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Aerosol particles generated by diesel-powered school buses at urban schools as a source of children's exposure

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Abstract

Various heath effects in children have been associated with exposure to traffic-related particulate matter (PM), including emissions from school buses. In this study, the indoor and outdoor aerosol at four urban elementary schools serviced by diesel-powered school buses was characterized with respect to the particle number concentrations and size distributions as well as the PM2.5 mass concentrations and elemental compositions. It was determined that the presence of school buses significantly affected the outdoor particle size distribution, specifically in the ultrafine fraction. The time-weighted average of the total number concentration measured outside the schools was significantly associated with the bus and the car counts. The concentration increase was consistently observed during the morning drop-off hours and in most of the days during the afternoon pick-up period (although at a lower degree). Outdoor PM2.5 mass concentrations measured at schools ranged from 3.8 to 27.6 μg m^{-3}. The school with the highest number of operating buses exhibited the highest average PM2.5 mass concentration. The outdoor mass concentrations of elemental carbon (EC) and organic carbon (OC) were also highest at the school with the greatest number of buses. Most (47/55) correlations between traffic-related elements identified in the outdoor PM2.5 were significant with elements identified in the indoor PM2.5. Significant associations were observed between indoor and outdoor aerosols for EC, EC/OC, and the total particle number concentration. Day-to-day and school-to-school variations in Indoor/Outdoor (I/O) ratios were related to the observed differences in opening windows and doors, which enhanced the particle penetration, as well as indoor activities at schools. Overall, the results on I/O ratio obtained in this study reflect the sizes of particles emitted by diesel-powered school bus engines (primarily, an ultrafine fraction capable of penetrating indoors).

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Traffic aerosol; PM2.5; diesel; school bus; outdoor; indoor

1. Introduction

Exposure to traffic-related particulate matter (PM) has been associated with various health effects, especially in children, including respiratory allergies, decreasedlung function, bronchitis, and exacerbation of existing childhood asthma (Brauer et al. 2007; Delfino et al. 2004; Gauderman et al. 2007; Gehring et al., 2010; Kim et al. 2004; McConnell et al. 2010; Morgenstern et al., 2008; Nordling et al., 2008; Ryan et al. 2005; Shima et al., 2002; Trenga et al. 2006). Diesel-engine exhaust is also listed as a potential carcinogen by the U.S. EPA (2002). Particles aerosolized by diesel-powered vehicles are mainly in the ultrafine size range (<0.1 μm), which makes them capable of penetrating the lower airways (Morawska and Salthammer, 2008a) and translocating through the blood-brain barrier (Block 2009).

A nationwide survey has shown that more than 30% of public schools in the US are located in close proximity (≤400 m) to major roadways, resulting in a potentially increased health risk (Appatova et al. 2008). In the Greater Cincinnati community 38.5% of public schools are located within 400 meters of major roads (Appatova et al. 2008). This distance also represents the area of elevated exposure to ultrafine PM (Martuzevicius et al. 2004; Morawska and Salthammer, 2003b; Reponen et al. 2003; Zhu et al. 2002a; Zhu et al. 2002b). A recent report by Richmond-Bryant et al. (2009), which established associations of PM2.5 and elemental carbon (EC) concentrations with traffic, idling, background pollution and meteorology during student dismissals at four New York City schools, singled out diesel idling and passing as significant factors contributing to EC and fine particles measured in the school vicinity.

More than 24 million American children are transported to and from school daily by over 600,000 buses most of which have diesel-powered engines (U.S. EPA 2008; Wargo et al. 2002). Children are also exposed to freshly emitted air pollutants when boarding and riding school buses. A recent case study demonstrated that school bus traffic significantly increases the total particle number concentration as well as the concentrations of diesel-associated elements in outdoor air in the school vicinity (Li et al., 2009). The effect is especially pronounced during arrival and dismissal times when dozens of buses may arrive and depart from the school within a short time. Temperature inversions, especially during the morning hours, and cold ambient air in winter may also increase traffic-related pollutant levels (Kim et al., 2002; Zhu et al., 2004; Ning et al., 2007). In cold weather, drivers often allow engines to idle for considerable time periods thus increasing emissions. Diesel engines emit 10 times more particles per mile than conventional gasoline engines and 30–70 times more than gasoline engines equipped with catalytic converters (OECD 1988). To reduce the exposure of children riding the school bus to diesel exhaust particles (DEP), the U.S. EPA supports programs such as the Clean School Bus USA. Exposures to DEP are also being reduced through nationwide and local efforts towards retrofitting school buses with diesel oxidation

catalysts, increasing the use of low-sulfur diesel fuel, and establishing anti-idling campaigns. These initiatives, however, are far from full implementation.

