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# The Association between Daily Mortality and Ambient Air Particle Pollution in Montreal, Quebec

## 1. Nonaccidental Mortality

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**This study was undertaken to determine whether variations in concentrations of particles in the ambient air of Montreal, Quebec, during the period 1984 to 1993, were associated with daily variations in nonaccidental mortality. Fixed-site air pollution monitors in Montreal provided daily mean levels of various measures of particulates and gaseous pollutants. Total sulfates were also measured daily (1986–1993) at a monitoring station 150 km southeast of the city (Sutton, Quebec). We estimated associations for $PM_{2.5}$, $PM_{10}$, total suspended particles, coefficient of haze (COH), extinction coefficient, and sulfates. We used coefficient of haze, extinction coefficient, and Sutton sulfates to predict fine particles and sulfates for days that were missing. To estimate the associations between nonaccidental mortality and ambient air particles, we regressed the logarithm of daily counts of nonaccidental mortality on the daily mean levels for the above measures of particulates, after accounting for seasonal and subseasonal fluctuations in the mortality time series, non-Poisson dispersion, weather variables, and gaseous pollutants. There were 140,939 residents of Montreal who died during the study period. We found evidence of associations between daily nonaccidental deaths and most measures of particulate air pollution. For example, the mean percentage increase (MPC) for an increase of total suspended particles of 28.57 μg/m³ (interquartile range, IQ), evaluated at lag 0 days, was 1.86% (95% confidence interval (CI): 0.00–3.76%), and for an increase of coefficient of haze (IQ = 18.5 COH units per 327.8 linear m) the MPC was 1.44% (95% CI: 0.75–2.14%). These results are similar to findings from other studies (the mean percentage increase in nonaccidental deaths for a 100 μg/m³ increase in daily total suspended particles was 6.7%). We also found increases for fine particles and for inhalable particles, but the confidence intervals included unity. All measures of sulfates showed increased daily mortality; e.g., the MPC for sulfates from fine particles (IQ = 3.51 μg/m³) was 1.86% (95% CI: 0.40–3.35%). We generally found higher excesses in daily mortality for persons 65 years of age and for exposures averaged across lags 0, 1, and 2 days. The slope of the association between daily mortality and ambient air particles in Montreal, which has lower levels of pollution than most major urban centers, is similar to that reported in most other industrialized cities. This study therefore provides further evidence that the association is linear and that any threshold effect, should it exist, would be found at lower levels of air pollution than those found in Montreal.** © 2001 Academic Press

*Key Words:* mortality; ambient air pollution; particulates; total suspended particles; $PM_{10}$; $PM_{2.5}$; sulfates; coefficient of haze; time series; epidemiology.

## INTRODUCTION

Concentrations of ambient air particles have been found to be associated with a wide range of effects on human health (e.g., Dockery and Pope, 1994; Goldberg, 1996; Schwartz, 1991, 1994a; Zmirou *et al.*, 1998). Although the cardiorespiratory system appears to be the principal target, positive associations

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have been observed across a wide range of endpoints and may reflect both short-term and long-term exposures. Positive associations between daily mortality and ambient concentrations of particle mass have been observed in several locales throughout the world having different chemical and physical compositions and mixtures of other gaseous pollutants. These associations are generally consistent with a linear exposure–response function and the range of increased daily mortality is from about 2 to 9% per 100 $\mu g/m^3$ increase in total suspended particles (e.g., Schwartz, 1994a; Zmirou *et al.*, 1998).

Critical research issues include determination of whether the association is causal, whether the exposure–response relationship found at lower levels of pollution is similar to that observed in the more polluted cities, and whether the association is stronger in certain subgroups of the population (Bates, 1992; Goldberg, 1996; Seaton *et al.*, 1995; Frank and Tankersley, 1997). The present investigation was designed to shed light on these questions. We exploit data from Montreal, Quebec, Canada, a large metropolitan area that experiences relatively low levels of air pollution. In this paper, we present a time series analysis of nonaccidental mortality; subsequent papers will focus on cause-specific mortality and the identification of frail subgroups by use of medical information collected before death.

## MATERIALS AND METHODS

The study population consisted of all residents of Montreal who died in the city during the period 1984 to 1993 and who were registered with the universal provincial health insurance plan (Goldberg *et al.*, 2000). Subjects were identified from the computerized provincial database of death certificates that provided identifying information on subjects, date of death, place of death, residence at time of death, and underlying cause of death as coded in the International Classification of Diseases, Ninth revision (ICD9).

Methods, time period of monitoring, number of monitoring stations, and approximate frequency of measurements of particulate and gaseous pollutants in Montreal are summarized in Table 1. Coefficient of haze (COH, a measure of organic and inorganic carbon) and the criteria gaseous pollutants (carbon monoxide (CO), carbon dioxide ($CO_2$), nitrogen oxide (NO), nitrogen dioxide ($NO_2$), sulfur dioxide ($SO_2$), and ozone ($O_3$)) were measured hourly. Total suspended particles (TSP), particulates having an aerodynamic diameter of 10 and 2.5 μm or less ($PM_{10}$, $PM_{2.5}$), and sulfates derived from these indices were measured at a frequency of every 6 days. The measurements of $PM_{10}$ and $PM_{2.5}$ were from two dichotomous sampler monitoring stations operating at a frequency of about every 6 days (Brook *et al.*, 1997a,b). From July 1992 to September 1995, the measurement schedule for these two pollutants was increased at one site (Ontario Street) to daily sampling (Brook *et al.*, 1997a).

Because sulfates were measured every 6 days, we used daily sulfate measurements (1986–1993) from a monitoring station of the Canadian Air and Precipitation Monitoring Network at Sutton, Quebec, a rural community about 150 km southeast from the city (Fig. 1). A filter-pack system was used and, thus, the measurements represent total sulfates. Use of these data is justified because the regional component of sulfates and fine particles is largely determined by long-range transport of pollutants, so that measurements at the Sutton station represent background levels throughout southwest Quebec, including Montreal (Brook *et al.*, 1997a). The correlation between sulfates measured at this station and the two Montreal stations was 0.9.

Concentrations of most pollutants from the different monitoring stations in Montreal were measured in laboratories of the Montreal Urban Community by use of methods approved by the U.S. Environmental Protection Agency. Most of these stations were included in Environment Canada's National Air Pollution Surveillance Program database, and Environment Canada regularly audited these stations for the gaseous pollutants. The quality assurance procedures for the Environment Canada dichotomous sampler measurements have been described previously (Brook *et al.*, 1997a). Quality of the Canadian Air and Precipitation Monitoring Network (CAPMoN) sulfate measurements was examined through regular testing of blanks, periodic calibration of mass flow controllers, and comparison with other networks (e.g., Canadian Acid Aerosol Measurement Program (CAAMP) sulfates (Brook *et al.*, 1997a) and USEPA CastNet sulfate (Clarke and Edgerton, 1997)).

