

U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
PUBLIC HEALTH SERVICE
CENTER FOR DISEASE CONTROL
ATLANTA, GEORGIA 30333

OFFICIAL BUSINESS

POSTAGE AND FEES PAID
U.S. DEPARTMENT OF HEW
HEW 396



THIRD CLASS
BLK. RT.

19898SNEE RD792525
MR RONALD D SNEE
DUPONT DE NEMOURS & CO
ENGINEERING SVC DIV
1007 MARKET ST
WILMINGTON, DE 19898

TEH 0532412

N33804

Lead-sealed cans account for half of food pollution

By Robert Locke

Associated Press

PASADENA, Calif. — Lead pollution pervading the atmosphere and the world's food chain is much worse than previously suspected, with lead-sealed food cans — such as tuna fish cans — accounting for half the lead in the American diet, researchers at the California Institute of Technology report.

The contamination, underestimated because it is so widespread, "masquerades as natural lead in foods before they are processed," two Caltech scientists said in the *Journal of Science* this week.

"Typical adult Americans have 500 times more lead in their bodies than their prehistoric ancestors," said Dorothy Settle, a senior scientist at Caltech, in an interview yesterday.

She and geochemist Clair C. Patterson have studied lead pollution for more than 10 years.

Their findings are based in part on the discovery that tuna in cans

sealed with lead solder contains 10,000 times more lead than the estimated level from pre-industrial times.

Tuna from lead-soldered cans was 1,000 times more contaminated than fish fresh from polluted oceans, they found, compared with only a 20-fold increase in non-soldered cans.

"Lead-soldered cans (for any foods) should be eliminated immediately because they constitute a known, readily identified principal source of lead in foods," Ms. Settle said.

Richard Atchison, president of Van Camp Seafood Co. which makes Chicken of the Sea tuna, said his company had "anticipated this problem about four years ago" and installed equipment to make and pack a one-piece molded steel can with a clipped on lid.

The research was "not really a surprise," according to Carolyn Campion, of Castle & Cooke in San Francisco, the parent company of Bumblebee Seafood. She said the company is looking into non-soldered cans.

She predicted there would be no substantial comments from the industry until a meeting Tuesday at which the Tuna Research Foundation Technical Committee will discuss the issue.

Lead pollution — from paint, auto fumes, industrial processes, canned goods — is known to cause symptoms ranging from headaches, weakness and constipation to abdominal cramps, convulsions and brain damage.

"For the present generation of adults I think little can be done," Ms. Settle said, citing inadequate controls on lead in the air and food. "It is for future generations that positive action should be taken."

Because few laboratory processes can distinguish between natural lead and industrial pollutants, both have been lumped together and falsely interpreted as the natural level, the researchers said.

They conducted unusually elaborate tests to eliminate as much industrial lead from their samples as possible, and worked in a laboratory designed to be virtually lead-free.

WILMINGTON JOURNAL
3/14/80

Long-Term Trends in Total Suspended Particulates, Vanadium, Manganese, and Lead at near Street Level and Elevated Sites in New York City

Paul J. Lioy, R. Peter Mallon and Theo. J. Kneip

Institute of Environmental Medicine
New York University Medical Center

Long-term concentration measurements of TSP, V, Pb, and Mn have been made in New York City. Three month moving averages have been constructed from integrated weekly data collected at the NYU Medical Center rooftop site from 1968-78. Rooftop vs. 1st floor concentration comparisons have been made at two locations for the periods 1968-70 and 1976-78. TSP, V, Pb and Mn have decreased in ambient concentration over the entire study. This reflects the controls incorporated on sources over the 10 yr period. The rooftop vs. 1st floor analyses have shown the influence of source distribution on concentration patterns. V levels showed no difference between the rooftop and 1st floor concentrations, which demonstrates the elevated nature of the sources and uniform mixing of material in the atmosphere. Pb, Mn, TSP, showed higher values at the ground, which would be indicative of ground level sources. During 1978, Mn and Pb increased throughout all (Pb) or part (Mn) of the year. The concentrations of these materials do not approach values which have been related to known health effects, although the present Pb values have exceeded the NAAQS.

