Section 2, Chapter 1
PARTICULATE MATTER MEASUREMENTS

INTRODUCTION

Fine particulate matter, or $PM_{2.5}$, refers to a mixture of solid and liquid atmospheric particles with an aerodynamic diameter (d_{ae}) less than 2.5 micrometers. It arises mainly from anthropogenic sources such as fossil fuel combustion by electric utilities and motor vehicles, wood burning, and the smelting or other processing of metals. $PM_{2.5}$ consists of sulfate, elementary carbon, nitrate, ammonium, organic compounds, trace elements, elemental carbon, and water.[1] The majority of $PM_{2.5}$ components are secondary materials, derived from the chemical reactions of gaseous precursors such as SO_2, NO_x, volatile organic compounds (VOCs), organic and elemental carbon, and a range of trace metals.

In 1997, the U.S. Environmental Protection Agency (USEPA) established a standard for $PM_{2.5}$ of 15 ug/m^3 for an annual arithmetic mean and 65 ug/m^3 for a 24-hour average concentration based on assessments of three consecutive years.[2] USEPA based this standard on an extensive review of numerous epidemiological studies. Recent studies suggest that $PM_{2.5}$ is more strongly correlated with adverse health effects than particles in other size ranges.[3] These health effects range from slight respiratory symptoms to increased mortality rates. In a study of six eastern U.S. cities, Schwartz et al. reported a 10 μg/m^3 increase in the two-day mean $PM_{2.5}$ concentration associated with a 1.5 percent increase in total daily mortality.[4] Certain population groups such as seniors, respiratory and cardiovascular patients, and children are most susceptible to particle pollution.

The association between ambient $PM_{2.5}$ concentrations and a variety of adverse health outcomes suggest that ambient concentration may be an indicator for personal $PM_{2.5}$ exposure, and ambient $PM_{2.5}$ should correlate with indoor and personal $PM_{2.5}$ concentrations.[5] However, epidemiological study results are questionable due to concerns involving inconsistent correlations between outdoor, indoor, and personal levels.[6] Numerous factors exist that can affect relationships including: spatial variability of outdoor concentrations, classification of $PM_{2.5}$ exposure into different microenvironments, the role of chemical components, influence of co-pollutants, and meteorological confounders.

Spatial Variability of Ambient $PM_{2.5}$ Concentrations

Generally, the ambient $PM_{2.5}$ concentration measured by a fixed central monitoring site is used as a proxy for individual $PM_{2.5}$ exposure in epidemiological studies.[7] For uniformly distributed pollutants such as secondary pollutants, a fixed-site monitor provides applicable data in estimating exposures for regional air pollution.[8]

Fine particles have low deposition rates and long atmospheric residence time (days or weeks) resulting in transportation over thousands of kilometers. Consequently, the distribution of ambient $PM_{2.5}$ tends to be homogeneous over urban areas and large regions. Several studies conducted in the U.S. and Europe have reported a uniform distribution of ambient $PM_{2.5}$ across multiple sites over large geographic areas.

Classification of $PM_{2.5}$ Exposures

Total personal exposure to $PM_{2.5}$ is the result of encounters with $PM_{2.5}$ in a variety of microenvironments. The $PM_{2.5}$ concentrations in these microenvironments are a combination of ambient and non-ambient sources. The nonambient $PM_{2.5}$ consists of indoor-generated $PM_{2.5}$ and personal-activity $PM_{2.5}$.

Separating exposure into ambient and non-ambient $PM_{2.5}$ is necessary due to their differences in sources, compositions and driving forces. The formation of ambient $PM_{2.5}$ depends on the photochemical smog processes, which increases with sunlight and temperature but is reduced dramatically indoors. Emission rate changes from mobile sources, utilities, and industrial processes, and meteorological conditions influence ambient $PM_{2.5}$.

Indoor $PM_{2.5}$ includes ambient outdoor $PM_{2.5}$ that has infiltrated indoors, and indoor-generated $PM_{2.5}$. Human activities, cooking, household appliances, and room ventilation conditions affect indoor $PM_{2.5}$ concentrations.[9] The mean indoor/outdoor (I/O) concentration ratio can increase dramatically during the day but decreases at night when human activities are reduced.[10] Three major studies evaluating indoor and outdoor $PM_{2.5}$ show increases of $PM_{2.5}$

concentrations ranging from 25-45 ug/m^3 due to smoking.[11] In the six-city and New York State studies, mean indoor concentrations were double the outdoor levels.[12] As a result of multiple sources, indoor $PM_{2.5}$ is often higher than ambient concentrations.

Personal $PM_{2.5}$ is measured by personal exposure monitors worn by subjects. It can also be obtained by integrating the time-weighted concentration of different microenvironments. Several studies reported increased personal exposure compared to either indoor or outdoor concentrations with exceptions of some studies of elderly or disabled subjects.[13] The increased personal exposure compared to indoor or outdoor concentrations is called personal-activity $PM_{2.5}$ or "personal cloud."[14] Studies also found that the personal cloud increases during the day when personal activity was high.[15] The origin of personal cloud is still unknown.[16]

According to USEPA, U.S. citizens spend about 90 percent of their time inside, and only about 6 percent outdoors.[17] Because a large proportion of personal exposure occurs indoors, indoor $PM_{2.5}$ is a great contributor to personal $PM_{2.5}$ exposure.[18] In a study of 10 subjects with chronic obstructive pulmonary disease (COPD), Bahadori found personal concentrations more closely correlated to the indoor levels (a Pearson's R= 0.72) than to the outdoor levels (a Spearman's R= 0.30).[19]

Correlations of Personal and Outdoor $PM_{2.5}$ Concentrations

Historically, correlations of ambient outdoor and personal $PM_{2.5}$ concentrations vary considerably with correlation coefficients (R) ranging from near zero to near one, which is due to the influence of non-ambient PM regardless the difference in study designs.[20]

The influence of indoor $PM_{2.5}$ provides a plausible explanation for the varying correlations between personal and outdoor $PM_{2.5}$ in epidemiological studies.[21] Relatively strong correlations result for subjects with limited nonambient $PM_{2.5}$ sources.[22] This trend is more obvious in a longtitude-designed study.[23] Longitudinal studies, tracking correlations on individuals over time, illustrate better correlations than studies with cross-sectional designs. [24] Strong correlations occur on subjects with limited nonambient sources due to their sedentary lifestyles, or no apparent indoor source exposure such as environmental tobacco smoke (ETS) exposures.[25] For example, studies in the 13 primary school children in the Netherlands documented that the median R-value for personal-outdoor correlation improved from 0.86 to 0.92 when children with smoking parents were excluded from the analysis.[26]

High correlations also occur for subjects who spend more time in outdoor microenvironments, or live in micronenvironments with high air exchange rates.[27] Indoor $PM_{2.5}$ concentrations decrease as air exchange rates or ventilation conditions improve.[28] In examining the impacts of ventilation

conditions, Sarnat et al. reported that the coefficient of determination (R^2) of personal and ambient concentrations in well ventilated, moderately ventilated, and poorly ventilated indoor environments were 0.88, 0.59, and 0.25 respectively.[29] In summary, correlation variations found in PM studies actually reflect the impact of individual lifestyles and the conditions of the microenvironments in which the subjects spend time.[30]

RESEARCH METHODOLOGY

This study was conducted in central and southeastern Ohio (Figure II.1.1) from January 1999 through August 2000. Fourth and fifth grade classes in two schools in Columbus (Koebel Elementary School and New Albany Elementary School) and one school in Athens (East Elementary School) comprised the monitoring sites. Approximately 30 students at each site were involved.

Figure II.1.1. Overview of Study Area

The three elementary schools are in residential neighborhoods. The location of these sites is illustrated in Figure II.1.2. Koebel is located on the south side of Columbus in the industrial center of the city. The industrial activities include foundries, plastic facilities, and gravel/quarrying operations. This site is also located within 0.5 km of a major transportation artery. New Albany is approximately 8 km northeast of downtown Columbus and is approximately 32 km northeast of Koebel. New Albany is a bedroom community of Columbus with few commercial facilities and no significant industrial operations within the municipal boundary. Since the prevailing winds are from the southwest, transport of $PM_{2.5}$ precursors from the Columbus area may influence the particle pollution at this site.

The third site, Athens, is approximately 120 km southeast of Columbus and is a rural location. Athens is a university town with a population of 20,000. The site is in a residential area and the only significant local stationary pollution source is Ohio University's coal-fired power plant. Athens is about 32 km west of the Ohio River Valley, which has numerous coal-fired power generation facilities, chemical manufacturing facilities, and industrial operations. Athens is an upwind remote site for the Department of Energy's Ohio River Valley $PM_{2.5}$ monitoring projects.[31]

The Koebel school is a one-story building while the New Albany and Athens schools are two-story buildings. Classrooms at each elementary school used for indoor monitoring were selected as far as possible from the kitchen facilities to reduce the impact of cooking-generated $PM_{2.5}$. The classrooms at Athens and New Albany are air-conditioned. Koebel elementary school has a central heating system but no central air conditioning system. All three schools use natural ventilation during the warm months, so classroom windows are typically open during the months of April, May, June, September, and part of October. With central air conditioning, Athens and New Albany may close their windows during very warm days. However, windows are open a majority of the school days during the spring and fall.

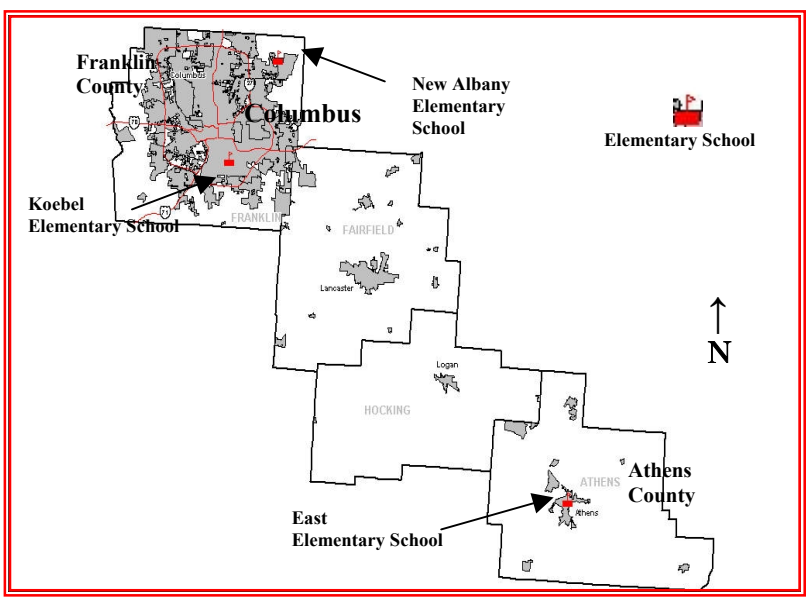


Figure II.1. 2. Locations of the three sites involved in the study.

Measurement Methods

$PM_{2.5}$ data were collected from January 1999 through August of 2000. Filter-based ambient, indoor, personal $PM_{2.5,}$ and continuous ambient $PM_{2.5}$

were measured concurrently at all three sites. The monitoring scheme is outlined in Table II.1.1.

Table II.1.1. Summary of the sampling time and samplers used.

Sampling Type	Sample Sampling Period	Schedule Sampling time	Monitor	Vendor
Continuous Ambient $PM_{2.5}$	Jan, 99 ~Aug, 00	Daily-24 h	TEOM® Series 1400a	Rupprecht & Patashnick (R&P) Co., Inc.
Filter Based Ambient $PM_{2.5}$	Jan~Jun, 1999	School Day- 8 a.m. – 3 p.m.	ACCU™ System	
	Sep, 99~ Aug, 00	Daily-24 h		
Filter Based Indoor $PM_{2.5}$	Jan, 99~Jun, 00	School Day - 8 a.m. – 3 p.m.	URG-2000-30EH URG-3000-02Q	University Research Glass (URG) Corporation
	Jun, 00~Aug, 00	Non-school day Daily-24 h		
Filter Based Personal $PM_{2.5}$	Jan, 99~Aug, 00	School Day - 8 a.m. – 3 p.m.	URG-2000-25F	

Ambient $PM_{2.5}$ Measurements

Continuous ambient $PM_{2.5}$ concentrations were measured using Tapered Element Oscillating Microbalances (TEOMs) series 1400a equipped with 2.5-µm inlets (URG-2000-30EH). Produced by Rupprecht & Patashnick (R&P), TEOM usage occurs widely in PM monitoring around the world.[32] Airflow through a cyclone at 16.67 L/min is isokinetically split into a 3 L/min main flow for the continuous ambient $PM_{2.5}$ sample, and a 13.67 L/min bypass flow. TEOMs were operated at 50 °C to reduce humidity, which might lead to some loss of semi-volatile materials.[33] The 3 L/min air stream passes through a Teflon-coated borosilicate glass fiber filter, which is on the narrow end of a hollow tapered tube. The frequency of the tapered element changes according to filter mass changes under the control of an electronic circuit. A precision electronic counter senses this frequency in a 2-second period and computes the mass of the particulate collected on the filter. The TEOMs were set to run 24 hours seven days a week.