Daily weather data, measured at Dorval International Airport, included visibility (from ground level), barometric pressure, temperature, total precipitation (distinguishing snow from rain), relative humidity, and dew point temperature. We calculated an extinction coefficient (a measure of light scattering and absorption, due mostly to sulfates) using visibility at noon unless there was precipitation. Otherwise, we searched for the first afternoon sighting when there was no precipitation; if sightings without precipitation were not found, we then

TABLE 1

Particulate and Gaseous Pollutant Data[a] Used in the Statistical Analyses, Montreal, 1984 to 1993

| Pollutant | Start year | No. of monitoring stations | Duration | Frequency | Sampling methods | Analytic methods |
|---|---|---|---|---|---|---|
| TSP | 1984 | 19 | 24 h | Every 6th day | High volume samplers (flow rate of 1.5 $m^3$ per min), midnight to midnight sampling | Washed glass filters, mass measured on Sartorius AC121S digital electronic balance |
| Sulfate from TSP | 1984 | 13 | 24 h | Every 6th day | High volume samplers (1.5 $m^3$ per min); midnight to midnight sampling | Soluble sulfates on filters extracted by hot water and analyzed by ionic chromatography |
| $PM_{10}$ | 1984 | 2 | 24 h | Every 6th day | Sierra-Anderson dichotomous (flow rate of 16.7 liters per min); midnight to midnight sampling | Electronic microbalance at constant temperature and relative humidity |
| Sulfate from $PM_{10}$ | 1984 | 2 | 24 h | Every 6th day | High volume samplers | Dionex ion chromatography |
| $PM_{2.5}$ | 1984 | 2 | 24 h | Every 6th day | Sierra-Anderson dichotomous (flow rate of 16.7 liters per min) | Electronic microbalance at constant temperature and relative humidity |
| Sulfate from $PM_{2.5}$ | 1984 | 2 | 24 h | Every 6th day | Same as $PM_{2.5}$ | Dionex ion chromatography |
| COH | 1984 | 11 | Hourly | Continuous, 2-h integrated sampling | AISI Sequential (RAC), using a continuous roll of white filter paper (flow rate of 0.4 $m^3$ per h) | Opacity as measured by photometer, in COH units per 0.8 $m^3$ of air |
| Sulfate measured at the Sutton acid rain monitoring station | 1986 | 1 | 24 h | Daily | Downward-facing, open-faced filterpack mounted at 10 m (25 $m^3$ per min mass flow controlled); 8 am to 8 am sampling | Dionex ion chromatograph |
| $SO_2$ | 1984 | 13 | Hourly | Continuous | Philips 9700, Monitor Lab 8850 | (1) Ultraviolet fluorescence and (2) electrical conductivity from changes in chemical composition of a bromine solution |
| $O_3$ | 1984 | 9 | Hourly | Continuous | Bendix 8002 | Chemiluminescence |
| NO | 1984 | 8 | Hourly | Continuous | Thermo Electron 14B | Chemiluminescence |
| $NO_2$ | 1984 | 8 | Hourly | Continuous | Thermo Electron 14B | Chemiluminescence |
| CO | 1984 | 12 | Hourly | Continuous | Thermo Electron 48 | Infrared absorption |

*Note.* CO, carbon monoxide; $CO_2$, carbon dioxide; COH, coefficient of haze; $NO_2$, nitrogen dioxide; NO, nitrogen oxide; $O_3$, ozone; $PM_{2.5}$, $PM_{10}$, particulate matter with an aerodynamic diameter of 2.5 and 10 μm; $SO_2$, sulfur dioxide; TSP, total suspended particles.

[a] PM was measured by Environment Canada as part of the National Air Pollution Surveillance Program. All other pollutants were measured by the Montreal Urban Community. The number of sites changed during the study period.

searched for valid sightings from noon into the morning. The extinction coefficient was calculated by use of the relationship [3.91/visual range (km)] × *f*(relative humidity), where *f*( ) is an empirical function of relative humidity (Özkaynak *et al.*, 1985; Kinney and Özkaynak, 1991; Delfino 1994). About 80% of the visibility data were from measurements made at noon, with other hours of the day (from about 9 am until 7 pm, depending on the season) being selected about equally

### *Interpolation of Missing Particulate Data*

We developed statistical models to estimate concentrations of fine particles and fine particle sulfate when measurements were not taken. We used the

FIG. 1. Map of the southwest region of the Province of Quebec, showing the location of the Island of Montreal and Sutton.

general linear regression prediction model,

$$
\begin{aligned}
E[\mathrm{PART}_i] = \alpha &+ \beta_1 \text{ Coefficient of haze}_i \\
&+ \beta_2 \text{ Extinction coefficient}_i \\
&+ \beta_3 \text{ Sutton sulfate}_i, \qquad (1)
\end{aligned}
$$

where $E[\mathrm{PART}_i]$ represents expected daily mass levels for $PM_{2.5}$ or sulfates from $PM_{2.5}$, i is an indicator for the day in the time series, and the coefficients ($\alpha$, $\beta$) were estimated separately in each regression model. Predictions of levels of particles for each day in the time series were thus calculated from the linear predictor of Equation (1). As measurements of sulfate at Sutton were available only since 1986, the years 1984 and 1985 were excluded from the analyses. The $R^2$ for the model predicting concentrations of $PM_{2.5}$ when measurements were available was 0.72 and for fine particle sulfates it was 0.80.

### *Estimates of the Effects of Particles on Daily Mortality*

We used quasi-likelihood estimation within the context of the Generalized Additive Models (Hastie and Tibshirani, 1990) to model the natural logarithm of the expected daily counts of deaths as a function of the predictor variables. These flexible regression models allow the estimation of nonparametric smooth functions for pollutants and potential confounding variables (time, weather variables). We used locally weighted regression smoothers (LOESS) that are akin to a moving average using information in a specified neighborhood around each data point (referred to as the span) to estimate an expected value for each data point. We assumed that counts of death were distributed approximately as a Poisson variate with constant over- or underdispersion. For each model the dispersion parameter was estimated and the covariance matrix was then corrected by multiplication of the usual Poisson covariance matrix by the estimated dispersion parameter.