Atmospheric aerosols can have significant effects on the environment. At current concentrations, the principal effects are associated with the degradation of visibility, local weather modification, soiling and material erosion. More importantly, however, airborne particulates have long been known to be associated with deleterious health effects.¹ Studies conducted at this laboratory^{2,3} have focused on the characterization of the Total Suspended Particulates (TSP) present in the New York City ambient atmosphere. The principal sources of the TSP are those common to most cities, and include power generation, space heating, incineration, industry and the automobile, plus natural and transported particles.

Studies have shown the effect on reported airborne lead concentrations of sampler location in relation to a roadway.^{4,5} In New York City, one study has shown that there is a difference in the time of the street level diurnal lead peaks for crosstown versus uptown-downtown streets at a given intersection.⁶ However, no data have appeared on the average long-term differences between street or low level and elevated

sampling sites in a large city. Since the automotive transport system contributes lead (Pb) and other metals contained in fuel additives to the TSP, a study was undertaken of the long-term differences observed between first floor and rooftop (12-14th floor) samplers.

Since sampling has been performed during two different periods in two locations in the city, the long-term trends in airborne particulate mass and composition were also considered in developing an understanding of the observed results. Concentrations of TSP and the elements vanadium, lead, and manganese have been evaluated for this study.

Methodology

Weekly composite air samples were collected continuously at a rooftop site from 1968-70 and 1972-78, and from 1976-78 at a first floor level site located at the New York University Medical Center. The rooftop site is situated above the 14th floor of the midtown NYU School of Medicine residence hall about 70 meters from the East River Drive and the lower sampler is about 25 m from and 4 m lower than the East River Drive. Each sample was collected on a Type A Gelman glass fiber filter using a moderate flow (0.57 m³/min) sampling system. The filters were weighed for TSP and portions were digested in HNO₃ and HClO₄. The samples were then analyzed for the trace elements using atomic absorption spectrophotometry. A thorough description of the sampling system and analytical procedures is found elsewhere.⁷

Results

Long-Term Trends at Rooftop Sites

Since 1969, there has been a steady decrease in the TSP detected at the NYU Medical Center Rooftop site* with the most significant reductions occurring between 1969-70 and 1972. This trend is a continuation of and consistent with the decreases recorded by the National Air Surveillance Network from the late 1950's through the 1960's. The 1978 TSP concentrations were approximately 40% of the 1969 values, and 25% of the values in the late 1950's.

* Figures depicting: 1) the long term trends in TSP, Pb, Mn, and V and 2) the rooftop ground TSP and V at the HASL and NYU sites are available (upon request) from the authors.

Copyright 1980 Air Pollution Control Association

Similarly, the concentrations of Pb, V, and Mn have declined as changes have occurred in fuel types and fuel quality. For instance, the reduction of the sulfur content of oil from >3% prior to 1967 to <0.3% since 1972 has accounted for much lower contributions of TSP and V concentrations due to oil burning.⁸ Several changes have occurred in regulations of Pb in gasoline and are generally reflected in the decline of the Pb concentration. The elimination of coal burning since 1967 has reduced Mn levels by 4 to 5 fold.

The overall reductions, however, may be deceptive as the atmospheric dispersion in New York City is highly variable.^{9,10} Both the morning mixing height and wind speed are highest during the winter period. An index of the potential average weekly volume into which pollutants can be dispersed has

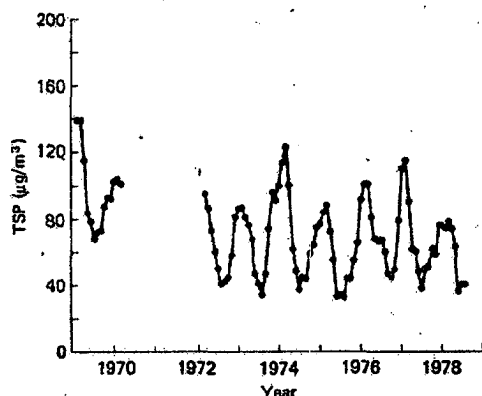


Figure 1. Dispersion normalized NYU Rooftop TSP concentrations for the period 1969 through 1978 as 3 month moving averages.

been computed as the product of morning mixing height and wind speed through the mixing height. This index is called the dispersion factor.⁹ It was shown that the dispersion pattern is cyclic with the winter having 2 to 3 times the ventilation capacity of summer.

