

BACKGROUND

The northwest-southeast trending 550 km by 100 km Central Valley of California is one of the richest agricultural areas in the world (Figure 1). In addition, there are two major metropolitan areas in the Valley, the Sacramento-Fresno regions each with about 1 million residents, and otherwise towns are generally medium (circa a few 100,000) to small and spread

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100

80

60 та/ш₃

40

20

0

FIG. 1. Map of the Central Valley of California. The northern part is referred to as the Sacramento Valley while the southern part is called the San Joaquin Valley. The areas within the dotted circumference vary in altitude from almost sea level in the Delta to a few 100 m in elevation. The small arrows denote the typical winter wind patterns (Hayes et al. 1984).

throughout the Valley. The mean population density is about 60 people/km².

High summer temperatures in the Valley (mean daily high, July, $36^{\circ}C \pm 1^{\circ}C$) lower the surface pressure and draw massive inputs of cool air into the Valley from the San Francisco Bay area, bringing with it oceanic aerosols and typical urban pollutants enhanced by the major petrochemical industrial sites at the Carquinez Strait. These latter sources lie directly in the path for the cold, dense winds from the San Francisco Bay. These winds funnel down the entire San Joaquin Valley, south of the strait, but rarely penetrate north very far in the Sacramento Valley, north of the strait. Rainfall in the Valley is rare during the period from May through October.

In winter, there is slow drainage of cold air off the Sierra Nevada to the Valley floor (mean daily low, January, $3^{\circ}C \pm 1^{\circ}C$) and then, at the rate of about 50 km/day, the air drains toward the lowest point, the Sacramento–San Joaquin Delta, directly east of the Carquinez Strait. Thus, little impact from the San Francisco Bay area is seen on most winter days in the Central Valley beyond the immediate Delta region. Winter rainstorms and strong northwest winds periodically clean out the Valley. Thus, the Valley tends to fall into two distinct and very different conditions: hot, dry summers with strong diurnal winds, and cool, foggy winters with long periods of stagnation. This results in two very different aerosol types: summer and winter (Figure 2).

In Figure 2, 24 h, one day in six PM_{10} mass at the Fresno 1st Street "Super-site" is shown (ADAM 2010). The site is in a largely residential neighborhood roughly 1 km east of the



FIG. 2. PM₁₀ mass at the Fresno "Super-site", 2002

89

Month of the Year 2002

10 11

12

The health effects data are derived from CORE (Kao et al. 1996) and CADHS (2010). CORE collected city (>20,000 in-habitant) and countywide data on mortality from stroke and IHD; the latter is the largest source of mortality in the California Central Valley accounting for about one fourth of all deaths. The CORE data were corrected for age, >35 years, and reduced to incidence per 100,000 residents. The results were then presented as a ratio to the California average values the both males and females. Individual city data were weighted and averaged to obtain countywide averages in order to match the air pollution data.

The effect of race was examined over the entire California dataset. The data were available only in three classifications: "White," "Black," and "Other," with "Other" in the Central Valley being largely Latinos. No statistically significant difference was seen in the IHD rates for the dominant >75 years age group, but a slight enhancement of the rates for Blacks was seen in the 64- to 75-year cohort. For all ages, the mortality/100,000, 95% confidence limit, was 196 \pm 3 for white males, 212 \pm 9 for black males, and 199 \pm 4 for other males; the corresponding values for females were 127 ± 3 , 166 ± 5 , and 135 ± 3 . The stroke deaths, on the contrary, while only roughly one third of the rate of IHD mortality, showed stroke rates roughly twice as high for the "Black" and "Other" cohorts than the "White" cohort. Consideration was given to cigarette-smoking patterns, but no significant difference was seen along the length of the Valley (Table 1).

