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Particulate Data from the First Year of Monitoring in Phoenix: Part I. Fine and Coarse Mass

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Abstract

As a result of recent findings of statistical relationships between ambient particulate matter concentrations and mortality, the EPA's Office of Research and Development has begun to establish monitoring sites to collect data which have more detail (in terms of size cuts and composition) than those used in previous epidemiological studies. The Phoenix site was designed to evaluate various particulate samplers in an area that is expected to be heavily influenced by a component of windblown dust.

This paper examines one year (February 1995-January 1996) of particulate mass data from that site. The data include 24-hr fine (PM2.5) and coarse (PM2.5 to PM10) mass fractions from three different samplers. In addition, 1-hr PM2.5 and PM10 measurements are available from continuous methods and are supported by 1-hr meteorological data. This paper also addresses method comparisons, precision and accuracy of the instruments, and characterization of the relation between fine and coarse mass. Seasonal and meteorological influences on the distributions of mass are explored. Special studies on the reliability of the continuous methods are examined.

Introduction

In February 1995 the National Exposure Research Laboratory (NERL) at Research Triangle Park, N.C. began the collecting particulate matter (PM) data with regard to aerosol size and composition in Phoenix, AZ. This study is part of an ongoing project identified as the National PM Research Monitoring Network which was initiated in response to the EPA's Office of Research and Development's (ORD) research strategy to examine PM associations with morbidity and mortality in various neighborhoods in different U.S. metropolitan areas with widely differing PM size and composition characteristics [1]. The Phoenix study primarily focused on the operational aspects of collecting and characterizing daily PM size and composition data. Along with meteorological data, daily fine (0-2.5 μm), PM10 (0-10 μm) and coarse (2.5-10 μm) particle mass ($\mu g/m^3$) including metals and organic carbons were collected. Particles were measured with four different makes of samplers: 1) Tapered Element Oscillating Microbalance (TEOM) (Rupprecht & Patashnick, Inc. Series 1400a), 2) Dual Fine Particle Sequential Sampler (DFPSS), 3) Versatile Air Pollutant Sampler(VAPS), and 4) Anderson Model SA 241 Dichotomous Sampler (Dichot). Three different TEOM instruments were used to provide integrated 1-hour mass measurements for particles. The first two were used to collect PM2.5 and PM10 mass. The PM2.5 TEOM was fitted with a University Research Glassware (URG) cyclone inlet and the PM10 TEOM was fitted with an Anderson Model 246B PM10 inlet. The third TEOM was used to conduct three special studies: to determine background noise using blank filters

(Feb. 7-Feb. 28), to collect PM2.5 with case temperature at 40° C instead of the prescribed 30° C (May 18-Aug. 24), and to collect PM1 (0-1.0 μm) mass (Feb. 28-May 17 and Aug. 8 -Jan. 22). A URG PM2.5 or PM1 cyclone inlet was used in these studies. TEOM coarse mass was determined by subtracting TEOM PM2.5 from TEOM PM10 mass. It should be noted that under certain conditions of rapidly fluctuating humidity, negative measurements are possible at low concentrations for the TEOM [2]. Negative hourly values are considered valid and are used to calculate 24-hour averages. (Note: Due to low humidity in Phoenix, TEOMS were operated at 30° C rather than the recommended 50° C.) The DFPSS, which also employs a URG cyclone inlet identical to the PM2.5 TEOM, provided 24-hour integrated PM2.5 mass on both quartz and Teflon filters (quartz for carbon analysis, Teflon for mass and X-ray analysis). The VAPS is a dichotomous sampler employing a virtual impactor to provide a cut point of 2.5 μm. It provides two separate channels to collect the PM2.5 samples- one employing quartz filters for carbon data and the other employing Teflon filters for mass and X-ray determinations. Coarse particles were collected on the third channel using polycarbonate filters. During this study, the VAPS operated on a 24-hour sampling interval at 33 LPM using an Anderson Model 246B PM10 inlet (twice the flow rate for which the PM10 inlet was designed). The Dichot employs the Anderson PM10 inlet along with a virtual impactor and operates on a 24-hour interval at a flow rate of 16.7 LPM collecting particles on two channels- one for fine and the other for coarse particles. For both dichotomous samplers, PM10 mass is determined by adding the mass from the coarse and fine channels.

Method Comparisons

Since the DFPSS, VAPS and Dichot all measure on a 24-hour interval, the TEOM hourly mass measurements (along with accompanying meteorological data) were averaged over a 24-hour period beginning at 7:00 a.m. to correspond to the changing of filters on the other instruments. At least 20 hourly measurements were required for a daily average. The frequency of sampling varied with methods. The TEOM and DFPSS sampled daily while the VAPS and Dichot sampled every third day with the exception of some special study periods: between February 26 and March 4 the VAPS was operated for seven consecutive days but the Dichot was not operated; between March 11 and March 17 both the VAPS and Dichot were operated for six consecutive days; and between September 14 and September 18 the VAPS and Dichot were operated for five consecutive days, and Teflon filters were used on the VAPS coarse channel.

A time series plot of 24-hour fine particle mass ($\mu g/m^3$) for each of the four methods is shown in Figure 1 along with wind speed (m/sec) plotted as a solid line. This is a scatter plot showing relative daily concentrations and sampling frequencies over a year-long period. The most noteworthy thing about this plot is that beginning in October the concentrations of fine particles begin to increase to the highest levels of the year. Corresponding to this increase in concentration is a decrease in wind speed. It should be noted that the drop in wind speed is accompanied by a shift to a predominately easterly direction during this period. This increase in concentration is also characteristic of coarse and PM10 particles plotted over time shown in Figures 2 and 3. Detailed information on the distribution of particle mass is given in Table 1.