Filter-based outdoor $PM_{2.5}$ concentrations were measured from an Automatic Cartridge Collection Unit (ACCU) System, which was connected to the TEOMs. The ACCU system redirects the bypass flow through one of eight flow channels that are controlled by the TEOM software. Sample media included 47 mm Whatman Teflon® filters (2-µm pores size), which were kept in standard 47 mm Teflon filter holders, at 13.67 L/min. The ACCU $PM_{2.5}$ were measured 24 hours Monday through Friday.

The TEOMs at Athens and Koebel were located on the roofs of the buildings used for the indoor $PM_{2.5}$ monitoring. The inlets for the monitors were within a 10-meter distance above the ground. At New Albany, the TEOMs were on top of a high school building approximately 200 meters from the site of the indoor monitoring. The monitor's inlet was within 13 meters of the ground. In addition, all three TEOMs were in secure limited access locations. A typical set-up of the ambient monitoring system is shown in Figure II.1.3.

Indoor $PM_{2.5}$ Measurements

Indoor monitors were operated at 10 L/min using flow-controlled indoor sampling pumps (Model 3000-02Q, URG). Measurements of indoor $PM_{2.5}$ concentrations were made using 2.5 µm cyclones (URG-2000-30EH). The inlets were specifically chosen to match the TEOMs & ACCU system inlets. The inlets were approximately 1.2 m above the floor. Whatman 37 mm Teflon filters with 2-µm pores size were held in 37 mm Teflon holders. Indoor monitors were timed to run from 8:00 a. m. to 3:00 p. m. from Monday to Friday throughout the school year.

Figure II.1. 3. A typical set-up of the ambient monitoring system.

Continuous ambient 24-hr $PM_{2.5}$ concentrations and filter based outdoor $PM_{2.5}$ concentrations were measured with TEOMs (middle) and ACCU system

(right) respectively from the roofs of the buildings where indoor were measured. Weather monitors (left) were set side-by-side with the $PM_{2.5}$ monitors.

Personal $PM_{2.5}$ Measurements

Personal $PM_{2.5}$ measurements were conducted using impactors (URG-2000-25F) with a $PM_{2.5}$ cut point. The inlets were placed within 0.25 m meters of the breathing zone directly below the shoulders of the subject minimizing the impact of expired air as a potential $PM_{2.5}$ source. Air sampling pumps (Model AirPro® 6000D, BIOS International Co.) were operated at a flow rate of 5 L/min and resided in an acoustic shell to reduce pump noise levels (Figure II.1.4). During non-sampling times, personal pumps were charged using direct plug-in converters. The personal $PM_{2.5}$ sampler devices were placed in personal backpacks (Camelback, Hydrate or Die®, 100oz. H.A.W.G.) that were worn by selected students during the school day (Figure II.1.4). Students were instructed to wear the sampling system throughout the sampling period. They were allowed to place the sampler nearby during indoor sedentary activities (i.e., reading, writing) or activities during which wearing the sampler would be inconvenient or impossible (i.e., sporting activities). The $PM_{2.5}$ samples were collected on Whatman 37 mm Teflon filters with 2-µm pores size, which were held in 37 mm Teflon holders.

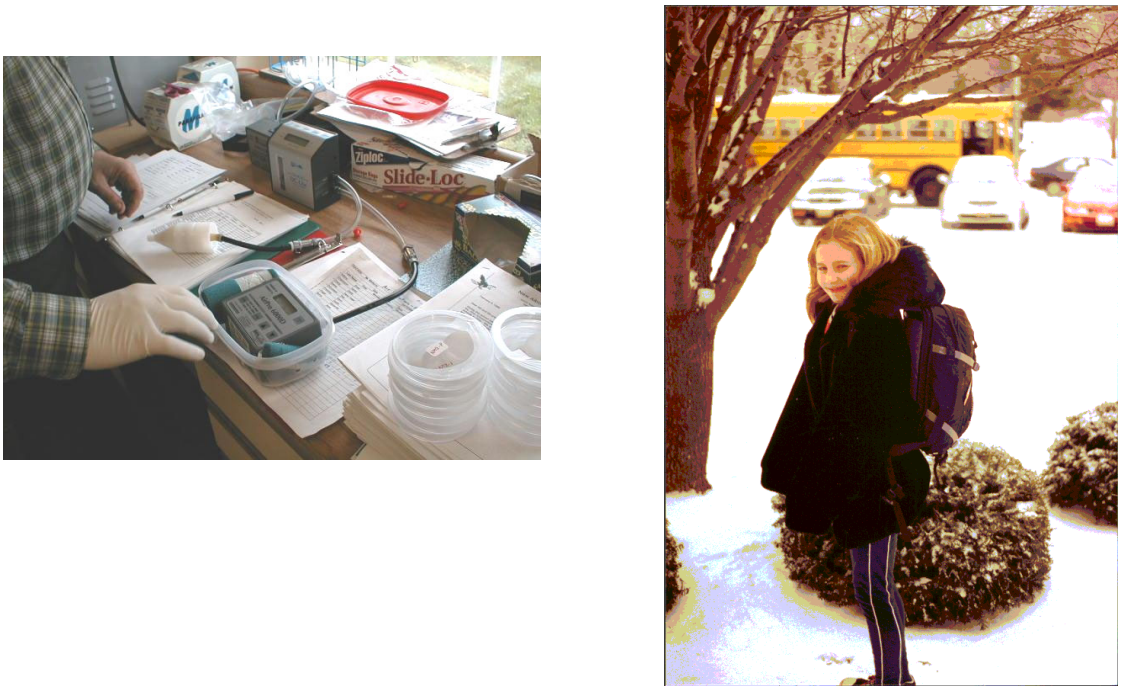


Figure II.1.4. Personal Samplers. Personal pump was placed in an acoustic shell (left) to reduce pump noise levels. Personal sampler devices were then placed in a personal backpack and could be carried by subjects (right).

Meteorological Monitoring

A MetOne AutoMET Meteorological Monitoring System station (Model 460 A, Serial U3439, Met One Instruments) was adjacent to the ambient $PM_{2.5}$ monitors at each site. Using Weather View 32 software, the weather stations recorded data including wind direction, wind speed, precipitation, barometric pressure, and solar radiation.

Gravimetric Measurements

Gravimetric measurements were conducted at Ohio University's Air Quality Laboratory. Filters were weighed in a temperature and humidity controlled microenvironment (environmentally controlled glove-box, PLAS-LABS, Figure II.1.5). As an alternative to the traditional weighing-room, the glove box provides an inexpensive, reliable, and convenient method for gravimetric measurements.[34] The specified temperature range was 20-23 ºC with a variability of no more than ±2 ºC over 24 hours, and 30-35% relative humidity with a variability of no more than ±5 percent over 24 hours.

Each filter was weighed in duplicate - both before and after sampling using a Sartorius analytical microbalance (MC5 UL), with a readability of 1 µg. When these two weights differ by more than 3 µg, the filter was reweighed until sequential weights agree within the specified range. The average of the two closest weights was the final weight.

Figure II.1.5. Humidity controlled microenvironment

Quality Control

In this study, $PM_{2.5}$ monitoring, filter handling and weighing protocols followed the standard quality assurance/quality control (QA/QC) procedures, which are on file at Ohio University. This section highlights the QA/QC procedures that were used during sampling (field procedures) and in the laboratory.

Field Procedures

For the purpose of chemical analysis, filter-based $PM_{2.5}$ mass measurements were collected concurrently with TEOM measurements. Ambient, indoor, and personal monitoring instruments were established in the same school building or at an adjacent building at each site. Prior to the study's commencement, all of the $PM_{2.5}$ monitoring instruments were calibrated. During the study period, trained field technicians visited each site weekly to collect data, transport samples, and perform quality control checks on the equipment.

During each site visit, continuous ambient $PM_{2.5}$ data were downloaded using a portable notebook computer. Filter-based samples were collected and transported in insulated coolers with frozen refrigerant gel packs to maintain a temperature below 4°C, which was monitored by a VWR brand® High/Low Memory Alarm thermometer. At this time, used personal impactors were also collected. New filters and cleaned personal impactors were deposited at the sites prior to the new sampling cycle. Quality control checks were also completed on the TEOMs, the ACCU systems, indoor, and personal monitoring instruments (on file at Ohio University).

One science teacher at each school was identified to support the monitoring program. The teachers attended a one-day training session at Ohio University prior to the initiation of the monitoring program to review quality control and monitoring protocols. The teachers were responsible for daily calibration of the personal pumps prior to and after the sampling period, setting up samples for the personal and indoor monitors, retrieving samples from the monitors, and maintaining sampling records.

Laboratory Procedures

Three sets of blank filters were used to insure the quality of the filter weighing and handling procedures. A lot blank was used to distinguish each batch of filters purchased. To test the stability of a filter batch, a randomly selected filter from each batch (lot blank) was weighed in a 5-day sucession. Only when the successive mass weighing of the lot blank was within 15 µg, was the new filter batch accepted. With each round of filters (i.e. two week sampling period) a lab blank and a field blank was also prepared. Lab blanks were used to assess the environmentally-controlled weighing chamber. Field blanks were used to evaluate the potential contamination during the processes of filter assembly, de-assembly, and transport.

Once the samples were preweighed, the lab blank remained in the controlled microenvironment. The field blank and field samples were placed into their sample cassettes inside an environmentally-controlled assembly hood (Captair Filtair®, 804C type, Captair Labx, Inc.). The assembly hood was cleaned with alcohol wipes (VWR Scientific Premoistened Clean-Wipes ™ ,

Alcohol/DI) prior to each assembly (and de-assembly) procedure. The filter cassettes were placed in individual petri dishes, parafilmed, double bagged, and transported to the morning sites.

After sampling, each filter was inspected before conditioning in the controlled microenvironment for post weighing. The filter holders were cleaned with alcohol wipes before reuse. After post weighing, the filters were placed in their corresponding petri dishes, parafilmed, bagged, and stored in the freezer at a temperature below 0°C. Outdoor and indoor $PM_{2.5}$ filters were sent by overnight service to Texas A&M University in Kingsville, Texas, for chemical composition analysis. The filters were transported in insulated Styrofoam coolers with frozen refrigerant gel packs to maintain a temperature below 4 °C.

Laboratory Qualification

Ohio University's Air Quality Laboratory is one of 9 laboratories participating in an ongoing Laboratory Inter-Comparison program sponsored by the USEPA/Harvard Particle Health Effects Center.[35] The round robin program is designed to assess the performance of each lab in handling weighing PM_{10} and $PM_{2.5}$ samples. The concluding remarks from the first and second rounds indicate that the Ohio University is proficient in analysis of low mass (25-1000 μg) gravimetric filter samples.

Data Validation and Analysis

In this study, data analyses complied with the published North American Research Strategy for Tropospheric Ozone (NARSTO) Data Management Handbook.[36]

The $PM_{2.5}$ concentration was determined by dividing the mass with the sample volume (the production of flow rate and sampling time). In this study, the continuous ambient 24-hr $PM_{2.5}$ concentrations were based on the 30-minute average readings of the TEOMs that were compiled by the TEOM software. Filter-based $PM_{2.5}$ concentrations were computed using the mass difference between the filter's initial and final weight obtained from gravimetric measurements.

All sample weights were corrected by subtracting the mean field blank weights. Samples from 1999 and 2000 were adjusted using the mean field blank of 1999 and of 2000 respectively. The detection limit (DL) was calculated as three times the standard deviation of the field blanks. Limits of detection (LODs) were defined as the ratio of DL to the sampling volume. Data points less than the LODs were included in the data analysis. Negative data points and data points due to operation and equipment failures (i.e. pump malfunction, tube disconnection, short sampling time) were not included in the analysis. Completeness was calculated as the number of collected samples divided by the target number of samples.

The mean, standard deviation, maximum, minimum and median were used to describe the distribution of $PM_{2.5}$ data. Linear regression was performed to determine the relationship between different sample methods, ambient $PM_{2.5}$ across different sites, and personal, indoor, and outdoor concentrations. The correlation coefficient (R) was used to indicate the strength of association between paired data. Also, the co-location data were assessed to detect the bias between each sample method by using paired t-test (at a significance level of 0.05). The results of the data analysis are displayed in tables and graphs. Units for $PM_{2.5}$ mass and $PM_{2.5}$ concentrations are reported in μg and $\mu g/m^3$ respectively.

RESULTS AND DISCUSSION

This section contains the results and the discussion of the data analysis. A short review of the field blanks, data completeness and co-locations is also provided. The data analysis includes; (1) evaluations of temporal and spatial variations of ambient $PM_{2.5}$ mass concentrations; (2) comparisons of personal, indoor, and outdoor $PM_{2.5}$ concentrations; (3) evaluations of indoor/outdoor (I/O) ratios of $PM_{2.5}$ mass concentration and I/O ratios of sulfate component of $PM_{2.5}$; and (4) correlations between outdoor, indoor, and personal $PM_{2.5}$ levels. In addition, the results from this study are compared with similar published studies to further understand the relationships between personal, indoor, and outdoor $PM_{2.5}$ exposures.