Four metrics for air pollution were studied, each with very different cardiovascular impacts: ozone (top 30 h and hours above 0.09 ppm), annual average CO, and annual average PM_{10} . The approximate cigarette-smoking patterns were calculated



County (north to south)	IHD rate vs. CA average (%)	Stroke rate vs. CA average (%)	Ozone top 30 h (ppm)	Ozone hours >0.09 (h)	CO annual average (ppm)	PM_{10} annual average $(\mu g/m^3)$	Smoking rate average (%)
Shasta	-14	+9	0.095	20	0.54	30	22.4
Butte	-27	-3	0.078	6	0.93	38	22.4
Sutter	-4	+26	0.090	4		44	22.8
Placer	-17	-4		125		30	22.8
Sacramento	-6	+2	0.107	153	1.29	44	23.4
Yolo	+2	+13		16		30	22.4
San Joaquin	+15	+17	0.096	36	1.13	51	22.8
Stanislaus	-7	-3	0.102	119	0.75	48	22.0
Madera	+11	-23		30		53	22.0
Fresno	+3	-12	0.129	369	0.87	77	22.0
Kings	+17	+20	0.106	21		61	22.5
Tulare	+22	+8	0.093	192	0.88	67	22.5
Kern	+33	-11	0.111	605	0.80	68	22.5

 TABLE 1

 Ischemic heart disease (IHD) and stroke mortality versus air pollutants in the Central Valley of California, 1989–1991

from cigarette sales data. Correlations were calculated for all parameters and are presented in Table 2 and Figures 3 and 4. At some sites, measurements were lacking. No data were deleted.

To further investigate the statistical robustness of these data, we used "proc glm" on SAS version 9.1 for the analyses. Because there are only 13 observations in total, we did not feel that imputation would prove fruitful. Therefore, we ran multiple regression models with "percentage change of IHD" and "percentage change of stroke" as the outcome variables. The possible explanatory variables were "number of hours of ozone above 0.09 ppm" ("ozone"), "annual average PM less than 10 μ m, measured in micrograms per cubic meter" ("PM₁₀"), and "percentage rate of smoking" ("smoking"). We checked the assumption of normality of the errors by formally using the Shapiro–Wilk test and the assumption of homoskedasticity informally using residual plots.

The overall stroke model was not significant (p = 0.1108). We did not proceed further for that outcome. However, the overall IHD model was significant (p = 0.0330). We used backward elimination as the model selection strategy, for example, that of Neter et al. (1996). Ozone and smoking were eliminated—in that order—leaving PM₁₀ as the sole significant explanatory variable (p = 0.0026). The resulting linear regression is given

by

estimated percentage change of IHD = $-41.5 + 0.87 \times PM_{10}$,

and the corresponding *r*-squared value is 0.576. It is noteworthy that Fresno appears to be an outlier. If this point is removed, the slope and *r*-squared values jump dramatically. We have no cause to remove that point and we therefore use the model, as indicated earlier, as our finding. However, the Fresno value includes semirural sampling sites at the urban fringe that are not present in other San Joaquin Valley cities, which had at that time only a single city center site.

However, the strong association between aerosol mass and IHD posed a problem. As additional data were developed in the Central Valley from the IMPROVE program (Malm et al. 1994) and from extensive California studies (CRPAQS 2001, Cahill et al. 2003), it became clear that the major aerosol species that dominated fine mass was ammonium nitrate, with much smaller contributions from organic matter, including a wood smoke component, and ammonium sulfate. These are however water soluble and have not been closely linked to IHD in animal and laboratory studies. (Devlin 2003; Lippmann 2009).

 TABLE 2

 Correlation of air pollution to mortality by IHD and stroke in the Central valley of California, 1989–1991

Mortality	Ozone average top 30 h (ppm)	Ozone number hours > 0.09 (ppm)	CO (ppm)	PM ₁₀ (µg/m ³)
Correlation to IHD	$r^2 = 0.18$	$r^2 = 0.27$	$r^2 < 0.05$	$r^2 = 0.56$
Correlation to stroke	$r^2 = 0.21$	$r^2 = 0.19$	$r^2 < 0.05$	$r^2 < 0.05$

IHD IND Annual average PM₁₀ mass Death rate versus state average 200 150 Average PM₁₀ 100 50 0 Nerced, Natera Sacramento Tulare 5an Joaqui tern Sutte Fresho Wings Placet Stanislat Butt Shas

FIG. 3. Comparison of death rate from IHD, compared to the California average, and annual PM_{10} inhalable aerosol mass. Shasta County is at the very northern end of the California Central Valley, and Kern County is at the southern end.



FIG. 4. IHD mortality rate relative to the California average versus PM_{10} . The parameters are associated by a regression $r^2 = 0.56$.