We assumed that yearly, seasonal, and subseasonal variations in the mortality time series represented unmeasured processes (e.g., influenza epidemics) that may confound the association between mortality and air pollution. The basic approach was to treat these temporal effects and the weather variables as nuisance terms in the statistical models. We thus regressed the natural logarithm of the daily number of deaths on a LOESS temporal filter that adjusted for seasonal and subseasonal variations, another term to account for annual trends in daily mortality, and LOESS terms to adjust for the potential confounding effects of relevant weather variables.

In developing these statistical models, we first identified the temporal filter that removed unwanted seasonal and subseasonal cycles to produce an adjusted mortality time series. As there was a clear secular increase in mortality we also added a term for calendar year. We selected the span for the LOESS function of time (in days) of the mortality time series (the temporal filter) that produced a filtered time series that was as closely consistent with a white noise process as possible, as determined from the cumulative periodogram using Bartlett's statistic (Priestly, 1981). This process reduced the residual serial autocorrelation considerably and usually minimized the Akaike Information Criterion (AIC; equal to the residual deviance plus the product of twice the number of used degrees of freedom and the estimated dispersion parameter).

We then extended the model to incorporate the effects of weather. We considered daily mean temperature, daily mean dew point temperature, change in maximum temperature from the previous day, relative humidity, and change in barometric pressure from the previous 24 h. We examined the AICs from models for each separate weather variable, lagged from zero to 5 days before the day of death. We accounted for losses in observations due to the taking of lags, selected for each variable the lag that produced the minimum AIC, and constructed a series of models that included all combinations of these lagged variables. We thus investigated combinations of two individual smooths (LOESS($A_j$) + LOESS($B_k$), where j and k are lags from 0 to 5), pairwise smooth interactions (LOESS($A_j$, $B_k$)), and combinations of three individual smooths (LOESS($A_j$) + LOESS($B_k$) + LOESS($C_l$)). The weather variables from models yielding the minimum AIC were used in all subsequent analyses.

Last, we assessed the effects of the air pollution variables on daily mortality after incorporating into a regression model the selected temporal filter, the term for calendar year, and the term for weather conditions. Single-pollutant models using daily mean values across the fixed-site monitoring stations were considered first. We also estimated mortality with the previous day's level of air pollution (lag 1 day) and with the average of lags 0 to 2 days (referred to as the "3-day mean"). For those pollutants not measured daily, lags were taken by the shifting forward of the mortality time series. Multiday averages could not be derived for pollutants that were not monitored continuously.

All nontemporal, continuous covariates (weather, pollution) were entered into the model nonparametrically using a LOESS smoother (span = 50%). Exposure-response functions for the particle variables were plotted from the fitted model. We used an approximate *F* test to determine whether the estimated function was consistent with linearity (Hastie and Tibshirani, 1990).

We also estimated parametric, linear terms for the relative change in the logarithmic number of daily deaths per unit increase in the pollutant. We calculated the percentage change in the mean number of daily deaths for an increase of the interquartile range (IQ) for each index of particles, i.e., as $[\exp(\beta \times IQ) - 1] \times 100\%$, where $\beta$ is the estimated regression coefficient. We refer to this quantity as the "mean percentage change" (MPC). Associated upper and lower 95% confidence limits on the mean percentage change were obtained under the assumption that the estimated regression coefficient was distributed normally, with the standard error corrected for non-Poisson dispersion.

## RESULTS

Table 2 shows correlations between monitoring stations and secular trends for each pollutant. As an

TABLE 2

Correlations and Trends in Measured Environmental Pollutants, Montreal, 1984–1993

| Pollutant | Units | Number of monitoring stations | Range of mean values between stations, 1984–1993 | Coefficient of variation of daily means, 1984–1993 (%) | Range of Pearson correlation coefficients between monitoring stations | Means of daily means across study years: Mean 1984–1985 | Mean 1992–1993 | Average annual change[a] |
|---|---|---|---|---|---|---|---|---|
| TSP | μg/m³ | 19 | 32.9–88.3 | 42.6 | 0.2–0.9 | 59.4 | 44.4 | –2.08 |
| $PM_{10}$ | μg/m³ | 2 | 27.8–44.5 | 54.7 | 0.7 | 40.0 | 26.4 | –1.98 |
| $PM_{2.5}$ | μg/m³ | 2 | 16.2–21.1 | 65.3 | 0.9 | 20.2 | 14.9 | –0.87 |
| Sulfate from TSP | μg/m³ | 13 | 2.3–6.0 | 68.7 | 0.3–1.0 | 5.2 | 3.6 | 0.21 |
| Sulfate from $PM_{10}$ | μg/m³ | 2 | 4.6 | 92.0 | 0.9 | 4.9 | 4.5 | 0.08 |
| Sulfate from $PM_{2.5}$ | μg/m³ | 2 | 4.0–4.2 | 97.7 | 0.9 | 4.3 | 4.1 | 0.06 |
| Sulfate from the Sutton monitoring station[b] | μg/m³ | 1 | 3.35 | 107.5 | | 2.83 | 2.9 | 0.03 |
| COH | 0.1 COH units per 327.8 linear meters | 11 | 1.2–4.2 | 63.9 | 0.2–0.7 | 2.6 | 1.9 | 0.10 |
| Extinction (corrected) | $m^{-1}$ | 1 | 0.15 | 66.7 | | 0.15 | 0.15 | 0.00 |
| $SO_2$ | μg/m³ | 13 | 11.1–30.7 | 63.2 | 0.1–0.8 | 22.6 | 14.0 | –0.96 |
| $NO_2$ | μg/m³ | 8 | 30.3–61.9 | 37.0 | 0.2–0.6 | 44.9 | 40.2 | –0.45 |
| NO | μg/m³ | 5 | 20.2–89.9 | 69.4 | 0.3–0.8 | 51.0 | 36.4 | –1.73 |
| CO | ppm | 12 | 0.5–5.8 | 58.8 | 0.1–0.8 | 1.1 | 0.6 | –0.77 |
| $O_3$ | μg/m³ | 9 | 16.6–43.1 | 59.0 | 0.5–0.9 | 29.3 | 28.9 | 0.08 |

[a] Calculated from a linear regression model

[b] For the period 1986–1993.

[c] For 1986.