Field Blanks and Completeness

The results of the analysis of the field blanks are outlined in Table II.1.2. The overall mean net mass gain was –0.53, 4.11, and 3.35 μg for the outdoor, indoor, and personal measurements respectively. The limits of detection (LOD) for the outdoor, indoor, and personal measurements were 1.41, 6.31, and 9.01 $\mu g/m^3$ respectively.

The data capture was evaluated for each sampling method used in this study. Continuous ambient $PM_{2.5}$, or TEOM $PM_{2.5}$, was measured for approximately 20 months. Each site experienced equipment malfunctions during the study. For example, from the middle of August to December 1999, the TEOMs at Athens were shut down several times due to malfunctions of the mass flow controller. At Koebel, the TEOMs were shut down in part of September, October, and November 1999 due to internal temperature malfunctions. At New Albany, the TEOMs were down for repair during the months of May, July, November, and December in 1999, and during short time intervals in January, and March of 2000.

Table II..1.2. Statistics of field blanks for personal, indoor, and outdoor $PM_{2.5}$ measurements.

	Mean[a]	SD[a]	Max[a]	Min[a]	Size	LOD[b]
1999						
Outdoor	3.11	5.33	19.50	-8.50	44	
Indoor	2.69	4.22	12.00	-5.00	36	
Personal	3.57	3.97	15.50	-2.50	38	
2000						
Outdoor	-4.02	10.77	10.00	-40.00	46	
Indoor	6.43	13.14	33.00	-14.00	22	
Personal	9.00	16.64	39.00	-12.00	16	
Total						
Outdoor	-0.53	9.23	19.50	-40.00	90	1.41
Indoor	4.11	8.83	33.00	-14.00	58	6.31
Personal[c]	3.35	6.31	19.00	-12.00	51	9.01

[a] :µg
[b] :$\mu g/m^3$
[c] :excludes three outliers. (when outliers included, LOD = 13.97 $\mu g/m^3$)

The filter-based measurements included outdoor (ACCU $PM_{2.5}$), indoor, and personal $PM_{2.5}$ concentrations. When sampling problems (i.e. pump or battery failures, tube disconnection) or laboratory irregularities were encountered, the corresponding data points were not included in the analysis. Data capture or completeness was calculated as the number of collected daily samples divided by the target number of samples. Eighty-six percent of the outdoor, 88 percent of the indoor and 82 percent of the personal target samples were completed (Table II.1.3).

Co-locations

Co-locations involved side-by-side sampling for personal, indoor, and outdoor samplers employed in this study. Co-locations were predominantly conducted at Athens with a few conducted at Koebel. The personal and indoor sampling devices were located adjacent to the TEOMs and the ACCU system for the co-locations and 24-hr samples were obtained.

Table II.1.3. Summary of $PM_{2.5}$ sampling completeness, number of days useable samples collected.

Completed Samples	Personal	Indoor	Outdoor	
			ACCU	TEOM
Athens	207	298	300	442
Koebel	194	251	303	493
New Albany	205	270	297	554
Total Completed Samples	606	819	900	1489
Target Samples	742	926	1036	1830
Total Completeness	82%	88%	87%	81%

The $PM_{2.5}$ concentration monitored by separate devices, were assumed to be independent from each other. Bias between each sampler was determined by using a paired t-Test (at a significance level of 0.05). In this study, the paired t-Tests included:

- TEOMs versus ACCU system
- TEOMs versus indoor monitor
- TEOMs versus personal monitor
- ACCU system versus indoor monitor
- ACCU system versus personal monitor
- Indoor monitor versus personal monitor

Results from the t-test indicated that $PM_{2.5}$ concentrations obtained from each sampler were not significantly different from each other at the 0.05 level. From the simultaneous sampling, determination of $PM_{2.5}$ concentrations resulted in the mean concentrations ranging from 10.34 to 15.58 $\mu g/m^3$ as presented in Table II.1.4. The difference between the two mean outdoor concentrations was 1.05 $\mu g/m^3$, while the difference between the indoor and personal mean concentrations was 2.47$\mu g/m^3$.

Table II.1.4. Summary of collocated monitoring data (µg/m^3) for each sampler monitored by each sampling methods used in the t-test.

	Mean	SD	Max	Min	N
Personal	10.34	6.63	22.85	1.39	13
Indoor	12.81	6.77	29.84	5.04	13
ACCU $PM_{2.5}$	14.53	7.40	29.85	2.57	13
TEOM $PM_{2.5}$	15.58	6.83	30.59	5.64	13

Correlations between each sampler were also evaluated using linear regression. The strongest agreement between different sampling methods occurred between the ACCU system and the TEOMs (R = 0.87) followed by the personal-indoor monitor (R=0.72). Only modest correlations between other paired methods were found (Figure II.1.6 and Table II.1.5). These moderate correlations may have been the result of problems encountered during the co-location sampling. The indoor and personal monitors were designed for indoor use. During the co-location sampling period, it rained frequently, and the indoor and personal monitors did not operate efficiently under the high humidity conditions.

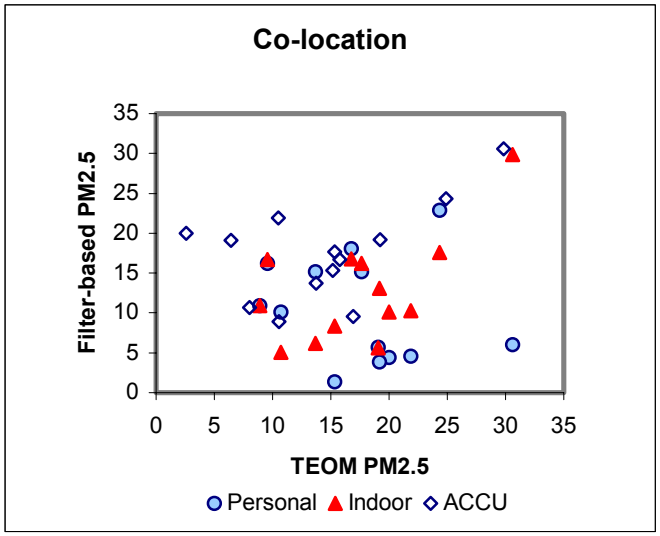


Figure II.1.6. Co-located ambient $PM_{2.5}$ concentrations (µg/m^3) measured by personal, indoor samplers, and the ACCU system plotted against those measured by the TEOMs (n=13).

Table II.1.5. Correlations between co-located samplers (μg/m^3).

	Intercept	Slope	R	N	P
ACCU vs. TEOM	2.48	0.82	0.87	41	<0.0001
Personal vs. TEOM	7.05	0.37	0.28	44	0.06635
Indoor vs. TEOM	8.83	0.41	0.28	35	0.10891
Personal vs. ACCU	5.05	0.48	0.57	30	0.00113
Indoor vs. ACCU	5.63	0.48	0.55	19	0.01486
Personal vs. Indoor	4.61	0.59	0.72	52	<0.0001

Regressing personal and indoor measurements with the ACCU system measurements resulted in similar R-values (0.57-0.55). Regressing personal and indoor measurements with TEOM measurements yielded an equivalent R-value (0.28). Personal and indoor measurements showed a stronger correlation with the ACCU system than with TEOMs. Since three stronger correlations were experienced between the filter-based measurements, the filter-based ambient measurements (ACCU system) were used to evaluate the correlations between personal, indoor, and outdoor concentrations.

Ambient $PM_{2.5}$

The highest ambient $PM_{2.5}$ occurred at the urban site (Koebel), while the lowest concentration occurred at the suburban site (New Albany). Ambient $PM_{2.5}$ illustrated a clear seasonal trend with high concentrations appearing in the warm months and low concentrations appearing in the cold months. The regression of TEOM $PM_{2.5}$ between the sites in the study area revealed a homogeneous distribution of ambient concentrations among the sites. At all three sites, daily ambient $PM_{2.5}$ did not exceed the 24-hr standard (65 μg/m^3). However, the annual standard of 15 μg/m^3 was exceeded at Koebel and Athens.

Ambient $PM_{2.5}$ Concentration

Table II.1.6 presents the results of TEOM monitoring for all three sites. The average annual ambient $PM_{2.5}$ concentrations ranged from 14.72 to 18.47 μg/m^3 for the entire sampling period. Annual average $PM_{2.5}$ concentration was ~3 μg/m^3 greater in the urban locations than in the rural and the suburban locations. Many historical and newly conducted research reviewed by the USEPA, report similar results.[37] These studies indicate that the annual $PM_{2.5}$ standard (15 ug/m^3) will be difficult to reach in most eastern states and in parts of California. The highest 24-hr ambient concentration occurred at Koebel (60.22 μg/m^3), which did not exceed the 24-hr standard of 65 μg/m^3.

Table II.1.6. Summary of ambient $PM_{2.5}$ concentrations ($\mu g/m^3$) for all study sites during the entire sampling period.

Total	Athens	Koebel	New Albany
Mean	15.35	18.47	14.72
SD	7.17	8.64	7.08
Max	40.99	60.22	47.81
Min	4.51	4.89	4.00
Median	13.44	16.61	13.07
25%	10.10	12.28	9.63
75%	18.73	22.44	18.48
N	438	491	552

As expected, the urban site, Koebel, had the highest ambient $PM_{2.5}$ concentration. Figure II.1.7 shows the diurnal variations in $PM_{2.5}$ concentrations. Koebel's ambient $PM_{2.5}$ showed a slightly different diurnal pattern than Athens and New Albany. Two prominent peaks, morning and evening, existed at Athens and New Albany corresponding with daily commuter traffic (from 8:00 a.m. to 10:00 a.m. and from 5:00 p.m. to 9:00 p.m.). The morning peak $PM_{2.5}$ concentrations at Athens were similar with New Albany. While the evening peak of Athens was greater than New Albany and about one hour later. In comparison, Koebel had several peaks during the day.

Overall Koebel's mean hourly ambient $PM_{2.5}$ was consistently higher (range= 15 to 22 $\mu g/m^3$) than the other two sites (range= 12 to 18 $\mu g/m^3$). This difference probably resulted from local sources. Recall that Koebel is located in an industrial center of Columbus and is close to a major transportation artery, so the ambient $PM_{2.5}$ concentration is unavoidably influenced by sources from the surrounding industry and nearby traffic. Röösli et al. reported a 30% increase of $PM_{2.5}$ and PM_{10} concentrations for sites exposed to traffic.[38]

In this study, seasonal trends of $PM_{2.5}$ concentrations existed at all three sites with the maximum $PM_{2.5}$ concentrations occurring during the summer months (Figures II.1.8 and II.1.9). Studies show $PM_{2.5}$ concentrations typically peak in the summer months.[39] This seasonal trend mainly results from the formation of major components of $PM_{2.5}$ such as sulfate (SO_4^{2-}). Sulfate is generated through the oxidation of SO_2 involving OH radicals, O_3, and H_2O_2 species. These species are generated during the photochemical smog processes and their concentrations increase with sunlight and temperature. The seasonal pattern of ambient $PM_{2.5}$ is more pronounced in the eastern U.S. where higher sulfate concentrations appear than the west.

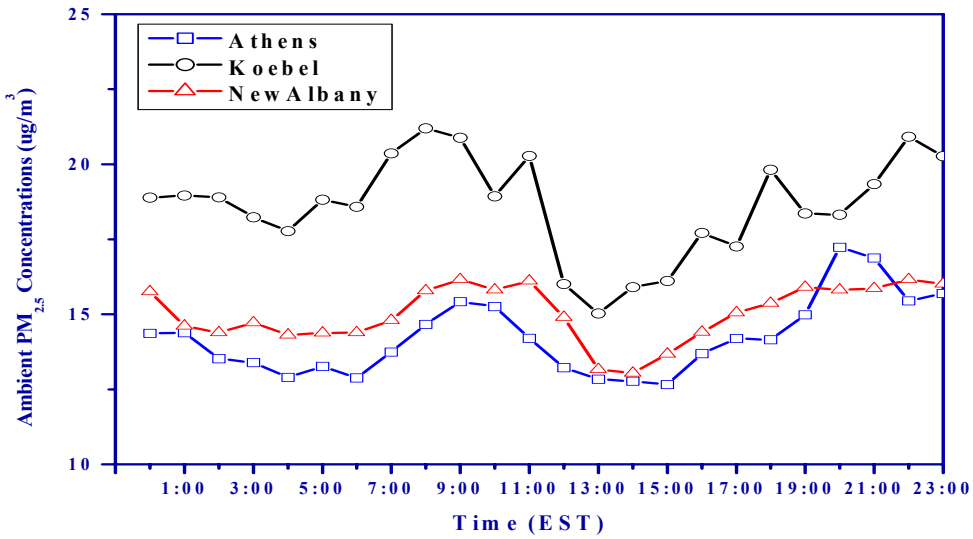


Figure II.1.7. Diurnal distribution of ambient $PM_{2.5}$ at all sites (1999 data).