To observe trends that might otherwise be masked by the meteorological fluctuations, the TSP data have been normalized to a 3-hr average dispersion factor.^{9,10} The normalized data are shown in Figure 1. The effect of increased winter emissions is evident in the figure with the normalized winter TSP now exceeding the normalized summer concentrations. The normalized TSP data show two winter peaks which ex-

ceed the others (Figure 1). These two peaks are associated with the periods of fuel variances during the extraordinary winters of 1973-74 and 1976-77 and show that the normalized concentrations for these two periods were similar to the normalized concentrations of the late 1960's.

While there was a general decrease in the ambient TSP concentrations the normalized TSP values had a less pronounced decline (excluding the winters 1973-1974 and 1976-1977). This suggests that a return to lower average dispersion conditions, combined with increased emissions, could result in higher actual ambient aerosol concentrations than those observed in recent years.

Rooftop Versus First Floor Level Site Concentrations

Since 1976, particulate samples have been taken at the first floor NYU Medical Center station simultaneously with the rooftop samples. This station is situated near the southbound lanes of the East River Drive; hence close proximity to a major source of automotive emissions. Data were also available for the period 1969 through 1970 from the DOE's Environmental Measurement Laboratory (EML, formerly the AEC Health and Safety Laboratory, HASL). These rooftop (13th floor) and first floor sites are located in lower Manhattan at Hudson and Varick Streets, and provide historical perspective for the rooftop versus first floor level concentration comparisons at NYU.

Statistical analyses were conducted to compare the data from the rooftop and first floor sampling sites at both the EML and NYU locations. The results of paired-t tests listed in Table I include the mean concentration $\pm 1 \sigma$, the level of significance, and the least square regression correlation coefficients for the roof and first floor.

The TSP concentrations are approximately 17% higher at the first floor site for both locations with a statistical significance level, $p < 0.005$. At the NYU sites, which reflect present emission patterns, the TSP concentrations at the street site presently average close to the primary NAAQS of $75 \mu\text{g}/\text{m}^3$. In addition, the general decline in rooftop TSP values observed from 1969 through 1978 is also evident from the two periods of first floor level sampling.

The trends in the NYU first floor level Pb concentrations correlated with the rooftop values ($p < 0.01$) over the 1976-78 period, and the concentrations (Figure 2) were statistically different between the two elevations with a $p < 0.005$. The Pb concentrations at the rooftop averaged about $934 \text{ ng}/\text{m}^3$, and were usually between 60 and 80% of the first floor concentrations. Similar results were observed for the EML (HASL) data, although the concentrations at the roof were usually above $1500 \text{ ng}/\text{m}^3$. Recent results from the NYU sites indicate

Table I. Statistical relationships between the roof and first floor sites at both the New York University Medical Center and Environmental Measurements Laboratory, New York City

Variable	Mean concentration $\mu\text{g}/\text{m}^3$		Paired t-test Roof vs. first floor Concentrations		Least squares corr. coef. Roof- Ground
	Roof (X_R)	First floor (X_F) Medical Center	Sign. Level (p)	$X_F - X_R$ > 0	
TSP	58.7 ± 12.7	68.4 ± 10.0	<0.005	Yes	0.50*
Pb	0.934 ± 0.287	1.196 ± 0.387	<0.005	Yes	0.43*
Mn	0.020 ± 0.005	0.026 ± 0.008	<0.005	Yes	0.20
V	0.053 ± 0.030	0.047 ± 0.032	No sig. diff.	No	0.66*
Environmental Measurements Laboratory (HASL)					
TSP	103.3 ± 23.0	125.9 ± 25.6	<0.005	Yes	0.24
Pb	1.816 ± 0.394	2.614 ± 0.811	<0.005	Yes	0.41
Mn	$0.066 \pm .022$	0.060 ± 0.018	No sig. diff.	No	0.05
V	0.666 ± 0.383	0.667 ± 0.276	No sig. diff.	No	0.94*

* Level of significance $p < 0.01$ indicating an association between roof and first floor trends.