Although children's exposure to DEP generated by school buses has recently received considerable attention, there is still a lack of information regarding characteristics of aerosol in the school vicinity. Very few studies have addressed the outdoor air quality at schools whose student population is transported by "yellow buses." Additionally, there is a lack of data regarding the association between indoor and outdoor aerosol characteristics in schools. In order to address this need, a public health partnership was formed between the University of Cincinnati and the Cincinnati Public Schools and Cincinnati Health Department to develop and implement a community-driven anti-idling campaign. The objective of this report was to establish the characteristics of indoor and outdoor aerosol at urban elementary schools, which use diesel-powered school buses for transporting students prior to the implementation of the anti-idling campaign. The aerosols were characterized with respect to their PM2.5 mass concentrations and elemental compositions as well as the particle number concentrations and size distributions. The association between indoor and outdoor aerosol concentrations was investigated at each school with respect to the particle number concentration, PM2.5, elemental carbon (EC), organic carbon (OC) and other relevant elements. Factors such as the school proximity to major highways, school building characteristics, the number of diesel-powered school buses, and the number of other vehicles during drop-off and pick-up times were also considered.

2. Materials and Methods

Four urban public schools – further referred to as I, II, III, and IV – were selected for this study in four different neighborhoods of the city of Cincinnati, Ohio, USA. The schools were *a priori* chosen to maximize variability in exposure to buses and nearby major highways. As seen from Table 1, three of the school buildings were built in 2005 or later and one was built and renovated in 1967. Schools labeled I and IV were within <1000 m of several industrial operations and/or wastewater treatment facilities. It was noted that site II was the only school that kept the windows and/or doors open during a significant portion of the school day. The schools also exhibited differences with respect to the number of cars that dropped off the students daily. Enumeration of school buses and cars was conducted manually during drop-off and pick-up time periods each school day. The number of buses operating in the morning (drop off time) and afternoon (pick up time) was the same on all monitoring days for a given school. The car counts during drop-off hours were much more consistent that those during pick-up hours; therefore, the former was chosen as the representative parameter to quantify the daily number of cars. All school buses were diesel-powered, of a similar age, make, and model, and operated by the same company. According to the Federal Motor Vehicle Safety Standard, the buses operated at the tested schools belonged to the following categories: Type A-1, Type B, and Multifunction School Activity Bus (MFSAB) (Codes of Federal Regulations, 2004).

Indoor and outdoor air monitoring was conducted at each site for seven school days – two in the fall of 2009 and five in the spring of 2010. Each day, the monitoring began approximately 30 minutes before the first school bus arrived in the morning, and ended

approximately 15 minutes after the last school bus left the school in the afternoon. While dependent on the specific school schedule, the sampling period generally covered nine hours – approximately from 7 AM to 4 PM. The students and staff at schools performed their normal activities during the course of the study. In addition, some days featured non-routine indoor events that resulted in extensive resuspension of particles. Such events included the opening of bi-fold doors between the gym and stage, as well as increased activity in the cafeteria and gym due to holiday celebrations. These days were recorded and excluded in some analyses, e.g., indoor-to-outdoor correlations.

Sampling stations were generally placed in locations of students' activities to represent children's exposure. Specific locations were selected in a manner that minimized disruption of student activities and prevented students tampering with equipment. At schools III and IV, the indoor sampling stations were close to the auditorium stage/cafeteria/gymnasium areas while at schools I and II these were farther away from these areas. The outdoor stations were closer to car drop-off areas at schools I and IV than at schools II and III. The outdoor station at school II was in very close proximity to the bus drop-off/pick-up area; distances were greater for schools IV, I, and III. Generally, the bus dimensions and the bus line configuration affected the proximity and introduced considerable challenges in defining the distance between the "source" and the sampling station as "points."