FIG. 5. $PM_{2.5}$ mass at the Fresno "Super-site," 2002, the same period used in Figure 2.



FIG. 6. IHD in the California Central valley, 1989–1991 versus 2003–2007. The 1989–1991 data are scaled to match the Shasta–Butte data, 2003–2007, as diagnostic protocols differed.

Reduction of IHD Rates: 2003-2007 Versus 1989-1991

Beginning around 1999, data on aerosol in the Central Valley were enhanced by the availability of PM_{2.5} mass and chemical speciation at some San Joaquin Valley sites. In Figure 5, the same period for the Figure 2, one day in six PM₁₀ mass is shown for 24 h daily fine PM_{2.5} mass (ADAM 2010). The old federal 24-h PM_{2.5} standard was 65 μ g/m³ and the current 24-h standard is 35 μ g/m³, which is routinely violated in winter. The annual average standard is 15 μ g/m³.

The question of IHD and aerosols was revisited for the period 2003–2007 (Cahill et al. 2010). From the recent data, it became



FIG. 7. Vanadium and nickel aerosols in Bakersfield, January 1976.



FIG. 8. (a) and (b) showing $PM_{2.5}$ mass closure and major species.

clear that a comparison of the IHD mortality, 1989–1991 versus 2003–2007, shows a major reduction, roughly 30%, that occurs in 2003–2007 for the counties north of Bakersfield (Figure 6). The trend for the northern half of the Central Valley is similar, while Bakersfield in Kern County is only slightly reduced. Thus, the question arises as to the causes for the improvement in the IHD rate.

The compositional analysis of Central Valley aerosols dates back to the early 1970s, including an innovative program of UC Davis for the California Air Resources Board (CARB), 1973–1977 (Cahill et al. 1974; Flocchini et al. 1976; Barone et al. 1978; Motallebi et al. 1990a, 1990b). A total of 14,100 daily aerosol measurements were taken in three size modes (15–3.5 μ m, 3.5–0.50 μ m, and 0.50–0.0 μ m) and analyzed for elemental content by particle-induced x-ray emission (PIXE) (Cahill 1995) at about fourteen separate sites, including the valley sites of Chico, Sacramento, Visalia, (100 km NNE of Bakersfield), and Bakersfield. Bakersfield had one of the highest levels of vanadium and nickel aerosols in California, but port areas (Oakland, Richmond, and Los Alamitos) were also high (Figure 7).

Neither vanadium nor nickel was seen in sizes above 3.5 μ m diameter. The radically different vanadium/nickel ratios occurred when winds came from the western (Taft, 40 km SW) or northern (Oildale, 5 km N) oil fields periods of stagnation, giving the highest pollution levels, favored sources in the much closer northern oil fields, some of which were within Bakersfield itself (Motallebi et al. 1990b). In addition, there was an oil refinery in Oildale, with potentially different emissions than the crude oil burned for enhanced oil recovery (EOR). Also it is noteworthy that while the vanadium and nickel concentrations can vary by over an order of magnitude, the lead levels were relatively



FIG. 9. Lead and sulfur, 1974–1976, versus sulfur, 2009, and vanadium, nickel, and zinc 1974–1976. Visalia lies between Bakersfield and Fresno.

constant, indicating a continuous impact of traffic-derived aerosols at Bakersfield.

Recent compositional analysis of the $PM_{2.5}$ aerosols in the southern San Joaquin Valley also became available through the IMPROVE site at Sequoia National Park (NP) starting in 1992 (Malm et al. 1994). The Sequoia NP Ash Mountain site was at low elevation, near 600 m, and thus received aerosols from the valley floor whenever the inversion lifted to that level.

IMPROVE had anticipated the value of PM_{2.5} mass and full speciation, using quality assurance protocols of "integral redundancy" to measure major parameters by two or more independent methods. Sulfur from PIXE on Teflon in Channel A (Cahill 1995) is compared with sulfates after a denuder on Nylon filters in Channel B. Organic matter measured by combustion from tandem quartz filters in Channel C is compared to organic matter estimates by nonvolatile hydrogen of Teflon in Channel A (Cahill et al. 1989). Iron was measured by both PIXE and x-ray fluorescence (XRF) on the Teflon filters of Channel A. Thus, the gravimetric mass can be directly compared to the sum of all species (Figure 8b) and all major components (Figure 8a).