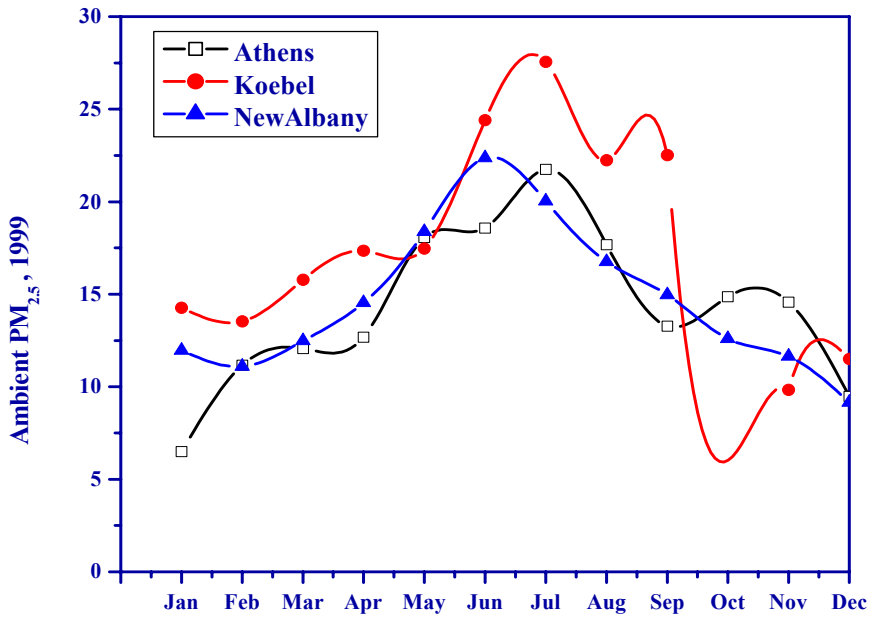


Figure II.1.8. Monthly ambient $PM_{2.5}$ concentrations ($\mu g/m^3$) monitored by TEOMs in 1999.

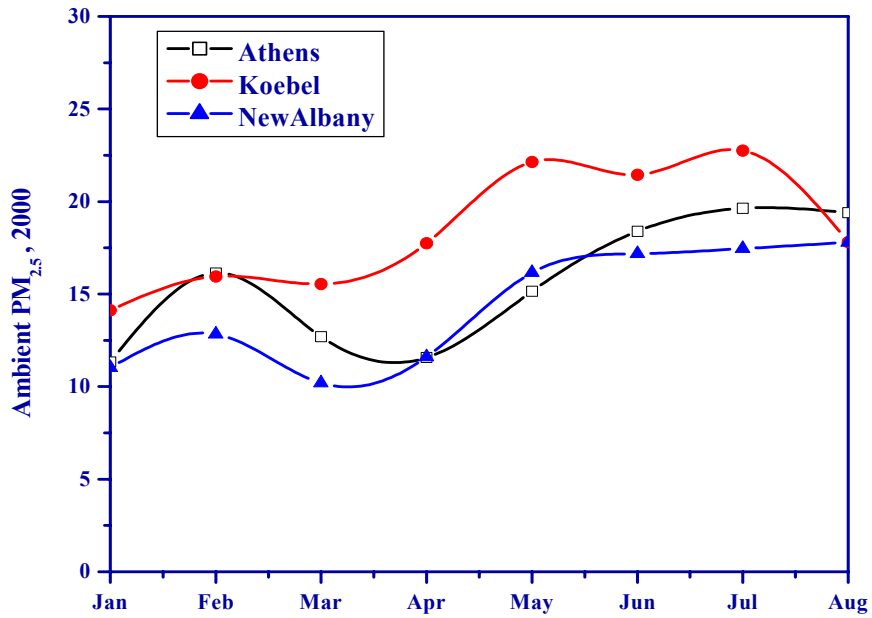


Figure II.1.9. Monthly ambient $PM_{2.5}$ concentrations ($\mu g/m^3$) monitored by TEOMs in 2000.

Spatial Distribution of Ambient $PM_{2.5}$

As shown in Figure II.1.10, strong correlations existed between the ambient $PM_{2.5}$ concentrations measured at all three sites, despite the differences in $PM_{2.5}$ concentrations at individual sites. The linear regression between paired sites produced coefficients (R) raging from 0.60 to 0.78.

When dividing the data into the warm season (April – September) and the cold season (October-March) as shown in Table II.1.7, higher correlations between the sites exist in the warm months (R range = 0.66-0.79) than the cold months (R range = 0.60-0.66), with the exception of the correlation between New Albany and Athens. Overall, the highest correlation occurred between Koebel and New Albany. These limited spatial variations of ambient $PM_{2.5}$ indicate the influence of regional $PM_{2.5}$ sources.

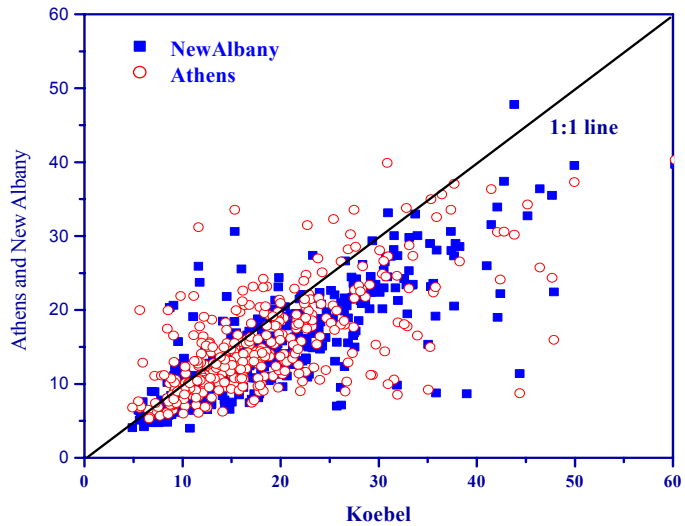


Figure II.1.10. Inter-site correlation of ambient $PM_{2.5}$. Ambient $PM_{2.5}$ concentrations measured at Athens and New Albany plotted against those measured at Koebel. A linear regression of Koebel vs. New Albany had (n=457) an R=0.80 with a slope of 0.62. A linear regression of Athens vs. Koebel (n=373) had an R=0.70 with a slope of 0.54. A linear regression of Athens vs. New Albany (n=411) had an R=0.60 with a slope of 0.58.

Table II.1.7. Correlations of ambient $PM_{2.5}$ concentrations($\mu g/m^3$) between three sites.

	Intercept	Slope	R	N	P
Warm					
Koebel vs. New Albany	3.53	0.62	0.79	281	<0.0001
Koebel vs. Athens	6.36	0.51	0.66	226	<0.0001
New Albany vs. Athens	8.52	0.49	0.51	257	<0.0001
Cold					
Koebel vs. New Albany	4.73	0.46	0.66	176	<0.0001
Koebel vs. Athens	5.34	0.48	0.60	147	<0.0001
New Albany vs. Athens	3.21	0.81	0.72	154	<0.0001

Koebel, the central urban location, had slightly higher concentrations than the other sites, which was potentially influenced by local sources. Figure II.1.11 shows a typical ambient $PM_{2.5}$ episode at the study sites. Koebel showed similar temporal patterns with the suburban site. Most strikingly, Athens, which was approximately 120 km from the two sites in Columbus, also depicted a very similar temporal profile over the entire sampling period.

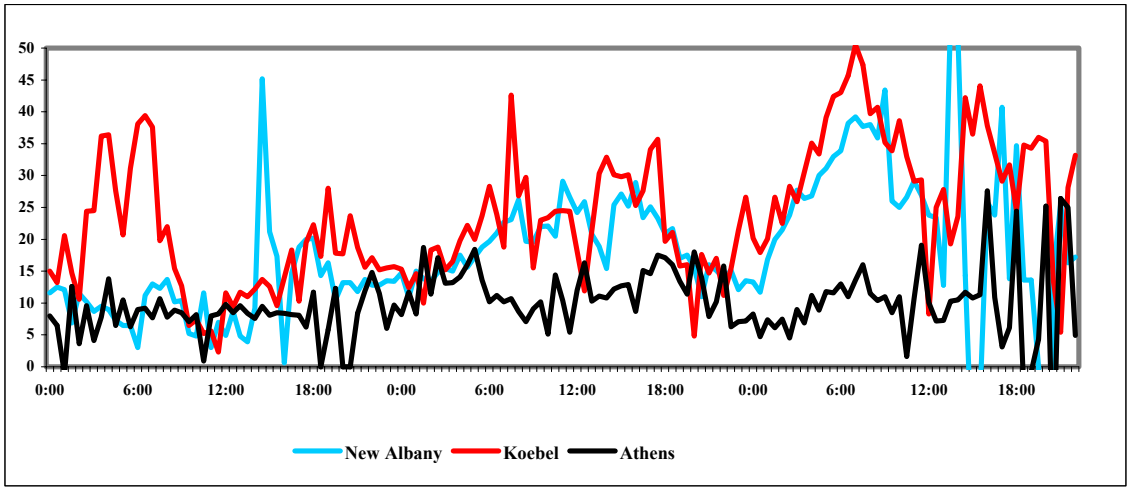


Figure II.1.11. Typical daily variations of ambient $PM_{2.5}$ at three sites. Half-hour TEOM $PM_{2.5}$ concentrations in a three 24-hr interval from May 16 to May 18, 2000.

Similar to the inter-site correlations, and comparisons of ambient $PM_{2.5}$ concentrations for the three sites in this study, sites within the region also showed strong correlations of ambient $PM_{2.5}$ concentrations as shown in Figure II.1.12.

The Advanced Technology System (ATS) monitor provides a comparable ambient $PM_{2.5}$ dataset for Athens. It resides at Gifford State Forest (39.44°N, 81.90°W) approximately 50 km northeast of Athens. Twenty-four-hr $PM_{2.5}$ mass was collected on the 47 mm Teflon filter every fifth day from June 1999 through August 2000. The ATS data depicted a mean ambient $PM_{2.5}$ concentration of 12.54 ± 6.10 μg/m^3 (N=89). While the Athens measurements, during the same periods, yielded a mean concentration of 16.01 ± 7.36 μg/m^3 (N=307). The correlation of ambient measurements at the two sites was strong with an R of 0.65 (Figure II.1.12).

For the New Albany and Koebel sites, datasets from nearby Ohio EPA monitoring sites, Maple Canyon and Woodrow, provide a comparable dataset. Maple Canyon is approximately 12 km west of New Albany and Koebel is about 1.5 km southeast of Woodrow. The mean ambient concentrations at New Albany and Maple Canyon as measured every third day from January to December 1999, were 14.86 ± 7.33 μg/m^3 and 16.90 ± 7.51 μg/m^3 respectively. The mean ambient concentrations at Koebel and Woodrow (daily measurement of 1999) were 18.51 ± 9.14 μg/m^3 and 17.38 ± 7.06 μg/m^3 respectively. Strong correlations exist between the paired ambient concentrations monitored at New Albany and Maple Canyon (R=0.73), and between Koebel and Woodrow (R=0.66) as shown in Figure II.1.12.

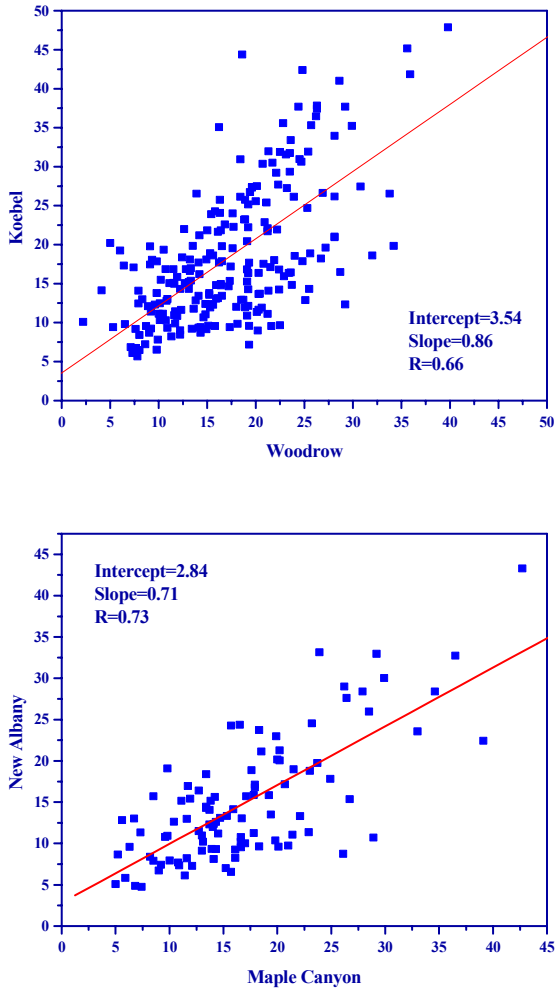


Figure II.1.12. Correlations of ambient $PM_{2.5}$ concentrations of the sites from this study and the corresponding sites within the region.

The uniform distribution of ambient $PM_{2.5}$ in this study is similar with results from other studies conducted in the U.S. and Europe.[40] These studies reported limited spatial variations of $PM_{2.5}$ and PM_{10} concentrations across multiple sites on a scale of 1-36 km with higher correlations occuring in the summer. Burton et al. reported that $PM_{2.5}$ across eight sites in Philadelphia was close to unity.[41] In the central region of Baltimore County, Williams et al. also found high correlation coefficients (R) ranging from 0.94 to 0.96.[42] In the USEPA's Particle Total Exposure Assessment Methodology (PTEAM) study, central-site $PM_{2.5}$ correlated well with $PM_{2.5}$ concentrations monitored immediately outside the participants' homes with the R-coefficients of 0.92 and 0.96 for daytime and overnight respectively.[43] Similar results were documented in the Air Pollution Exposure Distribution within adult urban populations in Europe (EXPOLIS) which showed good agreement between

$PM_{2.5}$ monitored at home outdoors and fixed site measurements.[44] The spatial homogeneity suggests that a fixed-site monitor can represent $PM_{2.5}$ concentrations throughout a large area (i.e. a metropolitan area, a town, small city, and a region).