Box plots in Figure 4 display the distributional characteristics of measurements for each method

and for each particle size. The shaded areas contain 50% of the measurements between the upper and lower quartiles (25th and 75th percentile). About 99% of the measurements are contained between the T-bars that extend from each end of the box. The horizontal lines represent potential outliers. The middle of the notch is the median or 50th percentile and the edge of the notches represent upper and lower confidence intervals for the medians. Any two notches that do not overlap indicate that the two methods being compared are significantly different. Statistical comparisons were based on the difference (in logs) between daily pairs of measurements for two methods. The TEOM and DFPSS were determined to be statistically the same (within 1%) for fine mass but both measured significantly higher (15% to 17% higher) than either the VAPS and Dichot which were within 2% of each other for fine particles. Box plots for coarse and PM10 are also shown in Figure 4. None of the methods were equivalent for coarse or PM10 mass based on percent differences that ranged in absolute value between 26% for TEOM-versus- Dichot on PM10 to 90% for TEOM-versus-VAPS on coarse mass. The VAPS median in Figure 4 is noticeably lower for coarse mass than the other methods. This is probably the result of a lower cut point for the VAPS coarse fraction caused by operating at a higher flow rate (33 LPM) compared to the usual flow rate of 16.7 LPM normally used for a PM10 inlet [3]. Particle stability on polycarbonate filters was reported to be a problem due to static charges developed during sampling. Coarse particle mass is a factor of 2 to 3 times higher than the fine mass for TEOM and Dichot samplers. This is the reciprocal of coarse/fine ratios typically found at sites not located in desert regions [4,5]. On the other hand, the VAPS collected about equal amounts of coarse and fine mass with a ratio of .97 which is significantly lower than the other two samplers. Here again this may be attributed to the high rate of flow. However, this ratio changed to a factor of 2 for the 5-day special study where Teflon filters replaced polycarbonate filters on the coarse channel.

Particles with a size fraction less than or equal to 1.0 μm (PM1) were collected as part of a special study on a third TEOM. The distributional scatter of daily mass is plotted over time in Figure 5 along with PM2.5 mass. As with other particle sizes there is also a rise in PM1 mass during the fall months compared to measurements made in the spring and late summer. Figure 6 is a scatter plot of differences between logs of daily pairs of PM2.5 and PM1 plotted against their mean. Although there is more scatter at the lower concentrations, a fitted line shows that on the average PM2.5 mass is about 40% higher than PM1 over a range of from zero to approximately 40.0 $\mu g/m^3$.

Instrument Precision and Accuracy

The error of a measurement is simply the difference between the measurement and the actual (or true) value as expressed in the following linear model:

$$\text{Measurement} = \text{True value} + \text{Measurement Error.}$$

The average of the measurement errors for repeated measurements at a given true value is a measure of the instrument bias or systematic error. The smaller the bias, the more accurate the instrument is considered to be. Although absolute bias cannot be estimated without knowing the true value, it can usually be controlled by proper calibration and therefore is not considered a serious problem. On the other hand, measurement error fluctuates around the bias in a random manner that is more serious and

difficult to control (or at least minimize) since this fluctuation can result from the combination of several different external influences and environmental conditions (e.g. weather, operators, handling, etc.) . The standard deviation of the measurement error is a measure of the precision of the instrument. Precision (or imprecision) is simply a measure of how well an instrument can repeat itself under the same conditions. For ambient instruments this is a question of how well two or more instruments of the same type - operating simultaneously in close proximity - can replicate each other over several days of sampling. The total uncertainty associated with a single daily measurement is a combination of the day-to-day variation between true values plus instrument precision. Therefore, it is important that the instrument precision be many times smaller (an order of magnitude as a rule of thumb) than the day-to-day variation in the true concentrations in order to have high efficiency in the measurement process [6]. Precision also determines the sensitivity of an instrument by controlling how quickly it responds to changes in daily concentration levels. None of the instruments used in this study were duplicated for the purpose of estimating precision. Nevertheless, estimates of precision for each individual instrument were determined using multivariate techniques on the available data [7]. For two instruments, this basically involved subtracting the covariance between paired measurements (which is an estimate of the variance of the true values) from the variance of the daily measurements for each separate instrument and taking the square root. For more than two instruments, calculations were slightly more involved [7]. The underlying assumption is that different instruments do replicate each other since they simultaneously measure the same true unknown daily concentration but with possibly different bias as well as different precision. This possibility exists even for duplicate instruments, but usually only the bias is hypothesized to be different while the precision is assumed to be same for both instruments. The average difference between daily pairs of measurements for any two instruments is an estimate of the difference in bias. The percent differences discussed in the previous section are a measure of bias between any two instruments relative to their average. The more precise the instruments are, the better we are able to detect small differences in bias between the instruments. Table 2 gives an estimate of precision for each different instrument and for each particle size. The original measurements were transformed into logarithms prior to analysis. Therefore, the estimates of precision are expressed as a percent of the concentration in the original units (usually referred to as the percent coefficient of variation). For example, the precision of the TEOM for a single 24-hour measurement was estimated to be $\pm 28\%$. This implies that about one third of the true values are measured with error greater than 28% of the concentration level. It also implies that if two TEOMs with the same precision were operated side-by-side then about half of the daily pairs would differ by more than $.95(28\%) = 27\%$ and, about one time out of 20, pairs would differ by as much as $2.77(28\%) = 78\%$ due to random error even if they had the same bias. Precision for the other instruments can be summarized in a similar way.

Background noise for the TEOM was estimated using blank filters to be $\pm .5\ \mu g/m^3$ (1-sigma) for 24-hour measurements. Since there is such a small probability of background noise contributing more than $1.5\ \mu g/m^3$ (i.e. 3-sigma) to a measurement, anything above this value must be a real quantity. According to the ACSCEI [9], $1.5\ \mu g/m^3$ would constitute a limit of detection (LOD) for TEOM 24-hour averages.