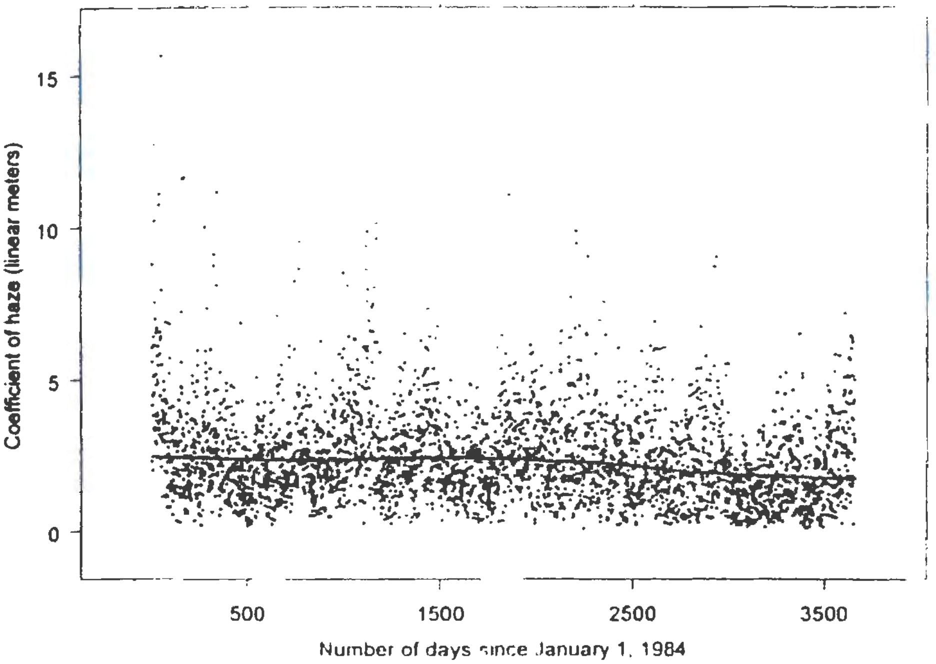

**FIG. 2.** Scatterplot of mean daily coefficient of haze. The solid line is the LOESS smooth representing the long term trend in the data (span of 50% of the data).

illustration, we show in Fig. 2 the time series plot of one of the principle particle pollutants, COH. (All other time series graphs are available from the first author at *http://www.epi.mcgill.ca.)* There was considerable temporal variation in mean daily levels of particles and gaseous pollutants. With the exception of $O_3$ and the extinction coefficient, mean levels of TSP, $PM_{10}$, $PM_{2.5}$, sulfates, COH, $SO_2$, $NO_2$, NO, and CO decreased during the study period. Fine and coarse particle mass, TSP, COH, extinction coefficient, and gaseous pollutants (except $O_3$) had peaks in the winter; sulfates and $O_3$ had peaks in the summer months.

During the study period, there were totals of 140,939 deaths and 133,904 nonaccidental deaths (Goldberg *et al.*, 2000). Table 3 shows the distribution of the number of nonaccidental deaths by two broad age groups and Fig. 3A shows the time series plot. The large amount of overdispersion (expected Poisson variance of 38.6 per day, observed variance of 52.8) arose mostly from dramatic seasonal fluctuations in daily mortality. This time series also exhibited substantial serial autocorrelation (Pearson correlation coefficients for lags 1 to 5 days: 0.26, 0.22, 0.22, 0.20, 0.20, respectively).

We found that a span of 91 of 3653 days (2.49% of the data) for the LOESS temporal filter minimized the Bartlett's test for white noise (Bartlett's statistic = 1.415, $P = 0.036$). Although this $P$ value suggests that the resultant time series was not entirely consistent with white noise, other spans (e.g., 31 and 151 days) led to even smaller $P$ values. The low $P$ values were due to a few days in which mortality was rather high (Fig. 3), but we did not find any compelling reasons to exclude these days. With this span, the serial autocorrelation (Pearson autocorrelation coefficients for lags 1 to 5 days: 0.04, 0.00, 0.00, −0.02, −0.03, respectively) was reduced dramatically and the AIC was a minimum compared to the other spans that were tested. Figure 3B shows the filtered mortality time series. After incorporation of temporal effects, the LOESS interaction of mean temperature and change in barometric pressure from the previous day, both evaluated at lag 0 days, had the lowest AIC. This last model, incorporating the temporal filter, calendar year, and the interaction term with mean temperature and change in barometric pressure, was used as the baseline model for consideration of the effects of air pollution.

### *Results by Particle Measure*

Across the indices of particles considered in our analyses, we found that the mean percentage change in daily nonaccidental mortality, for an increase in

TABLE 3

Distribution of Variables, Montreal, 1984–1993

| | Units | Number of days of measurements | Mean | Standard deviation | Minimum | Percentiles 25th | Percentiles 50th | Percentiles 75th | 100th | Interquartile range |
|---|---|---|---|---|---|---|---|---|---|---|
| All nonaccidental causes of death[a] | | 3653 | 38.6 | 7.27 | 0 | 33 | 38 | 43 | 91 | 10 |
| < 65 years old | | 3652 | 10.0 | 3.24 | 0 | 8 | 10 | 12 | 26 | 4 |
| ≥ 65 years old | | 3653 | 28.6 | 6.39 | 0 | 24 | 28 | 33 | 80 | 9 |
| TSP | μg/m³ | 603 | 53.1 | 22.6 | 14.6 | 37.0 | 48.7 | 65.6 | 211.1 | 28.57 |
| $PM_{10}$ | μg/m³ | 624 | 32.2 | 17.6 | 6.5 | 19.7 | 28.5 | 41.1 | 120.5 | 21.32 |
| $PM_{2.5}$ | μg/m³ | 636 | 17.4 | 11.4 | 2.2 | 9.4 | 14.7 | 21.9 | 72.0 | 12.51 |
| Sulfate from TSP | μg/m³ | 607 | 4.3 | 2.9 | 0.3 | 2.3 | 3.6 | 5.3 | 19.2 | 3.02 |
| Sulfate from $PM_{10}$ | μg/m³ | 437 | 4.7 | 4.4 | 0.3 | 1.9 | 3.6 | 5.7 | 30.7 | 3.84 |
| Sulfate from $PM_{2.5}$ | μg/m³ | 446 | 4.3 | 4.2 | 0.2 | 1.6 | 3.1 | 5.1 | 29.2 | 3.51 |
| Sulfate from the Sutton monitoring station[b] | μg/m³ | 2680 | 3.3 | 3.6 | 0 | 1.3 | 2.2 | 3.8 | 30.0 | 2.50 |
| COH | 0.1 COH units per 327.8 linear meters | 3653 | 2.4 | 1.5 | 0.1 | 1.3 | 2.1 | 3.2 | 15.6 | 1.85 |
| Extinction | $km^{-1}$ | 3454 | 0.15 | 0.10 | 0.01 | 0.06 | 0.15 | 0.17 | 1.87 | 0.11 |
| Predicted $PM_{2.5}$[b] | μg/m³ | 3653 | 17.6 | 8.8 | 4.6 | 11.5 | 15.4 | 21.0 | 71.7 | 9.51 |
| Predicted Sulfate from $PM_{2.5}$[b] | μg/m³ | 3653 | 4.1 | 3.6 | 0.02 | 1.9 | 3.1 | 4.8 | 30.1 | 2.88 |
| $SO_2$ | μg/m³ | 3653 | 17.8 | 11.2 | 3.9 | 10.3 | 14.6 | 21.8 | 105.7 | 11.50 |
| $NO_2$ | μg/m³ | 3653 | 41.7 | 15.4 | 8.8 | 30.9 | 39.5 | 50.2 | 143.5 | 19.34 |
| NO | μg/m³ | 3653 | 41.8 | 29.0 | 2.7 | 21.9 | 34.8 | 52.3 | 281.4 | 30.41 |
| CO | ppm | 3653 | 0.8 | 0.5 | 0.1 | 0.5 | 0.7 | 1.0 | 5.1 | 0.50 |
| $O_3$ | μg/m³ | 3653 | 29.0 | 17.1 | 2.8 | 16.6 | 26.0 | 37.9 | 163.9 | 21.34 |
| Mean temperature | C | 3653 | 6.4 | 11.8 | 27.3 | -2.6 | 7.5 | 16.5 | 28.8 | 19.10 |
| Change in maximum temperature from the previous day | C | 3653 | 0.0 | 4.7 | 25.0 | -2.5 | 0.4 | 2.8 | 19.4 | 5.30 |
| Mean dew point temperature | C | 3653 | 1.2 | 11.6 | -33.8 | -6.6 | 2.0 | 10.8 | 23.1 | 17.40 |
| Relative humidity | % | 3653 | 62.4 | 11.5 | 33.0 | 61.0 | 70.0 | 78.0 | 99.0 | 17.00 |
| Change in barometric pressure from previous day | kPa | 3653 | 0.0 | 0.9 | 4.2 | 0.5 | 0.0 | 0.6 | 4.4 | 1.11 |