Figure 8 shows periodic incursions of high aerosol levels throughout the winter whenever the valley inversion rises to the altitude of Ash Mountain. Other than during these incursions, aerosol mass levels are very low, a few μ g/m³. By choosing an arbitrary concentration of 10 μ g/m³, we can be assured that we are seeing San Joaquin Valley aerosols diluted by the high inversion levels necessary for the Ash Mountain incursions.

Finally, the excellent agreement between the mass of aerosol measured gravimetrically and the sum of species for all three IMPROVE channels (Figure 8a) shows that we are not missing any major component of the $PM_{2.5}$ mass.

Figure 5 showed that the violations of the annual and 24-h PM_{2.5} standards in the San Joaquin Valley are driven by the winter aerosols. The high PM_{2.5} values generally correspond to stable conditions with low inversions, and the clean periods to synoptic rainstorms passing through the Valley. Figure 8b shows that winter composition is dominated by fine $D_p < 2.5 \ \mu m$ nitrates with smaller contributions from organic matter, wood smoke and vehicular exhaust, and some sulfates (IMPROVE 2010). The IMPROVE data also include fine metals, thus aiding our intercomparisons.

Winter data from the earlier efforts (Cahill et al. 1974, 2003; Flocchini et al. 1976) can be roughly compared with the current work (Cahill et al. 2010) by summing the four finest DRUM stages to obtain $< 0.56 \ \mu$ m particles (Table 3 and Figure 6).

Addressing the potential health impacts of these data, vanadium and nickel have the highest mortality risk coefficients of seventeen fine particulate matter (FPM) components (Lippmann et al. 2006). The elevated levels seen in 1974–1976 were sharply reduced as early as 1992–1993, using the vanadium and nickel levels in the southern San Joaquin Valley from the Sequoia IM-PROVE site, although the Sequoia NP site's PM_{2.5} data are not directly comparable to the valley floor's very fine (<0.5 μ m) particle data. These data are shown in Figure 9. For comparison's sake, vanadium and nickel have current annual averages in FPM of 1.9 ng/m³ across sixty metropolitan areas (MSAs) (Lippmann et al. 2006).

Examining the potential sources for the very fine (in this case $<0.5 \ \mu\text{m}$) particles (Table 3 and Figure 6), we see sulfur plus metals associated with crude oil combustion (vanadium and nickel), as well as lead and other vehicular metals. Kern County has very large reserves of crude oil, most of which lie just north and west of Bakersfield, although there was and is some oil extraction within the city itself. The Kern County oil fields have a heavy crude oil that requires EOR techniques such as steam injection to extract. Initially, the steam was generated by burning the crude oil itself, resulting in high pollution levels. In 1979, violations of federal air quality standards required Kern County to apply strict controls on sulfur dioxide, nitrogen dioxide, and particulate emissions, which included metals vanadium and nickel. EOR by crude oil combustion dropped slowly, 1980–1990, and then dramatically with the increasing availability of natural gas, with an immediate improvement in air quality by the early 1990s (CA Almanac). Thus, the population subject to IHD mortality in Bakersfield 1989-1991 had at least two decades of exposure to elevated high levels of very fine to ultrafine transition metals, specifically vanadium and nickel at 10-20 times the current US averages.

Laboratory data on the impact of vanadium and nickel aerosols has become more extensive (Zhang et al. 2009). Thus, we can hypothesize that the effective control of the

TABLE 3

Comparison between January 1974–1976, averaged, and January 2009. The four finest DRUM stages of the 2009 data have been
summed to be PM < 0.56 μ m to better match the PM < 0.5 μ m data of the earlier ARB/UCD aerosol network. The Sequoia NP
and Fresno winter 2002–2003 data are of $PM_{2.5}$