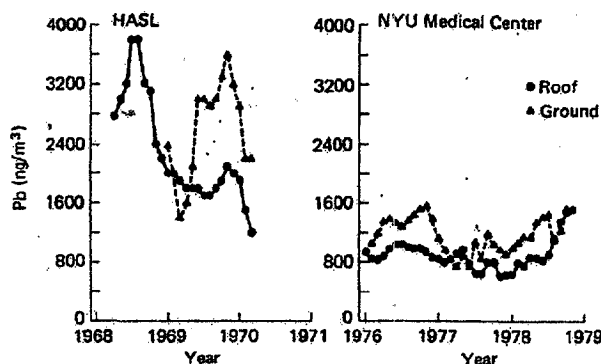


Figure 2. Rooftop and first floor (ground) elevation Pb concentrations for the HASL (EML) and NYU Medical Center Site as 3 month moving averages.

that the first floor and roof concentrations are not following the long-term trends and have been increasing toward 1600 ng/m³ (3 month moving average).

In contrast to the TSP and Pb results, no significant difference could be discerned between the concentrations of vanadium measured at the roof and first floor sites at each location. Moreover, V concentrations at both elevations had a significant correlation with $r = 0.66$ and 0.95 ($p < 0.1$) for NYU and EML, respectively.

The TSP, Pb, and V data have shown a general decrease in ambient concentration since 1968. This is also true for Mn at both the rooftop and first floor sites prior to 1976 with the first floor concentrations normally <25% higher than the rooftop concentrations (Figure 3). However, for a portion of 1976 and throughout most of 1978, the NYU first floor level Mn increased to 2.0–2.4 times greater than the rooftop concentration (Max. diff. = 25 ng/m³). In addition, for the 1976–78 period, the trends in rooftop and first floor Mn values did not correlate. However, when the two previously indicated periods are omitted, a correlation with significance $p < 0.01$ appears. In contrast, at EML (1968–70) no statistical difference was observed between the first floor and rooftop Mn concentrations. This is not apparent in the 3 month moving average because of the presence of two extreme excursions in Mn, one recorded at each elevation. These brief excursions can distort the moving averages while not affecting paired-*t* or statistical correlation tests. No such excursions in the monthly data were seen for the other elements.

Discussion

For both the rooftop and first floor level data, it is apparent that, since 1968, the concentrations of TSP, Pb, and V and Mn have been generally on the decline. Primarily, this has been the result of the use of low sulfur (and low ash) oil and the implementation of various control strategies on incinerators and automobile lead emissions. In addition, over the entire period, the concentrations of these materials have been below those associated with potential health effects, although at the first floor site TSP is still near the primary air pollution standard.

Comparison of the weekly average TSP and Pb concentrations recorded at the first floor level and the rooftop indicates that the first floor level concentrations are usually higher. This would be expected since the first floor level sampler is in closer proximity to the emissions which can be released directly or indirectly from the automobile or other ground level sources.

In contrast, there was no significant difference observed between the weekly average V concentrations measured at the two elevations for both locations. This is a consequence of the V being associated with power plants and residential and commercial boilers which have elevated emission points.

Under the influence of the atmospheric mixing processes, the V containing particles will mix downward and produce a fairly uniform column, which would lead to the observed pattern while ground level sources would yield a more pronounced vertical gradient. During the period 1968–70, the higher concentrations of V and TSP observed at the EML site were associated with fuels of high sulfur and V content and resulting emissions.

Manganese, however, exhibits two patterns. For the period, 1968–70, no difference was observed in the Mn concentrations measured at the rooftop and first floor. This result probably reflects the fact that Mn was being emitted by coal-fired energy sources, and the emission would occur at elevated points. It follows that the Mn distribution would be similar to the distribution described for V spatially if not temporally.