Two portable condensation nuclei counters (P-Trak, Model 8525, TSI Inc., St. Paul, MN) operated in parallel, one indoors and one outdoors, were utilized to quantify the number concentration of aerosol particles throughout the sampling periods. These devices are capable of a non-size-discriminative real-time counting of airborne particles from 20 nm to >1 μm. The data were recorded as time-series. The time-weighted average (TWA) of particle concentrations was calculated based on the P-Trak data for the entire school day as well as much shorter periods of intense traffic activities: morning drop-offs and afternoon pick-ups.

In addition, a Wide-range Particle Spectrometer (WPS, MSP Corp., St. Paul, MN) was deployed on selected days (2–3 times per school) to determine the size distribution of aerosol particles during the morning drop-off and afternoon pick-up times. The particle size resolved data obtained by the WPS's differential mobility analyzer were recorded in 24 channels between 10 and 460 nm. The time resolution was up to 2.5 min, which is considerably longer than the one provided by P-Trak.

At each school site, four Harvard-type PM2.5 impactors (Air Diagnostics and Engineering, Inc. Harrison, ME) operated in parallel – two indoors and two outdoors. The Harvard impactor features a cut size of 2.5 μm at a sampling flow rate of 20 L min^{-1}. The particles were collected on 37-mm filters: one Teflon filter (Pall Corp., Ann Arbor, MI) for elemental analysis and one quartz filter (Whatman, Inc., Clifton, NJ) for carbon analysis. The impactors were calibrated before and after air sampling using a flow meter (DryCal DC-Lite, BIOS International Corporation, Butler, NJ). Each impactor was placed on an aluminum tripod at a height of 1.5 m from the ground and at least 2 m away from any obstruction (U.S. EPA, 1998). The Teflon filters were analyzed for PM2.5 mass concentrations using gravimetric technique and for mass concentrations of different elements (Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Ge, As, Se,

Br, Rb, Sr, Y, Zr, Mo, Pd, Ag, Cd, In, Sn, Sb, Ba, La, Hg, and Pb) using X-ray fluorescence (XRF). The quartz filters were analyzed for EC and OC mass concentrations with thermal-optical transmittance (TOT) using the NIOSH-5040 method. All analyses were performed by the Chester LabNet Inc. (Tigard, OR). Two blank samples were also collected and analyzed for each sampling location.

Meteorological conditions, including the wind speed and direction, have been obtained from the Hamilton County (Ohio) Department of Environmental Services on hourly basis. Construction on nearby roads and other non-routine activities were recorded. No air monitoring was scheduled on the days with a forecasted precipitation exceeding 6 mm and unusual activities at the tested sites.

Statistical analysis was performed using SPSS 11.0 for Windows and Microsoft Excel. Some elements were detected in quantities below the analytical uncertainty level. In these cases, the measured level was replaced with half of the uncertainty level reported by the analytical laboratory. Descriptive statistical parameters, such as arithmetic average and standard deviation were used to present the data on the indoor to outdoor ratios of PM2.5, EC, EC/OC and the total number concentration. Linear regression was used to examine the association between (i) the number of buses and cars servicing each school and a 3-hour TWA of the total number concentration of outdoor aerosol particles, (ii) PM2.5 and carbon, and (iii) indoor and outdoor concentrations of PM2.5, EC, EC/OC and the daily average of the total number concentration. Spearman correlation coefficients were calculated to examine the significance of correlations between different elemental constituents of PM2.5 variation. Associations between the particle total number concentration and the wind characteristics (speed and direction) were examined using a t-test. A p-value of <0.05 was considered statistically significant.