Outdoor, Indoor, and Personal $PM_{2.5}$

$PM_{2.5}$ monitoring included outdoor (measured by the ACCU systems), indoor, and personal $PM_{2.5}$ measurements. Comparisons indicated that personal and indoor concentrations were higher than the outdoor concentrations at all sites. Comparisons between indoor and outdoor $PM_{2.5}$ mass concentration (I/O) ratios and $PM_{2.5}$ sulfate concentration I/O ratios indicated indoor $PM_{2.5}$ was greatly influenced by human activity.

In this study, measurements of indoor and personal $PM_{2.5}$ occurred when school was in session, typically from 8:00 a.m. to 3:00 p.m. From January to June 1999, the filter-based measurement (ACCU $PM_{2.5}$ concentrations) were also made from 8:00 a.m. to 3:00 p.m. Starting in September of 1999, through the completion of the study, the ambient filter-based measurements were made from 12:00 a.m. to 12:00 p.m., in order to capture sufficient mass for chemical speciation. For pairwise correlation, the 24-hr average ambient filter-based measurements were adjusted to match the 7-hr indoor and personal samples by using a weighting factor calculated from the TEOM measurements.

Comparisons Between Personal, Indoor, and Outdoor Concentrations

The results of filter-based measurements for all three sites are summarized in Table II.1.8. For all three sites, the mean indoor and personal $PM_{2.5}$ concentrations were consistently higher than the outdoor concentration. Outdoor $PM_{2.5}$ illustrated a clear seasonal trend with higher concentrations appearing during the warm months than cold months. However, personal and indoor concentrations did not demonstrate this apparent seasonal pattern as shown in Figure II.1.13. The dissimilarity of seasonal patterns between outdoor, indoor, and personal concentrations indicate the potential differences in their sources. Outdoor $PM_{2.5}$ differs from non-ambient $PM_{2.5}$ because it is influenced by regional-scale pollution emissions and meteorological conditions.[45] In comparison, nonambient $PM_{2.5}$ is highly influenced by the individual's activity-patterns and the ventilation conditions where the individual spends time.

Table II.1.8. Summary of filter-based $PM_{2.5}$ measurements ($\mu g/m^3$) for all study sites.

		Athens	Koebel	New Albany
Outdoor $PM_{2.5}$	**Mean**	13.66	13.89	12.72
	SD	8.91	9.29	8.86
	Max	61.12	77.01	69.30
	Min	0.50	0.24	0.05
	Median	11.49	11.65	10.87
	25%	8.30	8.12	7.62
	75%	17.08	16.65	15.43
	Size	332	315	327
Indoor $PM_{2.5}$				
	Mean	17.20	14.98	16.52
	SD	13.56	12.30	13.53
	Max	71.57	68.37	69.51
	Min	0.45	1.05	0.24
	Median	12.28	10.55	11.56
	25%	8.60	7.28	8.42
	75%	20.78	16.23	19.46
	Size	298	251	270
Personal $PM_{2.5}$				
	Mean	17.61	14.59	13.93
	SD	17.81	13.05	12.25
	Max	88.38	66.96	56.90
	Min	0.17	0.42	0.95
	Median	9.53	10.18	9.45
	25%	6.19	7.52	6.86
	75%	22.46	15.07	14.86
	Size	207	194	205

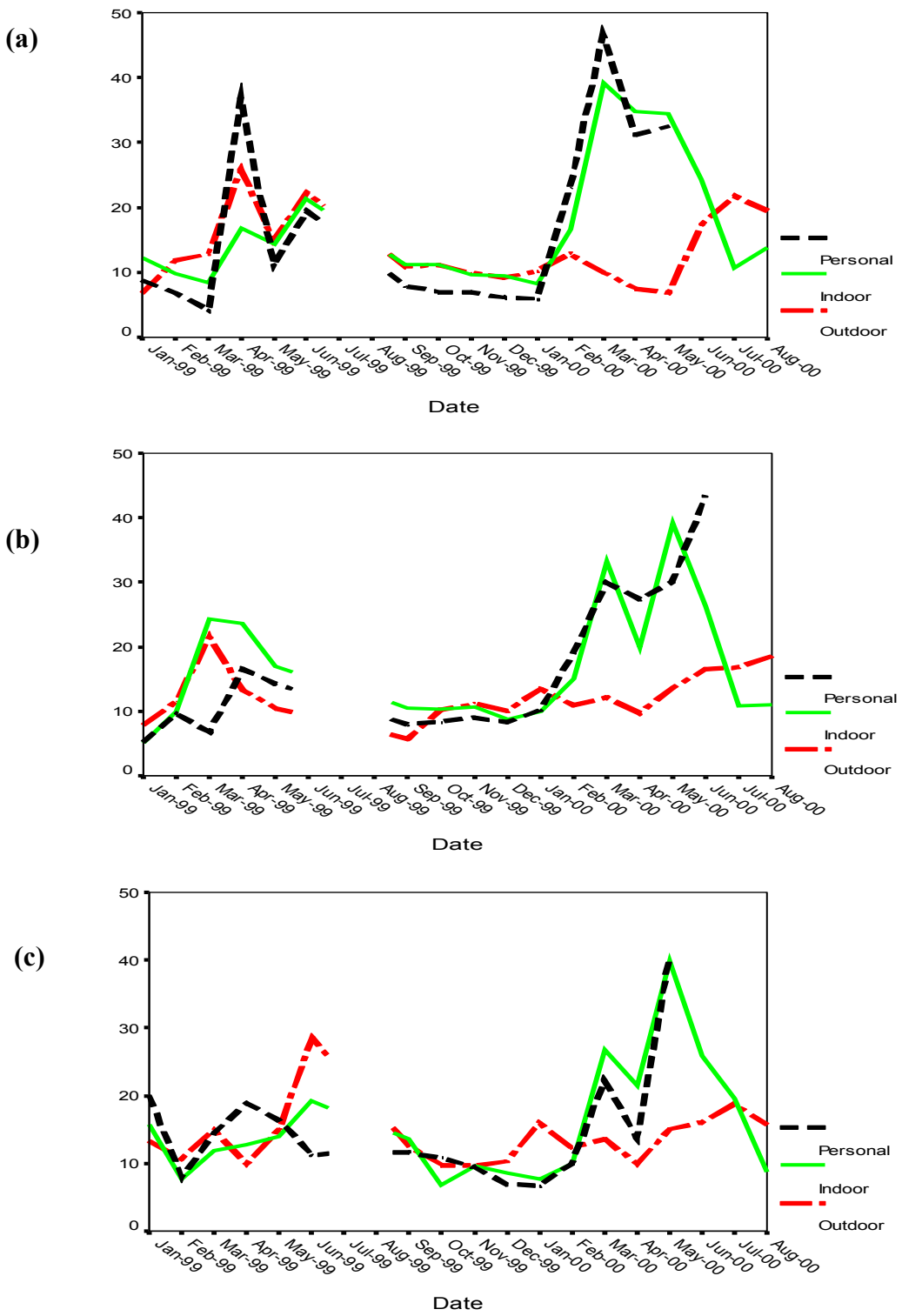


Figure II.1.13. Monthly average filter-based $PM_{2.5}$ concentrations ($\mu g/m^3$) at all sites.

Several reports have documented human activity as being responsible for high indoor and personal PM concentrations when no apparent indoor source exists.[46] Long et al. reported an increase in $PM_{2.5}$ concentration of approximately 23 ± 23 μg/m^3 and 12 ± 9.1 μg/m^3 associated with eleven dusting and fifteen vigorous walking events respectively.[47] In studying I/O ratios of PM (size range = 0.01- 2.5 μm) at Fresno, CA, Vette et al. found resuspension from activities during the day responsible for the observed diurnally I/O ratio variations for PM with size range > 1μm.[48] This study was designed to minimize the impact of indoor $PM_{2.5}$ sources including tobacco smoke and cooking activities. Consequently, the elevated indoor and personal $PM_{2.5}$ concentrations appear to be mainly related to human activity patterns. As the subjects of this study were active and spent most of their time in large groups, their activity patterns would influence resuspensions and reentrainment, resulting in elevated personal and indoor concentrations.

Comparisons Between Outdoor, Indoor, and Personal $PM_{2.5}$ by Sites

The comparison of personal, indoor, and outdoor $PM_{2.5}$ concentrations was unique for each monitoring location. Athens had the largest difference between mean personal and outdoor concentrations (3. 95 μg/m^3), followed by New Albany (1.16 μg/m^3), and Koebel (0.70 μg/m^3). Comparison of the differences between the mean personal and indoor $PM_{2.5}$ at Athens resulted in almost equivalent values (personal =17.61 μg/m^3, indoor= 17.20 μg/m^3). Athens had the highest personal and indoor $PM_{2.5}$ concentrations.

Koebel had the highest mean outdoor $PM_{2.5}$ (13.89 μg/m^3) concentrations and the lowest indoor $PM_{2.5}$ concentration (14.98 μg/m^3). The average indoor concentration exceeded the average outdoor concentrations at Koebel by 1.09 μg/m^3. While the personal concentration (14.59 μg/m^3) was similar with the indoor levels at Koebel.

New Albany, the suburban site, had the lowest average ambient $PM_{2.5}$ concentration (12.77 μg/m^3) and the lowest average personal levels (13.93 μg/m^3). The largest difference between the indoor and outdoor mean concentrations occurred at New Albany (3.75 μg/m^3), followed by Athens and Koebel. The average indoor concentrations exceeded the personal concentrations at New Albany by 2.59 μg/m^3.

Indoor and Outdoor Concentration (I/O) Ratios

The identification of indoor sources and their influence on total $PM_{2.5}$ exposures is important because the average U.S. citizen spends more time indoors than outdoors. Indoor air is influenced by human activities (i.e. cooking, cleaning, and general activities) and ventilation systems (i.e. open or closed windows and doors, air-conditioning systems). Evaluation of indoor and outdoor concentration (I/O) ratios is one method of characterizing the influence of indoor sources. In this study, two types of I/O ratios: $PM_{2.5}$ mass

concentration I/O ratios and $PM_{2.5}$ sulfate concentration I/O ratios, were evaluated. The analyses indicated that the indoor and personal $PM_{2.5}$ concentrations were highly affected by human activity. The influence of human activity on $PM_{2.5}$ levels was most prominent at New Albany and Athens.

Mass Concentration I/O Ratios

The I/O ratios were grouped into separate categories: school days and non-school days. During the school days, the mean I/O ratios ranged from 1.71 to 2.98 while during the non-school days, the mean I/O ratios ranged from 0.80 to 1.27 (Table II.1.9). As shown in Figure II.1.14, the I/O ratios dropped at all three sites when students were on vacation (starting from early June 2000). Overall, at Athens and New Albany, the I/O ratios of non-school days dropped 69% and 72% respectively compared to school days. However, Koebel only dropped 26%.

Table II.1.9. Summary data for $PM_{2.5}$ average concentrations ($\mu g/m^3$) and I/O ratios during school days and non-school days at all sites.