The standard deviation of the true values for this study was estimated to be $\pm 48\%$ (of the mean concentration level) for fine particles, and $\pm 40\%$ for coarse and PM10 particles. Therefore, in the

absence of measurement error, nearly all 24-hour fine particle concentrations would fall between zero and 30 $\mu g/m^3$ assuming that the overall mean is about 14 $\mu g/m^3$. For the TEOM, instrument imprecision accounts for about 25% (i.e. $.28^2/(.48^2+.28^2)$) of the total variation of a single 24-hour measurement. Taking into account instrument precision widens the interval to (-5, 33) $\mu g/m^3$ allowing for negative TEOM measurements. This agrees with the percentiles of the actual measurements shown in Table 1 for the TEOM. This increase in the upper limit is an example of how imprecision can lead to overestimation of peak values which may result in falsely claiming noncompliance with a standard when the true value is actually below the standard [10,11]. One way to reduce the effects of instrument error is through the diligent use of quality control procedures. Another way would be to deploy several instruments of the same type at a given site and use the average of the measurements. For example, the instrument precision for the VAPS ($\pm$6%) contributes only 2% to the overall variation in a single daily PM2.5 measurement. On the other hand, the imprecision of the TEOM ($\pm$28%) contributes about 25% to the overall variation. The ratio of the precision estimates of two different instruments is a measure of their relative efficiency. Based on the estimates of precision from this study, the average of twenty-two TEOMs would be required in order to be as efficient as the VAPS. This is impractical as well as expensive but it does illustrate the importance of instrument efficiency in achieving high quality data.

As previously mentioned, when duplicate instruments are used, the precision is usually assumed to be the same for both instruments. In this case, the error variance of a single instrument is estimated as half the variance of the difference between paired values. This may not always be a safe assumption even when the instruments are the same type since one instrument may have a significantly higher precision than the other. For example, except for the temperature levels at which the instruments were operated, the TEOM at 40° C may be considered a duplicate for the TEOM at 30° C for 24-hour averages. The precision for the TEOM (30°) was estimated to be $\pm$24%, and, for the TEOM (40°) , the precision was so low as to be statistically indistinguishable from zero. Assuming they have the same precision, the estimate for each instrument is $\pm$17%.

A final note regarding precision as it relates to linear regression: one of the necessary assumptions for regression is that the dependent variable be measured without error. Not accounting for error in the measurement will cause underestimations of the regression coefficient onto the true concentrations often referred to as the structural regression coefficient [12]. The magnitude of the underestimation depends on the imprecision of the measuring instrument being used. In this study, a precision of less than 10% reduces the regression coefficient by 4% for fine particles, but an imprecision of 28% reduces the estimate by as much as 1/4. This could seriously affect the estimates of mortality as it relates to particle loadings: the greater the imprecision, the lower the estimate of mortality for a given true concentration. Adjustments in regression coefficients due to error would help to normalize results for comparisons across sites where instruments measure with different precision.

Hourly TEOM Measurements

One of the unique things about the TEOM is the ability to collect particle data on a continuous basis which makes it possible to examine diurnal patterns. Hourly values of TEOM fine particles are plotted as a time series in Figure 7. The specific humidity - a measure of the water content in the air in units of grams H_2O per kilograms of moist air-is also plotted. During the time between Julian dates of 225

to 235 corresponding to calendar dates of mid to late August there is a noticeable scatter in the hourly fine fraction. This corresponds to the time when there is relatively high water content (>7.0 g/kg) in the air suggesting that the TEOM is unstable under such conditions. The operating temperature of the TEOM was 30° C. During this period of time a special study was being conducted with another TEOM sampling with a temperature of 40° C. Values from both instruments are plotted in Figure 8. Although there are spikes in the readings of both instruments corresponding to high values of specific humidity during this time period, the TEOM at 40° C appears more stable with spikes about 1/5 as great as the TEOM at 30° C .

The relative instability of the TEOMs is even more noticeable in Figure 9 where the differences in hourly fine particle mass between the TEOMs are plotted against the specific humidity. These differences are clustered about a fitted line (solid) at zero (broken line). The scatter about this line is extremely tight up to about 7 g/kg. About 75% of all hourly measurements were taken when the specific humidity was below 7 g/kg. In this range both monitors have an average concentration of 10.2 $\mu g/m^3$ with approximately 99% of the measurements for each monitor between limits (-9, 27) $\mu g/m^3$. Above 7 g/kg specific humidity this spread in hourly fine mass stays about the same for the TEOM at 40° C but the width of the interval increases to (-25, 47) $\mu g/m^3$ for the TEOM at 30° C. Above 12 g/kg specific humidity this spread increases to (-49, 64) $\mu g/m^3$ even though the average for both instruments remains approximately 10 $\mu g/m^3$. It appears that the higher temperature reduces the scatter in hourly measurements by a factor of 3 when the specific humidity is above 12 g/kg. Even with this nonhomogeneity in scatter, the averages remain comparable across the entire range of concentrations regardless of the water content. This comparability between averages was also evident for 24-hour values involving the TEOM and DFPSS where the difference was within 1%.

As was demonstrated above, diurnal patterns can be highly variable from hour to hour and across days. By averaging over all days for a given hour we are able to smooth the diurnal plots. These patterns for all particle sizes along with wind speed are shown in Figure 10. Peak levels occur around 7:00 to 8:00 a.m. and 10:00 to 11:00 p.m. with lowest levels generally occurring about 4:00 p.m.. Wind speeds are maximum in the afternoon causing dilution in the mass concentrations. When these plots are done by season, the diurnal pattern does not change; however, the vertical scales change to reflect a slowdown in wind speed during the fall and winter months and an increase in mass concentrations during this same period.