[a] The total number of nonaccidental deaths was 133,904 ( < 65 years. 31,756; ≥ 65 years: 102,148).

[b] For the period 1986–1993.

levels of particles across the interquartile ranges (Table 3), ranged from 0.8 to 2.3% (Table 4). COH, the extinction coefficient, predicted $PM_{2.5}$ (derived for the period 1986-1993 from the linear regression model including coefficient of haze, extinction coefficient, and sulfate from the Sutton monitoring station), and all measures of sulfates showed increases across all of the lags considered, with generally higher effects found for the 3-day mean. $PM_{2.5}$ and $PM_{10}$ did not show any significant increases and the effect of TSP at lag 1 day was reduced considerably from that observed at lag 0 days.

Figures 4A–4C show the results for each measure of particle according to age group ( < 65 and ≥ 65 years) and lag period. The figures show rather large confidence intervals that reflect the statistical noise inherent in these time series analyses and the relatively small samples for pollutants measured every 6 days. At lag 0 days, we found higher increases in daily mortality among the elderly, except for $PM_{2.5}$ and sulfates from $PM_{2.5}$. Consistently higher MPCs were found for all measures of particulates evaluated at the 3-day mean, especially among subjects age 65 years and over.

We assumed in these analyses that a parametric linear term adequately represented the pattern of exposure to particles and daily mortality. We evaluated this assumption using a partial *F* test and

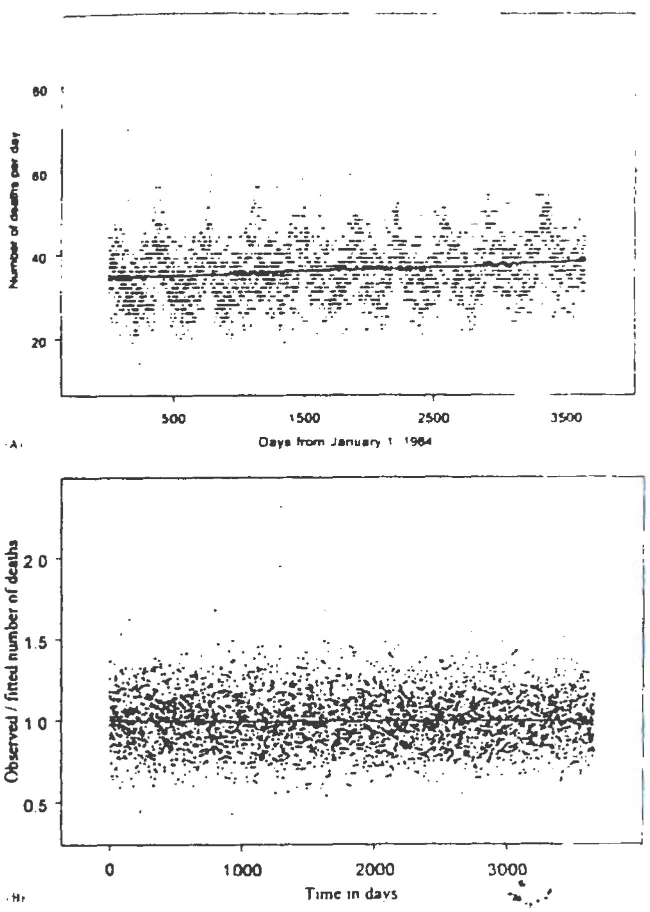

**FIG. 3.** (A) Scatterplot of daily number of nonaccidental causes of death; (B) scatterplot of the filtered time series for the daily nonaccidental causes of death. The solid line is the LOESS smooth representing the long-term trend in the data (span of 50% of the data). The total number of days in the time series is 3653.

inspected the graphs of the fitted functions to determine whether there were any important nonlinearities in the estimated exposure-response functions. We found "J-shaped" relationships for the coefficient of haze and predicted PM$_{2.5}$ evaluated at the 3-day mean.

### *Sensitivity Analyses*

We conducted a series of sensitivity analyses to determine whether the results reported above were modified when different temporal filters were used, when different weather variables were used, or when gaseous pollutants were included as covariates in the analyses (Table 5). Very few important differences were found between temporal filters having spans of 31 days of the data, 91 days, or 151 days. Using the 91-day filter, we also found that the estimates did not change dramatically across the different weather models, although exclusion of all weather variables increased the estimates. Estimates varied somewhat according to which copollutant was added to the model, with some pollutants increasing and others decreasing the estimates. For the coefficient of haze, the inclusion of sulfur dioxide decreased the estimates but the addition of ozone increased effects. For the analysis of sulfates and predicted PM$_{2.5}$, sulfur dioxide had little effect on the results, whereas the effects were reduced by ozone; this is likely due to the high positive correlation between these two regional pollutants, which often increase with similar types of weather conditions.