	School day			Non School day		
	Indoor	**Outdoor**	**I/O ratios**	**Indoor**	**Outdoor**	**I/O ratios**
Athens						
Mean	17.68	12.36	2.61	15.11	19.53	0.80
SD	14.22	8.51	5.76	10.05	8.34	0.70
Max	71.57	61.12	57.43	47.31	42.66	3.34
Min	0.45	0.50	0.17	1.01	4.95	0.10
Median	12.23	10.70	1.00	12.62	17.98	0.65
Size	242	272	235	56	60	31
Koebel						
Mean	14.44	13.44	1.71	17.46	16.04	1.27
SD	11.73	9.28	3.17	14.53	9.12	1.16
Max	68.37	77.01	31.69	55.15	48.16	4.94
Min	1.32	0.24	0.16	1.05	3.36	0.04
Median	10.54	11.30	0.98	12.85	15.22	0.96
Size	206	261	196	45	54	43
New Albany						
Mean	17.74	11.98	2.98	11.75	16.74	0.82
SD	14.47	8.59	5.47	7.27	9.09	0.60
Max	69.51	69.30	49.44	34.13	41.34	2.96
Min	0.71	0.05	0.05	0.24	3.10	0.13
Median	11.76	10.52	1.16	10.36	15.31	0.63
Size	215	273	208	55	54	54

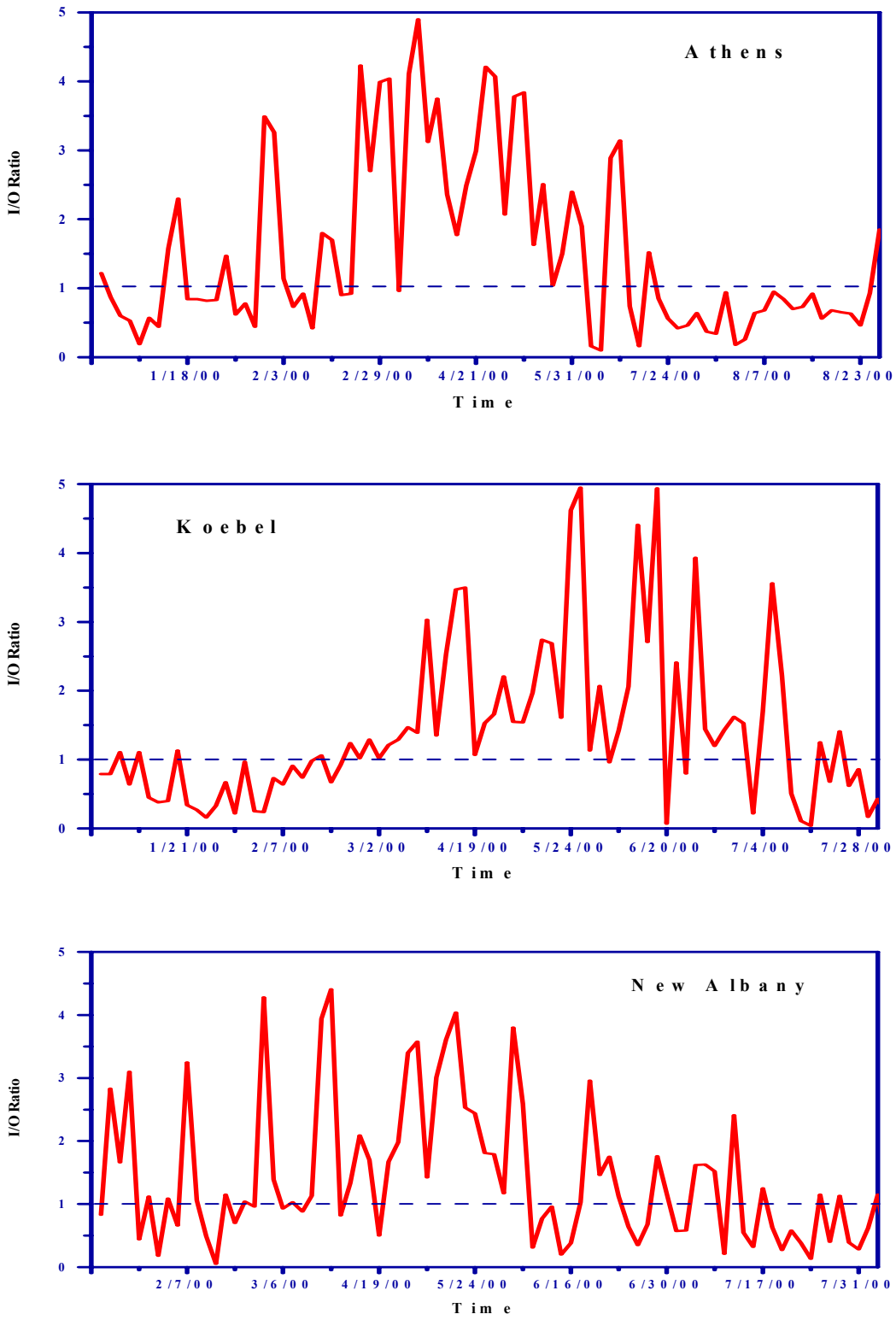


Figure II.1.14. School day I/O ratios vs. non-school day I/O ratios at three sites, 2000. I/O ratios greater than 5 are not included in the graph.

In general I/O ratios decreased when students were not present in the buildings. This is the result of reduced human activity and its impact on resuspension. However, the extent of the I/O ratio decrease varied at each site; the fluctuation pattern in the urban site, Koebel, differed from Athens and New Albany. During non-school days, I/O ratios at Athens and New Albany were less than 1, whereas Koebel's I/O ratios reduced slightly from 1.71 during school days to 1.27 during non-school days. Overall, I/O ratios at Koebel were greater than 1. The dramatic decline of indoor concentrations and I/O ratios associated with the absence of students was not experienced at Koebel. The activities of the summer school students at Koebel may contribute to the high indoor $PM_{2.5}$ concentrations.

These results are supported by several studies in which I/O relationships were highly influenced by indoor sources (i.e. human activities, ventilation conditions). This conclusion results from three observations. First, higher I/O ratios occur when human activity is high, while low I/O ratios occur when human activity is low. In nine homes in the Boston area, Long et al. reported the mean I/O ratios for $PM_{2.5}$ as 2.4 ± 14 for daytime while 0.74 ± 0.41 for nighttime. Second, high variability of indoor $PM_{2.5}$ concentrations occur during high I/O ratio time, which suggest the influence of short-term impacts of indoor source events.[49] In a fifth-grade classroom in Lindon, Utah, Patterson et al. reported prominent indoor $PM_{2.5}$ concentration peaks occurred from 9:00 a.m. to 4:00 p.m. each school day. No midday peaks existed during the weekend.[50] Third, in the absence of indoor sources, I/O ratios are less than or equal to 1. According to Wallace, an I/O ratio for $PM_{2.5}$ at homes with no apparent indoor sources was 0.65.[51]

Results similar to previous research were found in this study. First, higher I/O ratios appear during school days when human activity was high, while low I/O ratios occurred during the non-school days when human activity is minimal. Second, large variations of school day indoor $PM_{2.5}$ concentrations appeared in school days when I/O ratios were also high. Third, the I/O ratios were rarely below 1 during school days at all three sites. Since the indoor sources from smoking, cooking, and cleaning were minimized in this study, the indoor $PM_{2.5}$ was mainly generated by human activity.

$PM_{2.5}$ Sulfate Concentration I/O Ratios

Indoor $PM_{2.5}$ is the combination of ambient-origin $PM_{2.5}$ (ambient $PM_{2.5}$ that has infiltrated indoors) and indoor-generated $PM_{2.5}$. Sulfate (SO_4^{2-}) is commonly used as a marker of outdoor air in indoor environments due to its stability and absence of indoor origins. Particle sulfate results from atmospheric chemical reactions of gaseous sulfur dioxide, which is emitted from sulfur-containing fossil fuel combustion. Sulfate is not only the most homogenous pollutant in the ambient air, but also a strong marker for ambient-origin $PM_{2.5}$. Studies have reported sulfate as being the prime chemical

constituent of $PM_{2.5}$.[52] In the eastern U.S., sulfate constitutes approximately 30-40 percent of $PM_{2.5}$ compositions. Indoor aerosol sulfate arises through the resuspension of deposited outdoor sulfate tracked indoors by human activity.

In this study, two types of sulfate concentrations, indoor and outdoor, were available (refer to Section 2 Chapter 2). Similar to the $PM_{2.5}$ mass concentration I/O ratios, sulfate I/O ratios also varied between school days and non-school days. (Table II.1.10) Overall, the school sulfate I/O ratios were greater than 1.36, in contrast to the data of Lee et al. (0.81)[53], Li and Harrison (0.89).[54] During the non-school day, sulfate I/O ratios reduced to near unity (range = 0.98 ~ 1.25) at all sites. Studies suggest that for fine particles, the indoor/outdoor ratios appear lower or near unity in the absence of apparent indoor sources such as smoking, cooking and cleaning.[55] For example, Jones et al. reported sulfate I/O ratios of fine particulate matter (d_{ae} range = 1.1-2.1 μm) from 0.6 to 0.9 at twelve residences in Bringham, England.[56] The variations of sulfate I/O ratios with school day I/O ratios greater than the unity in this study, again, indicated the contributions of resuspension-related human activity to indoor $PM_{2.5}$.

Table II.1.10. Summary data for $PM_{2.5}$ sulfate concentration I/O ratios during school days and non-school days at all sites.

		Average	SD	Max	Min	Size
Athens	**School Day**	1.36	2.26	15.76	0.10	139
	Non School Day	1.25	1.25	4.69	0.16	29
Koebel						
	School Day	1.98	4.81	27.40	0.08	87
	Non School Day	0.98	1.83	10.35	0.01	39
New Albany						
	School Day	3.24	10.26	100.8	0.02	110
	Non School Day	1.06	1.34	7.88	0.10	49

The temporal patterns of sulfate I/O ratios were different at each site. At Athens and Koebel, the average indoor sulfate concentrations were lower than the average outdoor concentrations. For New Albany, the indoor sulfate was slightly higher than the outdoor concentration. While similar sulfate I/O ratios occurred at Athens (1.36) and Koebel (1.98) during school days, New Albany's sulfate I/O ratio (3.24) was significantly higher. The greatest difference between school day and non-school day sulfate I/O ratios occurred at New Albany (2.18), followed by Koebel (1.00). When the students were on vacation, the I/O ratio at New Albany reduced from 3.24 to near unity (1.06). However, only slight drop of sulfate I/O ratio values occurred at Athens (0.11). The low school day I/O ratios occurring at New Albany indicate that indoor

$PM_{2.5}$ at this site is strongly influenced by indoor sources generated by human activities.

Both Athens and New Albany have air-conditioning systems, and different patterns of I/O relationships exist in these schools. This may be the result of varying activity patterns between the student groups and of the frequency and time of open windows and open doors at each site. It may also result from the difference between the school buildings. The New Albany school was built in 1998, which is the newest building among the three school buildings, as such it will have less outdoor air infiltration. In addition, the classroom at New Albany was the only carpeted one among all three sites. Studies investigating indoor particle characters show that carpets act as long-term reservoirs for suspendable particles.[57] The variations of the I/O relationships indicate the complexity and difficulty in analyzing the influence of indoor sources, which may lead to the complexity of correlations between personal, indoor, and outdoor $PM_{2.5}$ concentrations.

Correlations between Personal, Indoor and Outdoor $PM_{2.5}$

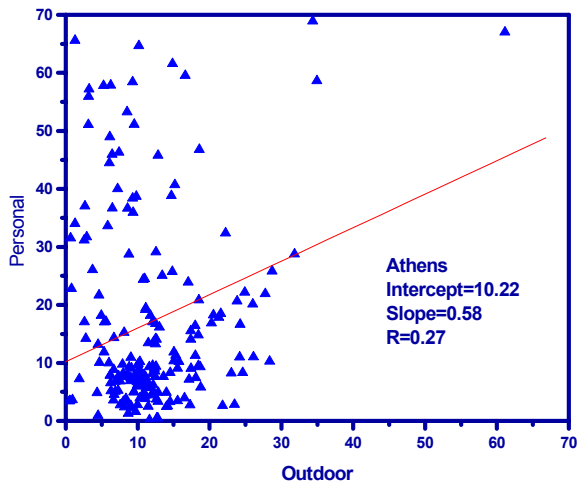
Results from regression analysis for personal, indoor, and outdoor $PM_{2.5}$ concentrations are presented in Table II.1.11 and Figure II.1.15, Figure II.1.16, and Figure II.1.17. The coefficients (R) of all pairwise comparisons were positive for all sites. Strong correlations existed between personal and indoor $PM_{2.5}$ concentrations at all three sites with R values ranging from 0.30 to 0.68. Weak to moderate correlations occurred between personal and outdoor $PM_{2.5}$ concentrations and between indoor and outdoor concentrations. The R-value ranged from 0.04 to 0.32 for personal-outdoor correlations, and from 0.13 to 0.30 for indoor-outdoor correlations.

Table II.1.11. Summary of correlations of personal, indoor, and outdoor $PM_{2.5}$ concentrations*.

	Intercept	Slope	R	N	P
Personal vs. Outdoor					
Athens	10.22	0.58	0.27	202	0.0001
Koebel	8.42	0.50	0.32	186	<0.0001
New Albany	13.37	0.05	0.04	200	0.59
Personal vs. Indoor					
Athens	2.03	0.87	0.68	196	<0.0001
Koebel	4.96	0.70	0.56	169	<0.0001
New Albany	8.19	0.32	0.30	182	<0.0001
Indoor vs. Outdoor					
Athens	14.28	0.19	0.13	292	0.03
Koebel	10.43	0.36	0.26	240	<0.0001
New Albany	10.77	0.44	0.30	265	<0.0001

* Data for personal-outdoor and personal-indoor correlation analyses are based on school day data. Indoor-outdoor correlation analysis includes both school day and non-school day data. Exclude data for non-school day yields indoor-outdoor correlations at three sites with R of 0.13, 0.32, 0.36 for Athens, Koebel, and New Albany respectively.

Correlations between personal, indoor, and outdoor $PM_{2.5}$ concentrations varied among the sites. Koebel, the urban site, had the strongest correlations for personal-outdoor and indoor-outdoor $PM_{2.5}$ concentrations. While Athens, the rural site, had the strongest personal-indoor correlation. The personal-outdoor correlations followed the pattern Koebel > Athens > New Albany. The indoor-outdoor correlations followed the pattern Koebel > New Albany > Athens. The order of personal-indoor correlations was Athens > Koebel > New Albany.