Conclusions

The results of this pilot study have great value in providing direction and expectations for future work in the National PM Research Monitoring Network. Comparisons between methods reported in this study form a baseline for future comparisons under similar conditions especially for fine particle mass. Monitors equipped with cyclones measured significantly higher concentrations than the dichotomous samplers for the same particle size. It should be pointed out that differences may be due in part to conditions under which the samplers were operated. The dichotomous samplers were entirely outside the sampling trailer while the TEOMs and DFPSS only had the inlets outside. Therefore it is possible that ambient temperature may have affected the observed differences in mass rather than the type of inlets. Measurement precision was estimated without benefit of duplicate samplers by using a model that takes

advantage of simultaneous readings from all instruments for each particle size. Verification of this procedure would require that duplicate samplers be operated under similar conditions; however, there is no guarantee that instruments of the same type have the same precision. For fine particles the VAPS and DFPSS had the best precision followed by the Dichot and TEOM. If the degree of imprecision relative to the total variability in a measurement is not taken into account, false conclusions concerning compliance testing or the statistical association between particle mass and mortality in urban areas could result. The fluctuations in TEOM hourly measurements caused by water in the atmosphere is effectively canceled by daily averaging. Raising the operating temperature in the housing to 40° C also reduces the instability in hourly measurements due to water (>7 g/kg) in the air. Peaks and valleys in daily concentration levels for all particle sizes appear to be seasonal and are inversely related to seasonal wind speed patterns. This is examined in more detail in Part II [13]. Diurnal patterns for all particle sizes show a similar behavior with respect to hourly wind speed with peak concentrations occurring in the morning and late evening regardless of the time of year.

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Table 1. Distributional statistics for 24-hr particle mass ($\mu g/m^3$).

		N	Min	5%	25%	50%	75%	95%	Max	Mean	Std Dev
	TEOM	335	-5.9	3.4	8.0	11.6	18.0	29.6	40.2	13.6	8.1
	DFPSS	284	2.1	5.1	8.0	10.6	16.0	27.5	37.4	12.9	7.0
Fine	VAPS	99	3.4	4.7	6.5	8.2	12.5	23.8	26.2	10.2	5.5
	Dichot	81	.6	4.4	6.8	8.6	12.3	24.8	27.3	10.9	6.4
	TEOM	326	-4.0	12.4	21.0	28.1	38.8	60.8	103.7	31.3	15.5
Coarse	VAPS	101	-0.2	3.8	5.4	8.0	11.9	26.1	43.4	10.2	7.7
	Dichot	81	3.9	9.1	16.6	23.3	28.8	48.9	81.4	24.7	13.1
	TEOM	327	-1.8	18.0	28.7	39.9	55.8	86.7	129.1	44.6	22.0
PM10	VAPS	99	6.9	9.0	12.3	16.3	23.1	52.2	67.2	20.4	12.5
	Dichot	81	11.3	15.0	22.9	32.0	40.8	70.8	104.6	35.8	18.5

Table 2. Precision of individual 24-hr measurements.

	TEOM	DFPSS	VAPS	Dichot
Fine	± 28%	±8%	±6%	±17%
Coarse	±18%		± 44%	±21%
PM10	±17%		± 26%	±14%