The 1976–78 sampling for Mn follows a different pattern than that above. The statistical analyses and visual comparisons of the curves indicate that there were significant differences between the rooftop and first floor concentrations which could be a result of increases in emissions from street level sources. This in fact was the case because of the use of methylcyclopentadienyl tricarbonyl (MMT) as an octane booster in no-lead gasoline. The resulting combustion products are primarily in the form of Mn₃O₄.¹¹ Since MMT reduced the efficiency of the exhaust catalyst, there was an initial demand for reduction in its use, with the EPA finally banning MMT in October 1978. Downward trends in the street level ambient concentrations have occurred in October–November 1976, and again in June–July 1978, which seem to reflect anticipation of the actual regulatory actions.

After the introduction of no-lead gasoline, the ambient Pb concentrations continued to decrease, and reached a low value of approximately 1000–1200 ng/m³ at the first floor site. The removal of MMT and its product Mn₃O₄ was not a singular event, as EPA permitted a phase-out period. Almost simultaneous with this activity, however, concomitant increases in the Pb were observed.

The health effects associated with ambient Mn are not clearly defined¹² although recent studies have indicated that effects should only occur at much higher concentrations.¹³ Conversely, Pb has been shown to accumulate in lung tissue when ambient concentrations are $>1.3 \mu\text{g}/\text{m}^3$ ^{14,15} (the new NAAQS is $1.5 \mu\text{g}/\text{m}^3$ monthly average). Presently, the Pb concentrations surpass these values, which indicates that the Pb increases could possibly, in the short term, result in the enhancement of a potential health hazard. Additional Pb increments to the air could yield greater risks to children in urban areas since there is believed to be a narrow margin of safety associated with current urban blood Pb levels.¹⁶ Further analyses to determine the reasons for the recent increases in Pb are planned, and hopefully will yield information which will be beneficial to the understanding of the present concentration patterns.

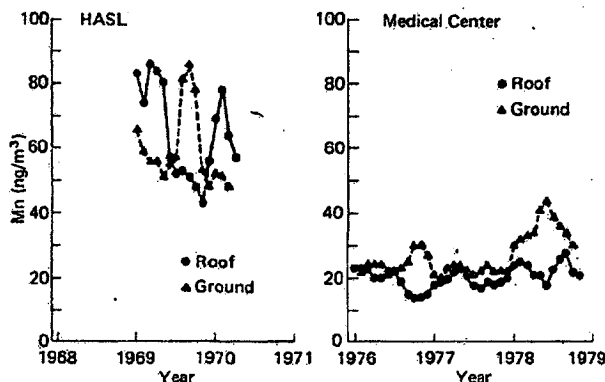


Figure 3. Rooftop and first floor (ground) elevation Mn concentrations for the HASL (EML) and NYU Medical Center Sites as 3 month moving averages.

Conclusions

The comparison of long-term rooftop versus first floor level airborne concentrations of various materials has been shown to be valuable in understanding the sources and distribution of TSP.

The comparison has also shown that it is possible to discern long term changes in ambient concentrations of materials at a first floor level site when there is an alteration in one of the additives used in gasoline. Since alterations in these and other source emissions can be significant, prudent examination of the potential health hazards associated with emissions from the combustion of new fuels must be continued.

Acknowledgments

The authors wish to thank Dr. Merrill Eisenbud who initiated the studies which have been conducted at the NYU rooftop, Dr. Morton Lippmann for his comments and suggestions, and James Miller, John Gorzynski and Bruce Naumann for their work on the sampling and analysis aspects of this study, and Dr. M. T. Kleinman of Rancho Los Amigos Hospital, CA, who supervised much of the sampling while at the Institute. The authors acknowledge for support of this research the American Petroleum Institute and the Electric Power Research Institute, Grant No. RP-1222-1 and RP-493-3, and as part of the center grant program supported by Grant No. ES-06260 from the National Institute of Environmental Health Sciences, and Grant No. CA 13343 from the National Cancer Institute.