## DISCUSSION

A principal finding of this investigation was that the daily mean number of nonaccidental deaths was higher when levels of ambient particles were higher on the day of death (lag 0 days), on the day before death (lag 1 day), and for the average of the 3 days preceding and including the day of death (3-day mean). For the most part, the associations were consistent with a linear exposure–response relationship. We found that the magnitude of the associations on the day of death did not differ in the two age groups considered (less than 65 years and 65 years and over), but larger effects in the elderly were found when the 3-day mean was considered.

### *Comparison with Published Results*

We have presented our results in terms of interquartile ranges so as to facilitate comparisons among the various indices of particulates. For comparisons with other studies, we reexpressed our estimates in terms of $100\ \mu g/m^3$ increases (Table 6). Our results for coefficient of haze and total suspended particles are similar to the results of other studies (Schimmel and Murawski, 1976; Mazumdar and Sussman, 1983; Fairley, 1990; Kinney and Özkaynak, 1991). For PM$_{2.5}$ and PM$_{10}$, our estimates are also consistent with published data, but tend to fall in the lower range of results; however, predicted PM$_{2.5}$ is more in accord with the published figures. We found that the percentage increases in daily nonaccidental deaths were (at lag 0 days) 32.7% for Sutton sulfates and 48.3% for predicted sulfates measured from PM$_{2.5}$. These results are similar to those of Schwartz and co-workers (1996) who found an overall increase of 24.5% in daily mortality for

### TABLE 4

**Summary Estimates of the Mean Percentage Change in Daily Nonaccidental Mortality across the Interquartile Range of Selected Indices of Ambient Air Particles, Montreal, 1984–1993[a]**

| | Lag 0 days | | Lag 1 day | | 3-day mean | |
|---|---|---|---|---|---|---|
| | Mean percentage change[b] | 95% CI | Mean percentage change | 95% CI | Mean percentage change[b] | 95% CI |
| Coefficient of haze | 1.44 | 0.75-2.14 | 1.12 | 0.42-1.82 | 1.98 | 1.07-2.90 |
| Extinction coefficient | 1.05 | 0.43-1.68 | 0.86 | 0.23-1.48 | 1.67 | 0.52-2.53 |
| Total suspended particles | 1.86 | 0.00-3.76 | 1.04 | 0.80-2.93 | NC | |
| $PM_{10}$ | 1.43 | 0.33-3.22 | 0.80 | 0.93-2.56 | NC | |
| $PM_{2.5}$ | 0.77 | -0.76-2.32 | 1.45 | -0.04-2.97 | NC | |
| Predicted $PM_{2.5}$[c] | 1.86 | 1.11-2.61 | 1.48 | 0.74-2.24 | 2.17 | 1.26-3.08 |
| Sulfates measured at the Sutton acid rain monitoring station[c] | 0.71 | 0.22-1.20 | 0.95 | 0.47-1.43 | 1.29 | 0.68-1.90 |
| Sulfates estimated from total suspended particles | 2.26 | 0.73-3.81 | 2.20 | 0.73-3.70 | NC | |
| Sulfates estimated from $PM_{10}$ | 1.89 | 0.32-3.48 | 1.58 | 0.12-3.08 | NC | |
| Sulfates estimated from $PM_{2.5}$ | 1.86 | 0.40-3.35 | 1.62 | 0.25-3.02 | NC | |
| Predicted sulfates from $PM_{2.5}$[c] | 1.15 | 0.59-1.72 | 1.15 | 0.59-1.71 | 1.59 | 0.90-2.28 |

[a] The statistical model was $E(\log(y_i)) = \alpha + \text{LOESS}(i, \text{span} = 91/3653) + \beta$ year + LOESS(mean temperature$_i$, change in barometric pressure from previous day$_0$, span = 0.5) + $\beta \times$ pollutant, where $i$ is an indicator for day. CI, confidence interval; NC, not calculated.

[b] MPCs calculated for an increase of exposure equal to the interquartile range of each separate pollutant.

[c] Time period covered in 1986–1993.

the cities that comprised the Harvard six-cities cohort. Burnett and colleagues (1998) also found that a 100 µg/m$^3$ increase in sulfates was associated with a 21% increase in nonaccidental mortality in Toronto over the 15-year period 1980 to 1994.

#### *Methodologic Aspects*

*Confounding.* Weather is associated with both daily mortality and air pollution. Our strategy was to select weather variables that minimized the residual variability in the mortality time series (minimum AIC criterion), after accounting for annual, seasonal, and subseasonal patterns. A wide range of combinations of weather variables across lags zero to 5 days were first considered before the final set of variables were selected. This strategy, which was driven by statistical considerations, has the advantage of being transparent and reproducible.

Montreal is subject to seasonal variations in temperature and humidity, but few persons suffer badly from the cold because all homes are heated and most are reasonably well insulated. Also, the social assistance support program of Canada protected individuals from the extremes of winter weather. Hot and humid conditions in the summer are substantially less severe than those experienced in the central and southern United States. Thus, in view of the sociodemographics and weather patterns in Montreal, weather should not be a strong confounding factor, and this is what was found, as the effects of particles were rather insensitive to the range of weather models considered. These observations are consistent with those of Samet and colleagues (1998) who considered a wide variety of options to control for weather in Philadelphia.

We adjusted for the effects of the gaseous pollutants. Many of these pollutants are generated from complex reactions in air (e.g., ozone), and some gaseous pollutants (nitrogen dioxide, sulfur dioxide) act as precursors in the formation of particles. These atmospheric chemical processes, plus the overarching influence of weather conditions, create high correlations among levels of most pollutants. As there is much overlap in the sources for the pollutants, care must be taken in the inclusion of these other pollutants in the statistical models, as their inclusion may lead to biased estimates of the effects of particles. We found differences in the estimates of the effects of particles after adjusting for the gaseous pollutants, with some copollutants increasing and others decreasing the estimates. Fortunately, the