	Sulfur	Vanadium	Nickel	Zinc	Lead
Valley transect 2009 [January 2009 (ng/m ³) $<$ 0.56 μ m]					
Redding	88.8	0.2	1.0	13.9	2.5
Chico	136.9	0.5	0.4	5.2	2.1
Sacramento	246.0	0.4	0.1	6.9	1.6
Fresno	261.4	0.3	0.5	40.3	4.9
Bakersfield	505.0	0.2	2.3	32.0	9.4
Fresno $PM_{2.5}$ (ng/m ³)					
Winter 2002	266.2	0.47	0.22	12.7	1.2
Sequoia NP (~600 m), 10 episodes $PM_{2.5} > 10 \ \mu g/m^3$					
Winter 2002–2003 (ng/m ³)	194	0.2	0.1	4.7	1.0
Winter 1992–1993 (ng/m ³)	562	0.7	0.1	6.0	2.0
Valley transect 1974–1976 [January 1974–1976 (ng/m ³) average	ge, $<0.5 \ \mu m$ di	ameter]			
Redding	na	na	na	na	na
Chico	170	7	1	10	483
Sacramento	375	0	1	18	720
Visalia	225	17	8	19	593
Bakersfield	1685	19	38	61	1714
Port of Los Angeles					
August-September, 2008	2565	16	4	13	6

crude-oil-derived metals in the late 1980s may be responsible for the decreased IHD death rate at sites north of Bakersfield in the 1990s and beyond. This hypothesis is supported by other studies, such as the sharp drop in mortality after a sharp drop in sulfur, vanadium, and nickel in Hong Kong when the sulfur levels in fuels were lowered by edict (Hedley et al. 2002; Lippmann et al. 2006). The presence of high levels of nickel in New York City from residual oil combustion in buildings is hypothesized to be a causal factor in the enhanced mortality



FIG. 10. Very fine $(0.26 > D_p > 0.09 \,\mu\text{m})$ vanadium and nickel in Wilmington, CA, downwind during daylight hours at the Port of Los Angeles.

associated with $PM_{2.5}$ (Peltier et al. 2009; Peltier and Lippmann 2010).

The current data also have relevance to the emission of sulfur, vanadium, and nickel by oceangoing ships. Recent data in New York and New Jersey (Peltier et al. 2009), and the Port of Los Angeles (Cahill et al. 2009), show the impacts of oceangoing ships burning residual or bunker oil. In the Los Angeles study, the average concentrations of fine sulfur, vanadium, and nickel in the coastal town of Wilmington in August 2008 were 2565, 16, and 4.4 ng/m³, respectively (Table 3). Southern winds bring aerosols from the Port of Los Angeles into Wilmington routinely during part of each day (Figure 10). Ultrafine particles were not collected in this study, so the actual values could have been significantly higher than these values. Thus, on the basis of the Bakersfield data, the enhanced rates of IHD are predicted in the area.

CONCLUSION

IHD mortality in the Central Valley of California, corrected for age, race, sex, and smoking, was almost 60% greater at the more polluted southern end of the Central Valley than the less polluted northern end in 1989–1991 despite similar meteorology and land use patterns throughout the Valley. A significant association, $r^2 = 0.56$, is seen between IHD mortality and PM₁₀, but negligible correlation, $r^2 < 0.05$, is seen with stroke mortality. When PM₁₀ data are converted to an estimated PM_{2.5} mass by using more recent measurements, these results are similar to other studies such as the "6 Cities" study (Dockery et al. 1993) in mortality increase per particulate mass; 1.9% mortality increase per μ g/m³ of PM_{2.5} for "6 Cities," while 1.8% mortality increase per μ g/m³ of estimated PM_{2.5} for this study, despite major differences in meteorology and pollutant mix. No association was seen between CO and any health impact. Ozone was only weakly correlated with IHD and stroke. However, strong east-west ozone gradients were not corrected for in this work, and thus, these results for ozone must be viewed as merely indicative of the possible impacts.

The sharp reduction seen, when the IHD data were reexamined in the period 2003–2007, was coincident with a reduction in the high levels of vanadium (19 ng/m³) and nickel (38 ng/m³) due to oil field operations from before 1974 to after 1990, when new protocols were adopted. The vanadium and nickel levels in 2009 were close to the national average values, both roughly 1.9 ng/m³. The relatively smaller reduction in IHD rate in Bakersfield itself is associated with the continuing impacts of vehicular very fine and ultrafine metals (Cahill et al. 2010, this issue).

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