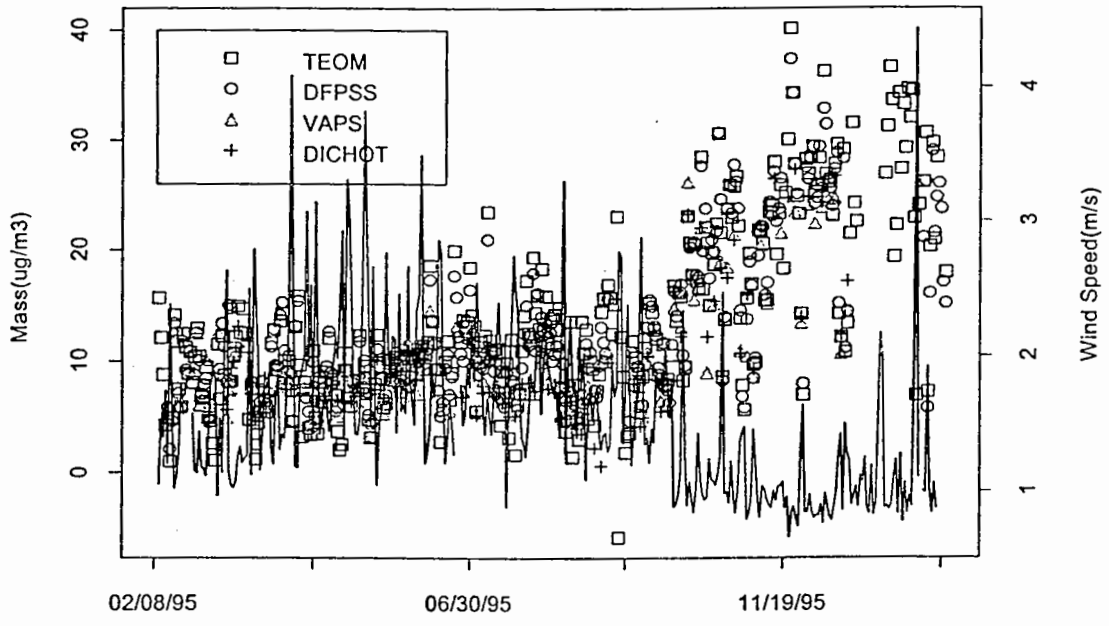


Figure 1. Time series of fine particle mass

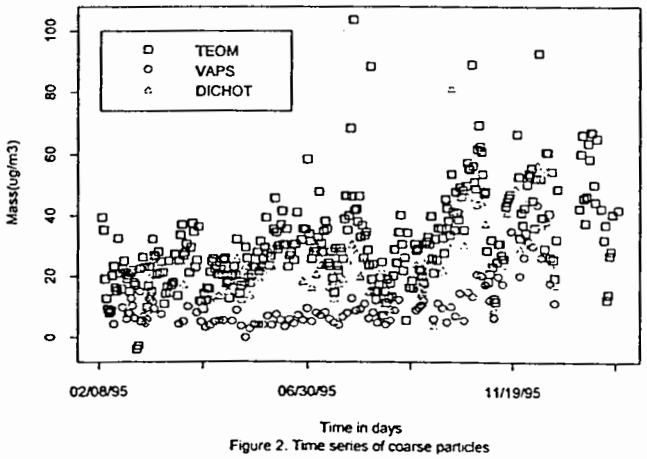


Figure 2. Time series of coarse particles

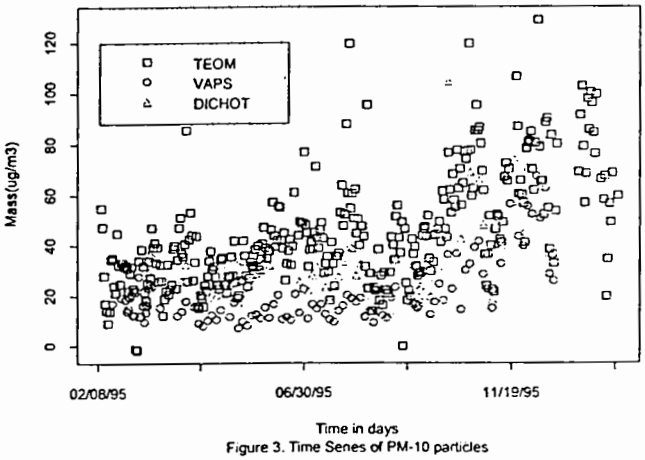


Figure 3. Time Series of PM-10 particles