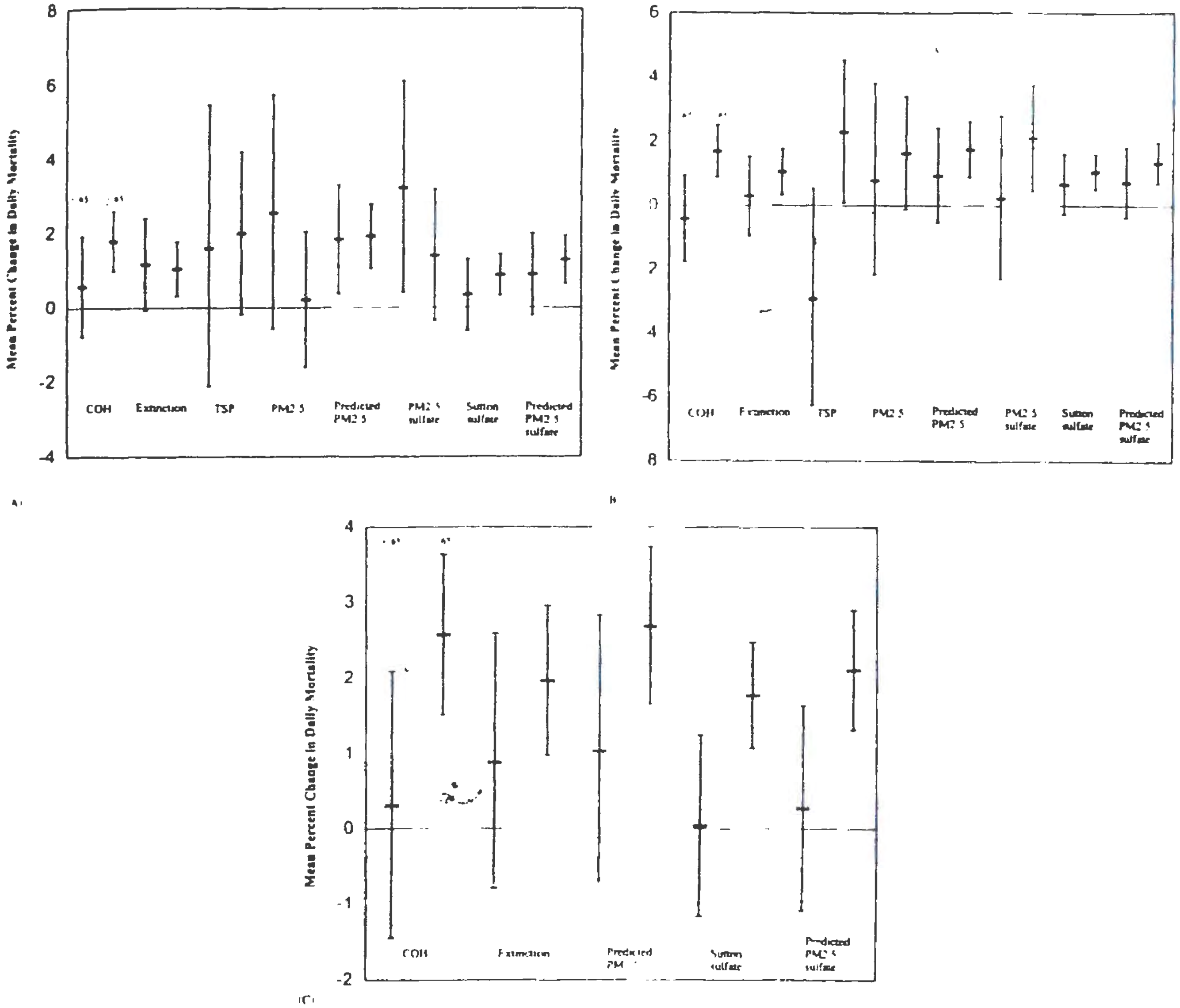

**FIG. 4.** (A) Associations between nonaccidental deaths and selected measures of particulate air pollution evaluated at lag 0 days, by age group; (B) associations between nonaccidental deaths and selected measures of particulate air pollution evaluated at lag 1 days, by age group; (C) associations between nonaccidental deaths and selected measures of particulate air pollution evaluated at the 3-day mean, by age group. Horizontal bars within the vertical lines are the estimated mean percentage change in daily nonaccidental mortality for an increase in levels of pollution across the interquartile ranges, and the vertical lines are the 95% confidence intervals.

differences were not profound, so our conclusions from the unadjusted analyses are unaltered.

We could not control for the effects of infectious disease epidemics (e.g., influenza, which occurs mostly in the fall and winter, when particle levels are increased) because there are no data bases that could be used for this purpose. Because these epidemics generally follow seasonal and subseasonal weather patterns, it is likely that some or all of the confounding effects were removed during the temporal filtering. However, some residual confounding effects remain possible. In a few studies, adjustment for influenza epidemics did not remove associations between mortality and suspended particles or sulfur dioxide (Spix *et al.*, 1993; Anderson *et al.*, 1996; Vigotti *et al.*, 1996).

*Components of ambient particles.* Despite the infrequent use of the coefficient of haze in time series analyses, it is a reliable measure of the

TABLE 5

**Sensitivity Analyses for Nonaccidental Deaths Evaluating the Effect of Different Temporal Filters, Different Weather Variables, and Adjustment for Different Gaseous Pollutants on the Estimates of Excess Relative Risk for Selected Pollutants across the Interquartile Range at Lag 0 day, Montreal, 1984–1993**

| | Temporal filter (span, in days/3653) | | | Weather Variables | | | Simultaneous adjustment for different gaseous pollutants evaluated at lag 0 days | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | 31 | Primary analysis (91) | 115 | Primary analysis[a] | Single variable[b] | None | Primary analysis ($M_1$) | $M_1$ + lo($SO_2$) | $M_1$ + lo(CO) | $M_1$ + lo($NO_2$) | $M_1$ + lo(NO) | $M_1$ + lo($O_3$) | $M_1$ + lo($SO_2$) + lo($CO_2$) + lo($NO_2$) + lo(NO) + lo($O_3$) |
| Coefficient of haze | 1.19* | 1.44* | 1.57* | 1.44* | 1.46* | 1.50* | 1.44* | 1.11* | 1.48* | 1.31* | 2.05* | 1.66* | 1.64* |
| $PM_{2.5}$ | 1.37 | 0.77 | 0.89 | 0.77 | 0.76 | 1.82* | 0.77 | 1.12 | 0.80 | 1.01 | 1.16 | 0.91 | 1.16 |
| Sulfate from the sutton monitoring station | 0.63* | 0.71* | 0.84* | 0.71* | 0.93* | 1.29* | 0.71* | 0.61* | 0.57* | 0.58* | 0.69* | 0.40 | 0.25 |
| Predicted $PM_{2.5}$ | 1.91* | 1.86* | 1.95* | 1.86* | 2.00* | 2.33* | 1.86* | 1.89* | 1.49* | 1.96* | 1.91* | 1.66* | 1.42* |

[a] Included the interaction term: LOESS (mean temperature$_0$, change in pressure$_0$).

[b] Included mean temperature evaluated at lag 0 days as the single weather variable having the lowest AIC (across lags 0 to 5 days).