References

1. *Airborne Particulates*, Subcommittee on Airborne Particles, Committee on Medical and Biologic Effects of Environmental Pollutants, National Research Council, University Park Press, Baltimore, MD, 1979.
2. T. J. Kneip, M. T. Kleinman, and M. E. Eisenbud, "Studies of Trace Substances in an Urban Aerosol," in *Proceedings of the International Symposium of Recent Advances in the Assessment of Health Effects of Environmental Pollution*, Paris, (June 24-28, 1974) 1975.
3. T. J. Kneip, M. T. Kleinman and M. E. Eisenbud, "Relative Contributions of Emissions Sources to the Total Airborne Particulate in New York City," *Proceedings of the Third International Clean Air Congress*, Dusseldorf, Germany, October, 1975.
4. T. A. Cahill and P. J. Feeney, "Contribution of Freeway Traffic to Air-borne Particulate Matter," Report No. ICD-CNL169, Crocker Nuclear Laboratory, University of California, June 1973.
5. R. H. Daines, H. Motto, and D. M. Chilko, "Atmospheric lead: its relationship to traffic volume and proximity to highways," *Env. Sci. Technol.* 4:318 (1970).
6. S. E. Bauman, E. T. Williams, H. L. Fenston, A. H. Bond, Jr., P. N. S. Lesser, and E. F. Ferrand, "Suspended Particulate Matter in NYC: Element Concentrations as a Function of Particle Size and Elevation above the Street," *Proceedings of the Third Int. Conf. on Nuclear Methods in Environ. and Energy Research*, NTIS No. 771072, University of Missouri, Columbus, MO, Oct., 1977.
7. M. E. Eisenbud, T. J. Kneip, M. T. Kleinman, and D. M. Bernstein, "Trace Elements in Urban Aerosols," Final Report to EPRI (October 1975), NTIS Publ. No. PB-248-324, 1976.
8. M. T. Kleinman, "The Apportionment of Sources of Airborne Particulate Matter," Ph.D. Thesis, New York University, June, 1977.
9. M. T. Kleinman, T. J. Kneip, and M. Eisenbud, "Seasonal patterns of airborne particulate concentrations in the New York City," *Atmos. Environ.* 9:9 (1976).
10. M. T. Kleinman, D. M. Bernstein and T. J. Kneip, "An apparent effect of the oil embargo on total suspended particulate matter and vanadium in New York City air," *J. Air Poll. Control Assoc.* 27:65 (1977).
11. D. P. Chock, "General Motors Sulfate Dispersion Experiment: assessment of the EPA Hiway Model," *J. Air Poll. Control Assoc.* 27:39 (1977).
12. *Manganese*, Committee on Biologic Effects of Atmospheric Pollutants, National Research Council, National Academy of Sciences, Washington, D. C., 1973.
13. C. E. Ulrich, N. Rinehart, and M. Brandt, "Evaluation of the chronic inhalation toxicity of a manganese oxide aerosol. III—pulmonary function electromyograms, limb tremor, and tissue manganese data," *J.A.I.H.A.* 40:349 (1979).
14. D. M. Bernstein, "The Influence of Trace Metals in Dispersed Aerosols on the Human Body Burden of Trace Metals," Ph.D. Thesis, New York University, October 1977.
15. D. M. Bernstein, T. J. Kneip, M. T. Kleinman, R. Riddick, and M. Eisenbud, "Uptake and Distribution of Airborne Trace Metals in Man," in: *Trace Substances in Environmental Health VIII. A Symposium*. Ed. D. D. Hemphill, University of Missouri, Columbus, MO, pp. 329-334, 1974.
16. "Air Quality Criteria for Lead," U.S. EPA, EPA-600/8-77-017, Washington, D. C., December, 1977.

Dr. Liroy is Assistant Professor of Environmental Medicine, Mr. Mallon is a graduate student and Dr. Kneip is Research Professor of Environmental Medicine, at the Institute of Environmental Medicine, New York University Medical Center, 550 First Avenue, New York, NY 10016.