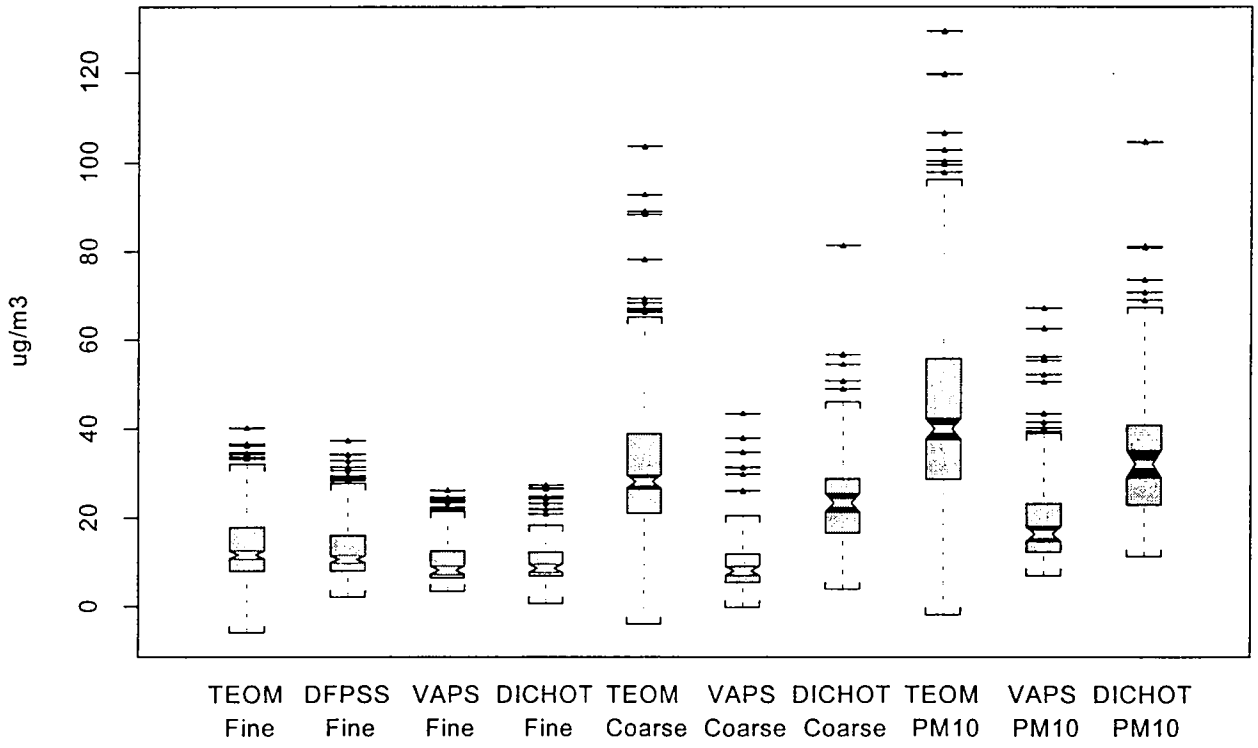


Figure 4. Box plots of particle mass

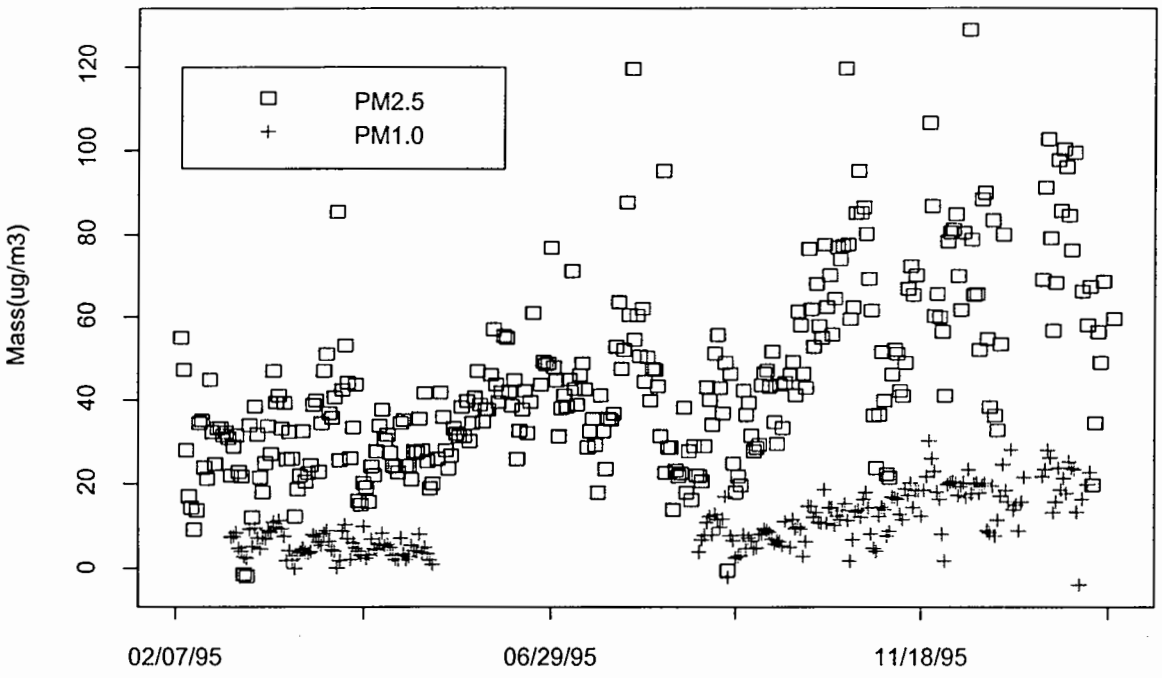


Figure 5. Time series of TEOM fine particles

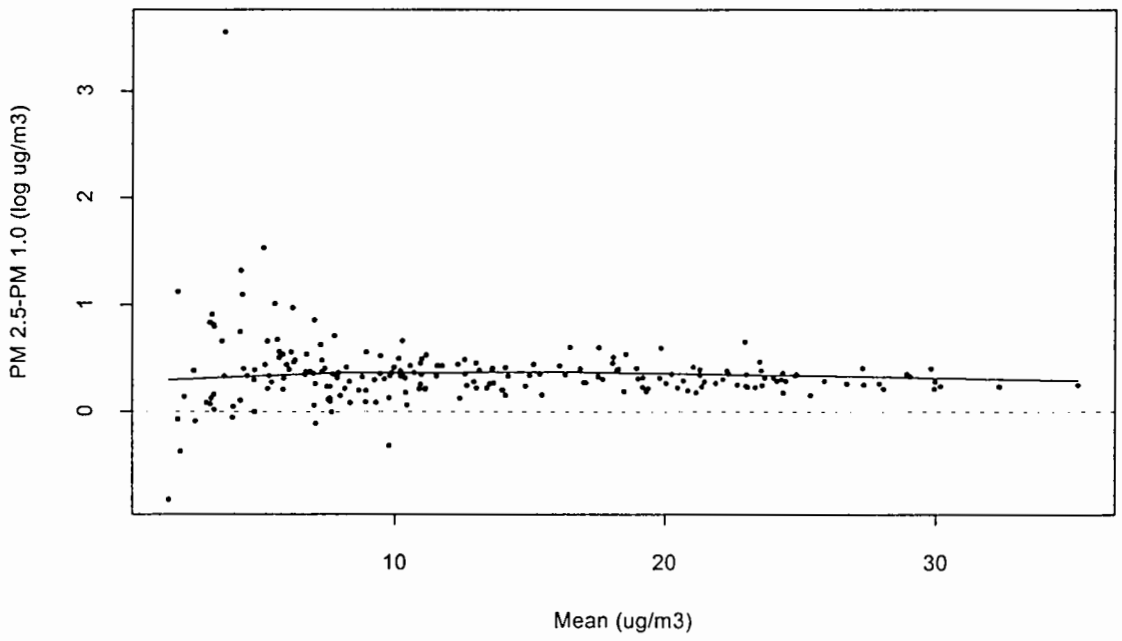
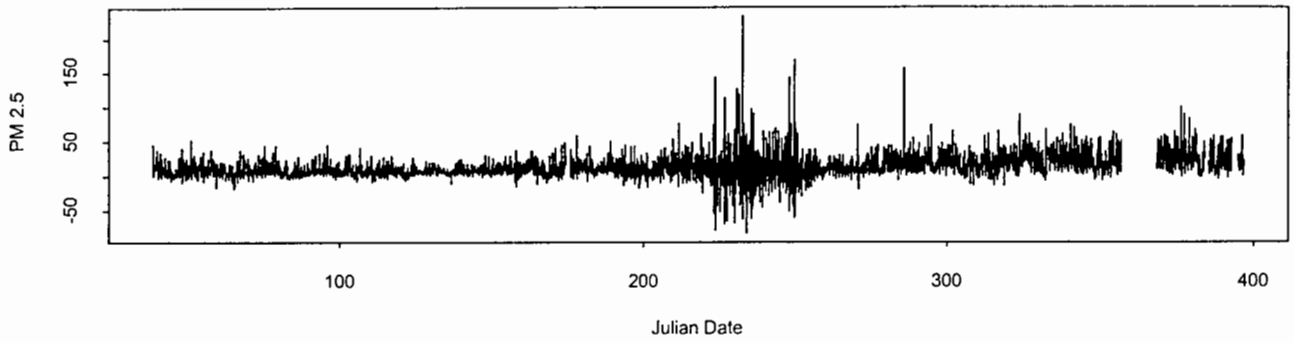