*Indicates a corrected *t* value > 1.96.

concentration of ambient carbon particles (generally from internal combustion), with only limited contributions from other pollutants, such as sulfates, nitrates, or particle mass (J. Brook, personal contribution). Thus, information from this measure is complimentary to other measures of particle mass and sulfates regarding the effects of air pollution on human health.

The extinction coefficient, as calculated from line-of-sight distance visibility measures at the airport, is a rather crude index of particles and may lead to a higher degree of misclassification than the measurements of particle mass. Measurements of sulfates at the Sutton acid rain monitoring station captured regional sulfate levels but, despite the high correlation with measurements of sulfate in Montreal, was likely misclassified because it would not capture local fluctuations. Moreover, statistical power in the analyses of Sutton sulfate was reduced because only 8 years of measurements were available. Reduced statistical power is a more serious concern in the analyses of total suspended particles and PM because the nominal 6-day sampling schedule led to only about 600 days of available measurements.

As we did not have a complete series of fine particle measurements, we created one using a linear regression model that included coefficient of haze, sulfate from the Sutton monitoring station, and extinction coefficient. We found models for $PM_{2.5}$ and for sulfate from $PM_{2.5}$ that were reasonably good ($R^2$ of 72 and 80%, respectively) and, due to the inclusion of regional sulfate, local visibility (influenced largely by sulfate and nitrate particles), and local COH measurements, the modeled measures of particles (e.g., predicted $PM_{2.5}$) reflect the main components expected to influence urban levels of $PM_{2.5}$.

It has been speculated that acid aerosols may be a key factor mediating the association between particulate air pollution and adverse health outcomes (Spengler *et al.*, 1990; Lippmann and Thurston, 1996). On the other hand, no associations have been found in the two published mortality studies reporting this exposure (Dockery *et al.*, 1992; Schwartz *et al.*, 1996). As daily measurements of acid in Montreal were available for only 2 years, we could not test this hypothesis. As in other mortality and morbidity time series studies (Burnett *et al.*, 1995), we also found associations with sulfate taken from measurements of total suspended particles, PM, and total sulfates from the Sutton acid rain monitoring station.

Our estimates of the effect of sulfate obtained from total suspended particles may well be attenuated, as sulfates from total suspended particles will have a sulfur dioxide artifact. In our Toronto mortality study (Burnett *et al.*, 1998), we reported that the slope of the regression line between sulfate from high-volume samplers using glass fiber filters and sulfate from the dichotomous monitors that use Teflon filters was 0.87. The correction to our data would

### TABLE 6

**Comparison of Percentage Increases for Nonaccidental Deaths in Relation to Increases of 100 μg/m³ for TSP, $PM_{10}$, and $PM_{2.5}$ between the Present Investigation (Unadjusted for Gaseous Pollutants) and Selected Other Studies**

| City (reference) | TSP (%) | $PM_{10}$ (%) | $PM_{2.5}$ (%) |
|---|---|---|---|
| Present study—lag 0 days | 6.7 | 6.9 | 6.3 |
| Present study—lag 1 day | 3.7 | 3.8 | 12.2 |
| Present study—predicted $PM_{2.5}$, lag 0 days | | | 21.4 |
| Present study—predicted $PM_{2.5}$, lag 1 day | | | 16.7 |
| Present study—predicted $PM_{2.5}$, 3-day mean | | | 25.3 |
| North and South American studies[a] | | | |
| Birmingham (Schwartz, 1993; Samet *et al.*, 1995) | | 10.7-11.0 | |
| Cincinnati (Schwartz, 1994b) | 6.0 | | |
| Cooke County, Illinois (Ito and Thurston, 1996) | | 5.0 | |
| Cooke County, Illinois: ≥ 65 years of age (Styer *et al.*, 1995) | | 5.5 | |
| Detroit (Schwartz, 1991) | 6.0 | | |
| Eastern Tennessee (Dockery *et al.*, 1992; Samet *et al.*, 1995) | | 17.4 | 26.0 |
| Harvard six-cities, mean (Schwartz *et al.*, 1996) | | 8.3 | 16.1 |
| Los Angeles (Kinney *et al.*, 1995) | | 5.0 | |
| Mexico City (Borja-Aburto *et al.*, 1997) | 2.0–6.0 | | |
| Minneapolis (Schwartz, 1994a) | 5.0 | | |
| Philadelphia (Moolgavkar *et al.*, 1995) | 1.6 | | |
| Philadelphia (Samet *et al.*, 1995)[c] | 2.7–8.6 | | |
| Philadelphia (Schwartz and Dockery, 1992a) | 7.0 | | |
| Salt Lake County, Utah: ≥ 65 years of age (Styer *et al.*, 1995) | | 0.0 | |
| Utah Valley (Pope *et al.*, 1992; Samet *et al.*, 1995) | | 16.0–17.4 | |
| Sao Paulo (Saldiva *et al.*, 1995) | | 13.0 | |
| St. Louis (Dockery *et al.*, 1992; Samet *et al.*, 1995) | | 16.2 | 19.0 |
| Steubenville (Moolgavkar *et al.*, 1995) | 2.0 | | |
| Steubenville (Schwartz and Dockery, 1992b) | 4.0 | | |
| European Studies | | | |
| Amsterdam (Verhoeff *et al.*, 1996) | | 2.3-6.2 | |
| Athens (Touloumi *et al.*, 1996)[b] | | 5.0 | |
| Barcelona (Sunyer *et al.*, 1996)[b] | | 7.0 | |
| Bratislava (Bacharova *et al.*, 1996) | 0.0 | | |
| Bratislava (Bacharova *et al.*, 1996) | 1.0 | | |
| East Berlin (Rahlenbeck *et al.*, 1996)[c] | 4.6–6.2 | | |
| Koln (Spix and Wichmann, 1996) | 5.0–6.9 | | |
| London (Anderson *et al.*, 1996)[b] | | 12.8–21.8 | |
| Lyon (Zmirou *et al.*, 1996)[d] | | 2.0 | |
| Rome (Michelozzi *et al.*, 1998)[c] | 3.9 | | |
| Santiago, Chile (Ostro *et al.*, 1996) | | 8.0 | |
| Valencia, Spain (Ballester *et al.*, 1996, 1997)[b] | | 9.4 | |

[a] Various models, not accounting for season.

[b] Assuming black smoke approximately equivalent to $PM_{10}$ (Dockery and Pope, 1994; Muir and Laxen, 1995).

[c] Assuming suspended particles approximately equivalent to TSP

[d] Assuming $PM_{13} \sim PM_{10}$.

thus be to multiply the regression coefficient for sulfate from TSP by 1.1494.

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