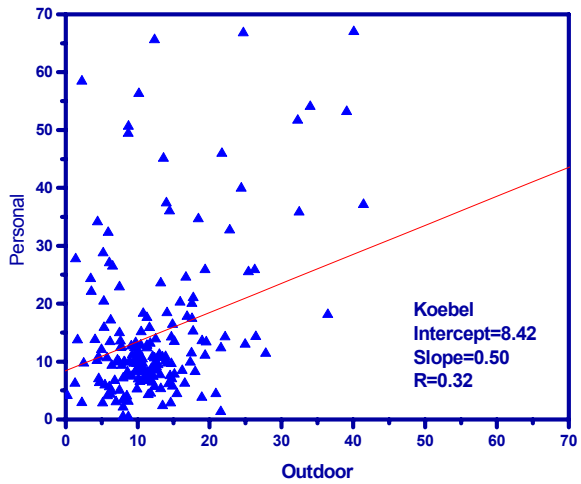


Athens

Intercept=10.22

Slope=0.58

R=0.27

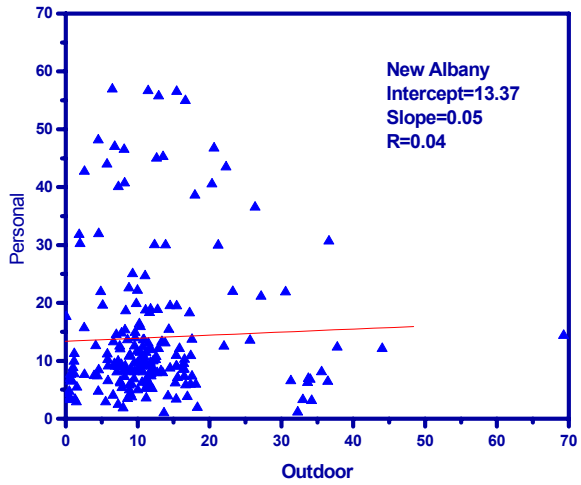


Koebel

Intercept=8.42

Slope=0.50

R=0.32



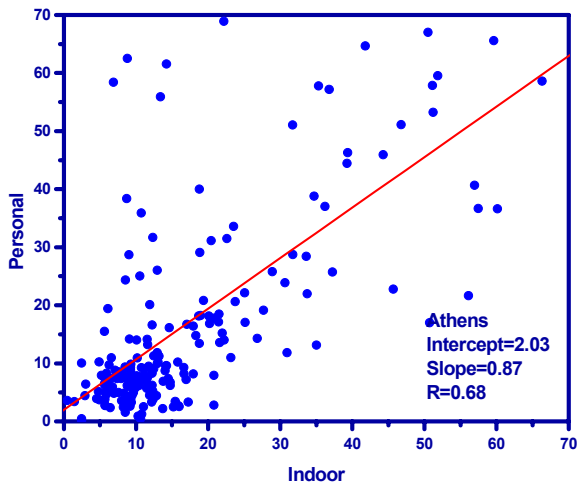
New Albany

Intercept=13.37

Slope=0.05

R=0.04

Figure II.1.15. Personal-outdoor relationships during the sampling period.

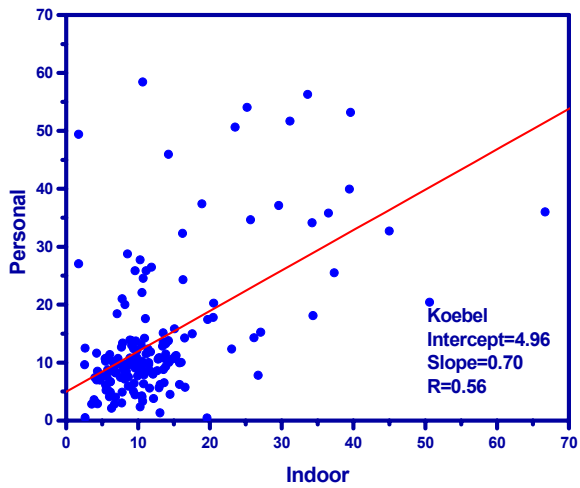


Athens

Intercept=2.03

Slope=0.87

R=0.68

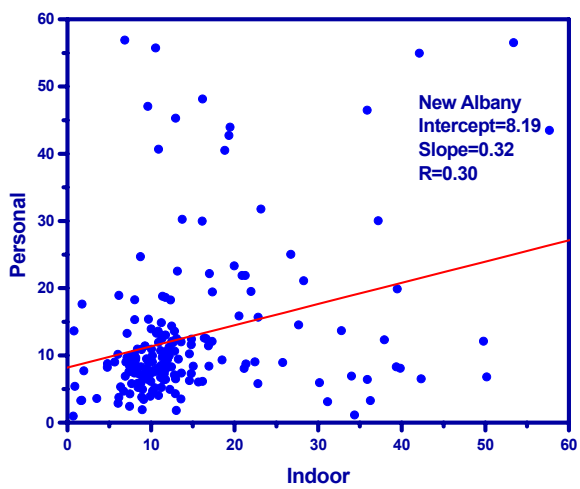


Koebel

Intercept=4.96

Slope=0.70

R=0.56



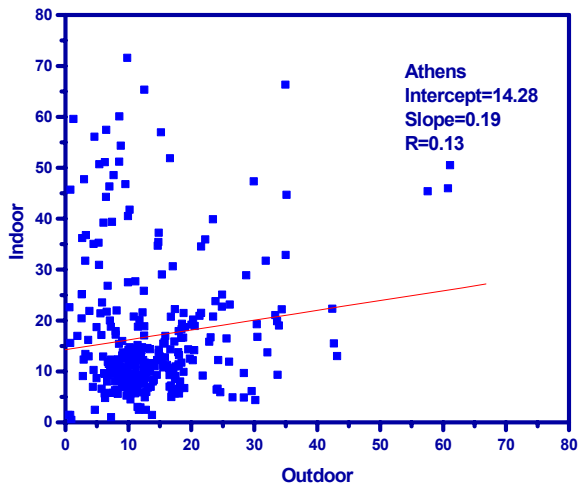
New Albany

Intercept=8.19

Slope=0.32

R=0.30

Figure II.1.16. Personal-indoor relationships during sampling period.

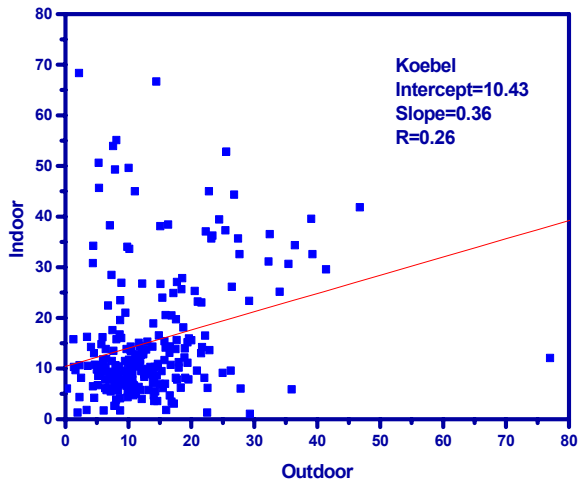


Athens

Intercept=14.28

Slope=0.19

R=0.13

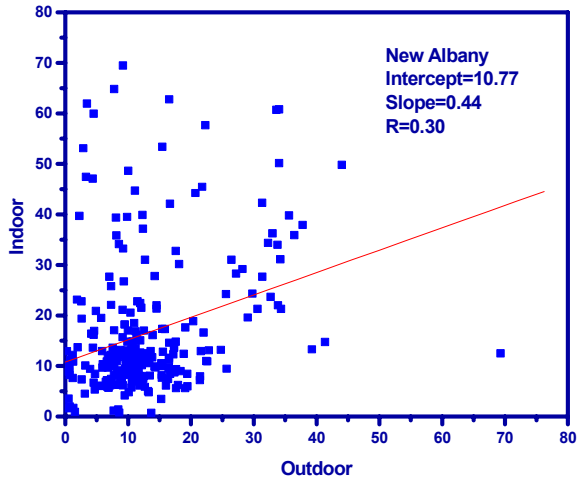


Koebel

Intercept=10.43

Slope=0.36

R=0.26



New Albany

Intercept=10.77

Slope=0.44

R=0.30

Figure II.1.17. Indoor-outdoor relationships during sampling period.

Correlations by Sampling Years and Seasons

Most PM exposure studies have been conducted during a single season, which leads to limitations in characterizing the variations of ambient concentrations, air exchange rates, and personal activities across seasons. The results of this study indicated large variations in correlations between personal, indoor, and outdoor $PM_{2.5}$ concentrations over multiple seasons. As shown in Figure II.1.18, all pairwise correlations fluctuated from month to month during the two-year study period with R-values ranging from below zero to near unity. No apparent patterns were found between different pairwise correlations. This illustrates the complexity of personal-indoor-outdoor correlations.

To examine temporal and seasonal correlation trends on personal, indoor, and outdoor $PM_{2.5}$ concentrations, data analysis of sampling years (1999 and 2000) and by seasons (warm; April to September; and cold; October to March) were examined separately. Strong and consistent correlations existed between personal and indoor $PM_{2.5}$ levels. This finding supports the hypothesis that indoor $PM_{2.5}$ is an important determinant of the total personal $PM_{2.5}$ exposure. Personal-outdoor and indoor-outdoor correlations varied during different time frames. All pairwise correlations were significant at Koebel regardless of sampling periods or seasons. More complex correlation patterns were experienced at Athens and New Albany. The results of correlations by sampling year and by season are summarized in Tables II.1.12, II.1.13, and II.1. 14.

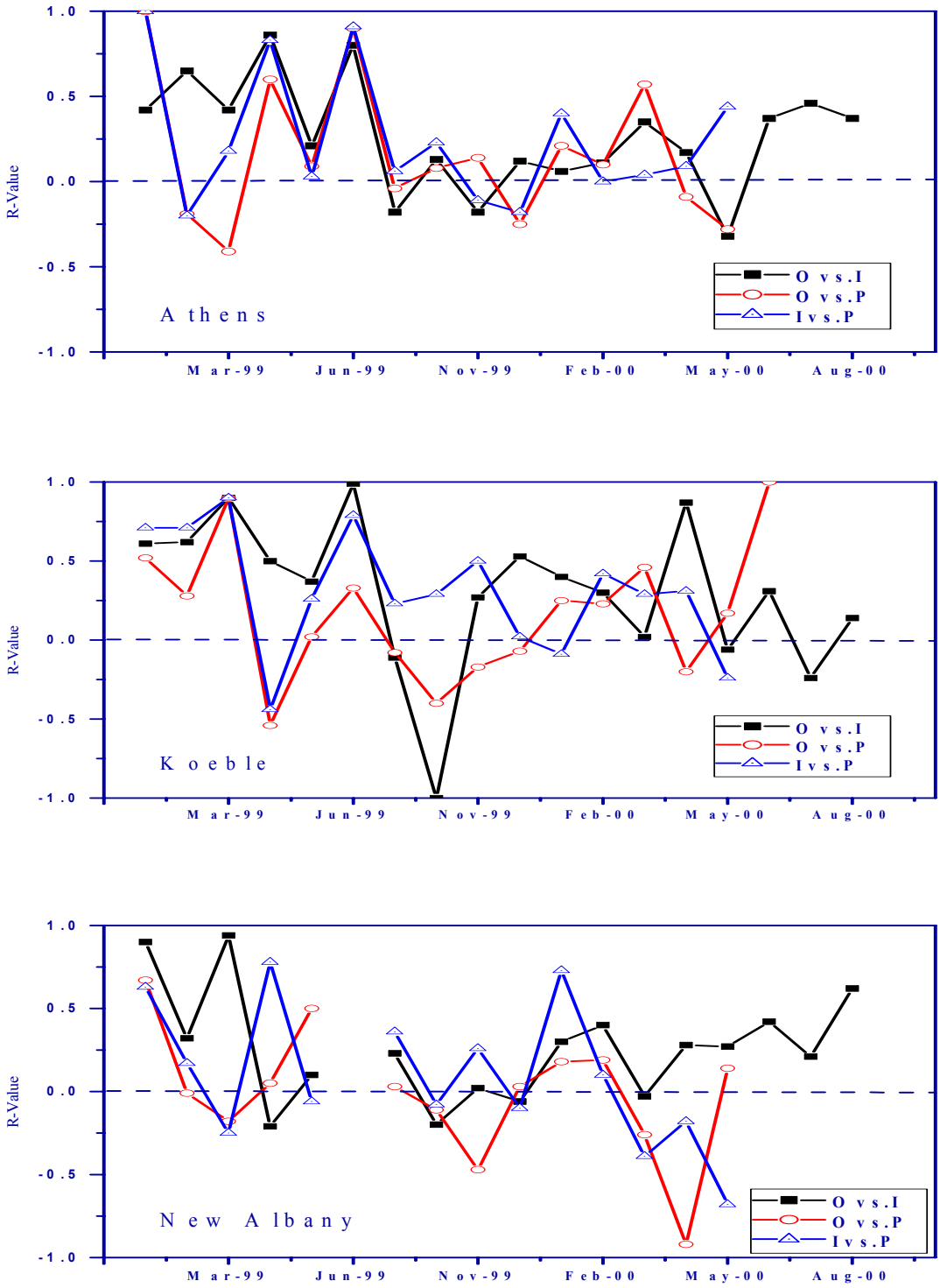


Figure II.1.18. Monthly variations of correlation coefficients of personal (P), indoor (I), and outdoor (O) concentrations at all three sites

Table II.1.12. Summary of correlations of personal and outdoor $PM_{2.5}$ concentrations ($\mu g/m^3$) by year and by season.