Figure 6. Paired Differences in logs between PM25 and PM1 plotted against means



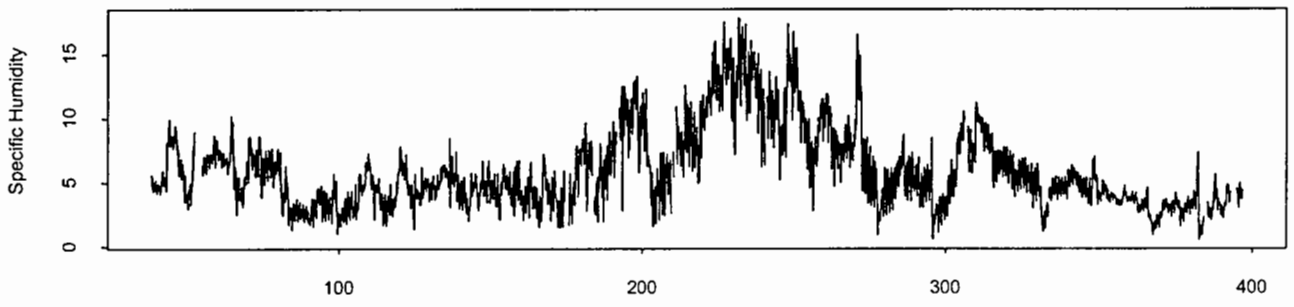
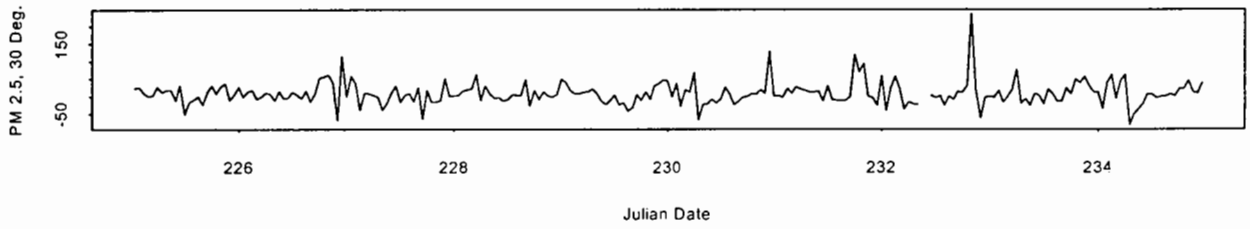
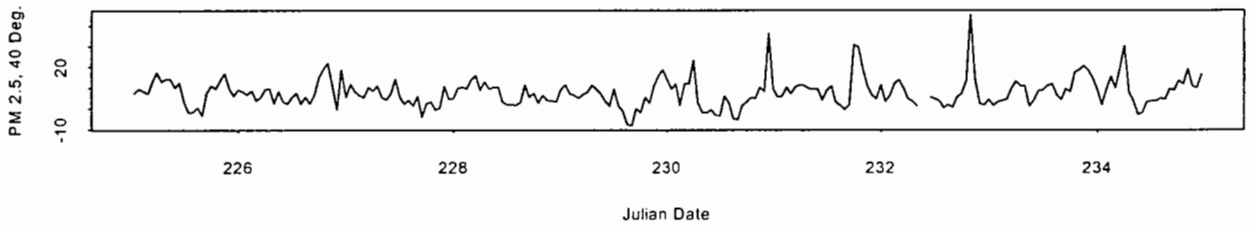


Figure 7. Hourly values of TEOM fine particles (ug/m3) and specific humidity (g/kg)





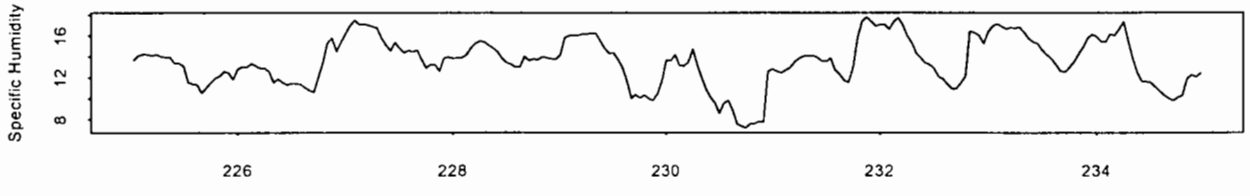


Figure 8. Hourly values of TEOM fine particles (ug/m3) and specific humidity (g/kg)

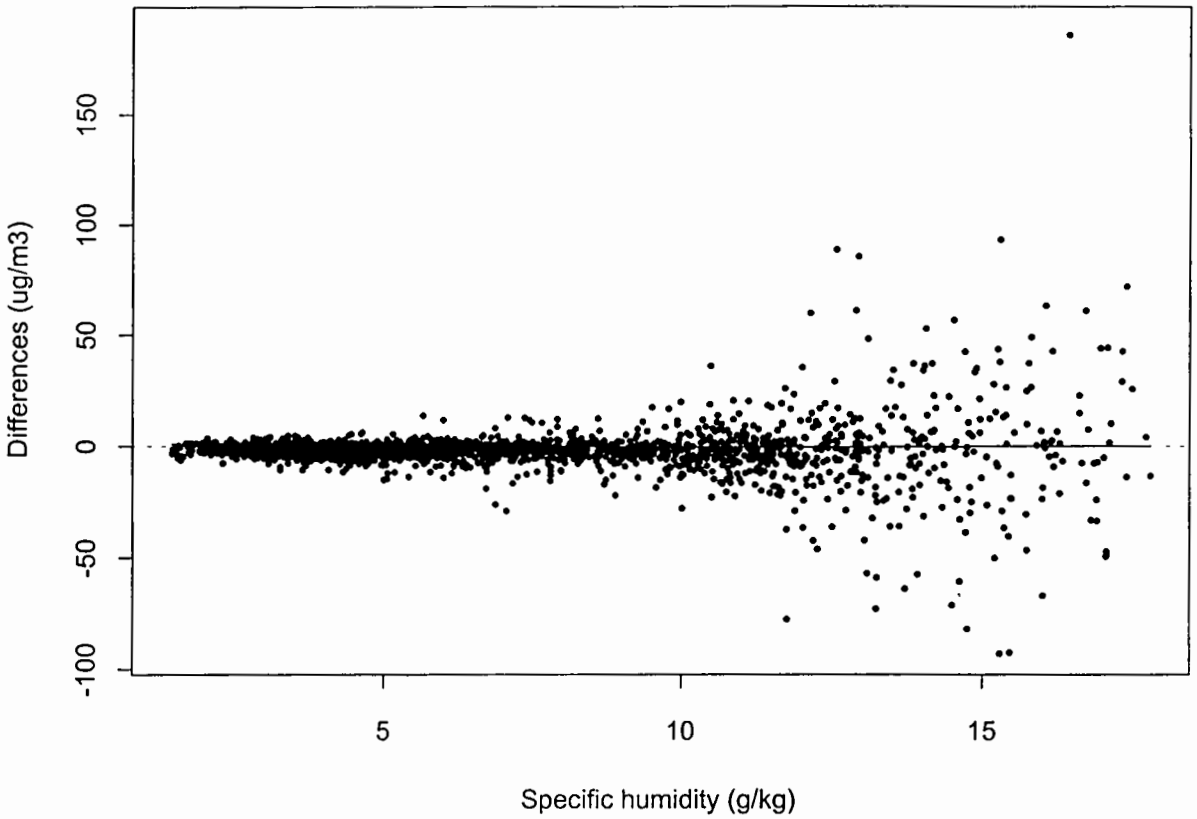


Figure 9. Plot of fine particle differences between TEOMS operated at 30 and 40 degrees C

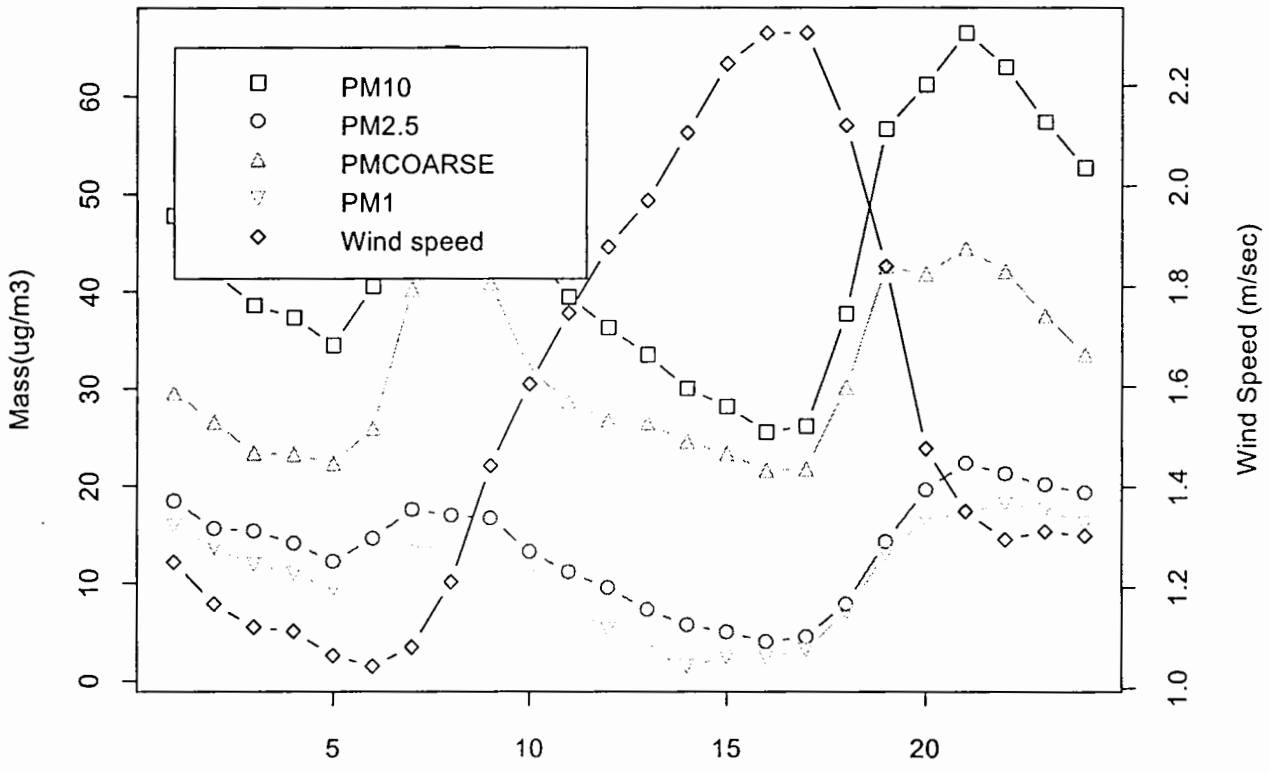


Figure 10. TEOM diurnal plots

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15. SUPPLEMENTARY NOTES

16. ABSTRACT

As a result of recent findings of statistical relationships between ambient particulate matter concentrations and mortality, the EPA's Office of Research and Development has begun to establish monitoring sites to collect data which have more detail (in terms of size cuts and composition) than those used in previous epidemiological studies. The Phoenix site was designed to evaluate various particulate samplers in an area that is expected to be heavily influenced by a component of windblown dust.

This paper examines one year (February 1995-January 1996) of particulate mass data from that site. The data include 24-hr fine(PM2.5) and coarse(PM2.5 to PM10) mass fractions from three different samplers. In addition, 1-hr PM2.5 and PM10 measurements are available from continuous methods and are supported by 1-hr meteorological data. This paper also addresses method comparisons, precision and accuracy of the instruments, and characterization of the relation between fine and coarse mass. Seasonal and meteorological influences on the distributions of mass are explored. Special studies on the reliability of the continuous methods are examined.

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