Personal vs. Outdoor	Intercept	Slope	R	N	P
1999					
Athens	-3.70	1.12	0.69	135	<0.0001
Koebel	6.15	0.60	0.37	139	<0.0001
New Albany	9.87	0.00	-0.01	138	0.94
2000					
Athens	28.88	-0.05	-0.02	68	0.90
Koebel	15.20	0.29	0.20	47	0.18
New Albany	22.85	0.04	0.03	62	0.83
Warm Months					
Athens	18.38	0.45	0.26	84	0.02
Koebel	14.14	0.46	0.23	65	0.07
New Albany	13.92	0.48	0.21	69	0.08
Cold Months					
Athens	9.71	0.21	0.07	119	0.48
Koebel	6.10	0.46	0.39	121	<0.0001
New Albany	11.08	0.01	0.01	131	0.89

Table II.1.13. Summary of correlations of indoor and outdoor $PM_{2.5}$ concentrations ($\mu g/m^3$) by year and by season.

Indoor vs. Outdoor	Intercept	Slope	R	N	P
1999					
Athens	3.86	0.58	0.74	156	<0.0001
Koebel	4.34	0.60	0.65	137	<0.0001
New Albany	5.45	0.77	0.54	150	<0.0001
2000					
Athens	26.78	-0.31	-0.16	136	0.05663
Koebel	19.52	0.02	0.01	103	0.88724
New Albany	16.50	0.19	0.13	115	0.17058
Warm Months					
Athens	20.09	0.03	0.02	154	0.76124
Koebel	15.54	0.28	0.16	111	0.10401
New Albany	8.31	0.53	0.43	137	<0.0001
Cold Months					
Athens	10.79	0.19	0.09	138	0.27813
Koebel	7.16	0.35	0.38	129	<0.0001
New Albany	14.40	0.29	0.16	128	0.06469

Table II.1.14. Summary of correlations of personal and indoor $PM_{2.5}$ concentrations ($\mu g/m^3$) by year and by season.

Personal vs. Indoor	Intercept	Slope	R	N	P
1999					
Athens	-3.59	1.27	0.65	131	<0.0001
Koebel	3.21	0.88	0.51	130	<0.0001
New Albany	7.47	0.17	0.27	137	0.00
2000					
Athens	10.04	0.64	0.59	65	<0.0001
Koebel	5.76	0.60	0.68	39	<0.0001
New Albany	16.46	0.31	0.25	45	0.10
Warm Months					
Athens	7.86	0.71	0.58	79	<0.0001
Koebel	12.23	0.35	0.32	54	0.02
New Albany	8.81	0.44	0.43	56	0.00
Cold Months					
Athens	-0.73	0.98	0.71	117	<0.0001
Koebel	1.03	0.99	0.69	115	<0.0001
New Albany	9.33	0.13	0.12	126	0.17

Factors Influencing Personal, Indoor, and Outdoor Correlations

Limited information is available regarding correlations between personal, indoor and outdoor $PM_{2.5}$ concentrations. Most studies focus on senior subjects and respiratory patients.[58] A few studies investigate children's $PM_{2.5}$ exposures and their relationships with indoor and outdoor levels.[59] The studies that focus on children are often conducted in homes, an environment quite different from the classroom. Therefore, making direct comparisons between this study and similar investigations is difficult. However, this study provides valuable information in examining the relationship between personal, indoor, and outdoor $PM_{2.5}$ levels. The main factors that influence the correlations are indoor $PM_{2.5}$, study design, and measurement bias.

Influ*ence of Indoor $PM_{2.5}$*

Personal $PM_{2.5}$ concentrations and their relationships to indoor and outdoor levels vary between studies, subjects, and environments. A majority of research evaluating personal, indoor, and outdoor correlations have indicated indoor $PM_{2.5}$ greatly influences personal exposures.[60] The influence of indoor $PM_{2.5}$ in these studies may result in higher indoor and/or personal concentrations than the outdoor levels, and stronger correlations for personal-indoor concentrations than personal-outdoor or/and indoor-outdoor correlations.[61]

In the PTEAM prepilot study conducted in Azusa, CA, mean 24-hr personal $PM_{2.5}$ exposure doubled the indoor (36.3 ± 2.6 μg/m^3) and the outdoor (42.6 ± 3.0 μg/m^3) concentrations.[62] For the 18 participants, pooled data showed personal concentrations were uncorrelated with outdoor levels (cross-sectional correlations). In the main PTEAM study (178 nonsmoking subjects, aged from 10-70), both indoor and personal were weakly correlated with outdoor concentrations.[63]

Study from Toronto, Canada also reported higher personal and indoor concentrations compared to ambient levels.[64] Median $PM_{2.5}$ concentrations for personal, indoor, and outdoor were 28.4, 15.4, and 13.2 μg/m^3 respectively. Pooled data for all subjects resulted in low personal-outdoor correlations (R = 0.19 to 0.27, P<0.01). However, strong agreement occurred on personal-indoor concentrations (R= 0.79, P <0.01), and the indoor-outdoor correlation was moderate (R = 0.21 to 0.33).

In EXPOLIS study conducted at Basel, Switzerland, Oglesby et al. documented higher 48-hr mean personal $PM_{2.5}$ concentration (23.7 ± 17.1μg/m^3) compared to outdoor concentration (19.0.7 ± 11.7μg/m^3).[65] Pooled data of 44 subjects (ETS and non-ETS) yielded poor personal-outdoor correlations (R = 0.07).

In Boston, Rojas-Bracho et al. found higher 12-hr mean personal (21.6 μg/m^3) and indoor (17.5μg/m^3) $PM_{2.5}$ concentrations than the outdoor levels (14.2 μg/m^3) for 17 adults.[66] The longitudinal correlation coefficients for these subjects were 0.61, 0.74, and 0.87 for personal-outdoor, indoor-outdoor, and personal-indoor correlations respectively.

This study supports much of the research described previously. Personal and indoor concentrations were higher than the outdoor levels with higher I/O ratios occurring during school days than non-school days. This study focused on 4th and 5th grade students who were generally active during the school day. Their exposures to ambient $PM_{2.5}$ were similar at each site and even similar inter-sites resulting from homogenous distribution of ambient $PM_{2.5}$. However, their indoor $PM_{2.5}$ exposures were highly variable. Personal $PM_{2.5}$ more closely correlated with indoor correlations than the outdoor levels due to the impact of indoor $PM_{2.5}$. While personal-outdoor correlations varied among sites and in different time periods, strong indoor-personal correlations occurred at all sites and all the time periods.

Influence of Study Designs

Study design is another reason for inconsistent correlations between personal, indoor, and outdoor PM concentrations. Generally, longitudinal studies result stronger personal-outdoor correlations than cross-sectional studies. In the prepilot PTEAM study, the cross-sectional personal-outdoor correlations (R^2 = 0~0.02) were lower than the longitudinal regressions in

which individual correlation was performed (R^2 = 0.01~0.58).[67] In some recent longitudinal-designed research, strong personal-outdoor correlations were found. For example, Williams et al. reported the R-value of daily average personal-outdoor correlation of 0.89 on subjects 72 to 93 years old.[68]

However, fluctuations of personal-outdoor correlations also take place in the longitudinal studies with better correlations occurring on subjects. Higher correlations in longitudinal studies are either due to choosing elderly and COPD patients who usually have low incidence of indoor sources, or to exclusion of data with high indoor source events. On the other hand, cross sectional correlations can also be strong in the absence of indoor sources. Tamura et al. found that pooled PM_{10} personal-outdoor correlations for 7 elderly adults in Japan were significant with R of 0.83.[69] These subjects were not exposed to apparent indoor sources. The data analysis also excluded days with high indoor source events (i.e. incense burning, smoking visitors). In sum, omitting indoor-generated sources from the study design and data analysis can strenghthen personal-outdoor correlations.

In general, this study was longitudinal because measurements over multiple days for the subjects were obtained. For daily personal monitoring, one subject was chosen randomly from the class. Although each subject tended to have similar activity patterns with the whole group, different personal exposure to nonambient PM sources existed due to discrepancies in individual's activity-pattern. Because of the large number of subjects involved and the length of the study, no activity diaries were maintained. This may limit the correlation analysis in differentiating the subjects from limited non-ambient $PM_{2.5}$ to those with high non-ambient $PM_{2.5}$ exposures, and result in poor to moderate personal-outdoor correlations.

Influence of Measurements

Other factors influencing correlations between personal, indoor, and outdoor $PM_{2.5}$ concentrations measurement protocols. These influences included measurement bias of sample devices used and measurement errors, including equipment malfunction and random errors. Following standard QA/QC procedures minimized measurement errors from the monitoring procedures. Refer to the *Quality Assurance Handbook* for more details on QA/QC procedures.

The possible reasons for measurement bias included the differences between monitoring devices, sampling times, sampling frequencies, and monitoring device locations. In this study, several factors minimized measurement biases. These factors include inlet type, monitoring schedule, monitor placement, inspection of the filters, and QA/QC procedures. The same inlet type was used for outdoor and indoor monitors. All sample types were monitored concurrently at each site and the monitoring devices were set up within the same building. New Albany, a special case, the outdoor $PM_{2.5}$

monitor resided on a building roof about 200 meters from the building where the indoor and personal $PM_{2.5}$ monitoring occurred). However, differences existed between monitoring devices used for personal, indoor, and outdoor measurement. More fibers or visible particles resided on personal sample filters than indoor and outdoor sample filters. It is possible the impactors, used in personal monitoring, collected some coarse particles due to low flow rate or to design capability. Moderate correlations between personal and indoor samplers and ACCU systems may contribute to the observed poor to moderate correlations.

Another common effect is the existence of "personal cloud." Personal cloud or personal-activity $PM_{2.5}$ is an increased personal exposure derived from personal measurements in comparison with a stationary monitors or a time-weighed average of indoor and outdoor concentrations. The cause of personal cloud is still unknown, however, since the chemical composition of personal cloud is similar to the indoor particles, personal cloud is related to indoor activity-generated sources. In comparing retirement centers to apartments, Rodes et al. suggested a personal cloud might be a result of "resuspended particles from carpeting, collection of body dander and clothing fibers, personal proximity to open doors and windows, and elevated PM levels in nonapartment indoor microenviroments."[70] Electrostatic effects which happen within a close vicinity of a subject to the sources may also contribute to the cloud. Although resuspension effects are more pronounced in coarse-model particles, increased $PM_{2.5}$ concentration in resuspension events (i.e. physical activity) were also recorded for small size subjects. The resuspension-related $PM_{2.5}$ increase in this study could be quite sizable due to large group (~30 subjects) involved. Studies show personal cloud as a major reason for the poor personal-outdoor correlations since the personal cloud for healthy persons could count for up to 50 $\mu g/m^3$ during their active period.

CONCLUSIONS

As part of the Air Pollution and Pediatric Health Impact Assessment (APPHIA) project, this section provides information on correlations between personal, indoor, and outdoor $PM_{2.5}$ levels and comparisons between the sites. $PM_{2.5}$ mass measurements occurred at three elementary schools in central and southeastern Ohio from January 1999 through August 2000. Outdoor, indoor, and personal $PM_{2.5}$ measurements were obtained at three locations representing a rural, urban, and suburban setting.

At all three sites, personal and indoor $PM_{2.5}$ concentrations exceeded outdoor levels. Personal $PM_{2.5}$ exposures were significantly affected by indoor $PM_{2.5}$, presumably the result of resuspension by human activity. The I/O ratios of $PM_{2.5}$ mass concentrations and of sulfate concentrations were greater than unity at all sites when school was in session. Lower I/O ratios associated with lower indoor sources were found during non-school days when the students were absent.

In addition, the personal-outdoor and indoor-outdoor correlations varied significantly throughout the study due to high variability of nonambient $PM_{2.5}$ caused by activity patterns and ventilation conditions. At all sites, indoor $PM_{2.5}$ concentrations moderately correlated with outdoor concentrations at significant levels with R ranging from 0.13 to 0.30. However, only moderate personal-outdoor correlations existed at two sites, Koebel and Athens. Weak personal-outdoor correlations occurred at New Albany. In comparison, there were strong personal-indoor correlations at all the sites.

The analysis of correlations between personal, indoor, and outdoor $PM_{2.5}$ are crucial in understanding the health effects in epidemiological studies. The central assumption of epidemiological studies is that ambient PM concentration, monitored by a fixed site, is a sufficient surrogate for personal exposure. In this study, ambient $PM_{2.5}$ distributed homogeneously throughout the study area. This indicates that ambient $PM_{2.5}$ concentrations monitored from a fixed site can represent the average ambient $PM_{2.5}$ level over a large area. However, high variations occurring with indoor and personal concentrations indicates that the ambient $PM_{2.5}$ may not be a strong indicator for indoor concentration or total personal exposures.

NOTES

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