3. Results and Discussion

3.1. Size distribution and number concentrations of aerosol particles

Figure 1 shows representative examples (one per school) of the outdoor particle size distributions measured immediately prior to (dotted line) and during (solid line) the afternoon pick-up time period. Each curve represents a 2.5-min integrated snapshot. In spite of similarities, some differences were observed suggesting the influence of other –stationary and mobile – combustion sources in the areas. At all four sites, presence of school buses affected the outdoor particle size distribution in the submicrometer fraction. A remarkable increase was observed especially in the number concentration of ultrafine particles (<100 nm). Some distributions were pronouncedly bi-modal. These findings can be attributed to the diesel emission. Number concentration of diesel-exhaust particles have been shown to exhibit a bi-modal distribution with 98% of particles in the submicrometer range (ARB, 1997; Morawska, 2003b), including significant ultrafine fraction (Burtscher, 2005; U.S. EPA 2002a).

The most prominent peak representing ultrafine particles was found at school I with a notably greater concentration than at the other three schools (Fig. 1). This tendency was observed in both the WPS and P-Trak measurements. The particle concentration increase is

consistent with the number of school buses (and cars). Both were considerably greater at school I than the respective numbers at other schools.

Figure 2 presents the relationship between the TWA of the outdoor total particle number concentration (measured with the P-Trak during the morning period of the most intense bus traffic) and the bus and car counts at a given school. Solid squares represent the four schools monitored in this study. In addition, similar measurements obtained from an earlier study performed at another public school in the Cincinnati area (Li et al., 2009) are represented by a hollow square. This addition increases the sample size and extends the ranges of the average daily number of diesel engine buses (to 75) and cars (to 173). To be consistent with the quoted study, the TWA was calculated over a 3-hour period.

As shown in Figure 2, the total number concentration of the outdoor aerosol at schools during morning hours is significantly associated with both the bus and the car counts (R^2 = 0.85 and 0.90, respectively). The TWAs were calculated only for the morning drop-off periods. The afternoon periods were often less than representative, primarily because the afterschool activities generated additional aerosol particles on some days. In addition, the outdoor P-Trak had several failures that occurred during the last three hours of a daily session. As a sensitivity analysis measure, the associations presented in Fig. 2 were re-examined with the fifth point (taken from the Li et al. data) being excluded. After its removal, bus and car traffic were not anymore significantly associated with the total number concentration of outdoor aerosol particles. This likely reflects the limited sample size and variation in traffic at the four schools.

Day-to-day changes in the wind speed and direction were also analyzed as to their influence on the TWA daily variability for each school. No statistically significant associations (t-test: $p>0.05$) were found. This suggests that meteorological parameters such as wind characteristics have no major effect on the results presented in Fig. 2.

Figure 3 presents typical time series of the total number concentrations measured outdoor and indoor at each school (one example per site). Purposely, different dates were chosen for this illustration as compared to Fig. 1. It is seen that although the total number concentrations outdoors sometimes reached high levels (e.g., nearly 60,000 per cm^3 at school IV in the morning of February 24, 2010, and in some instances even higher), the concentration was much lower for the remaining part of the days. The concentration increase was consistently observed during the morning drop-off period and in most of the days during the afternoon pick-up period (although at a lower degree). In spite of the similarities, the time series recorded for outdoor aerosols were different at different sites on different days, which may be attributed to variations in the school bus traffic as well as the influence of other PM sources. Day-to-day and school-to-school differences were also observed in terms of how the indoor concentration followed the outdoor changes. For instance, while high peaks were recorded outdoors during morning and afternoon bus traffic at school I, the indoor number concentration minimally followed this trend (Fig. 3A). This may have occurred because the recently built building of school I had a particularly solid structure with no visible cracks and windows and doors closed throughout the day. The indoor concentration at school II showed some response to the change in outdoor particle count

consistent with the morning drop-off (Fig. 3B). School II also opened doors and windows between 11.30 AM and 12 PM on the day of April 9, 2010, which explains the increase of the indoor concentration and the subsequent overlap of the indoor and outdoor concentration levels shown in Fig. 3B. A pronounced peak in the outdoor particle number concentration (of approximately 45,000 particles per cm^3) was recorded in school III during the morning bus drop-off period (Fig. 3C). A less pronounced peak of nearly 30,000 particles per cm^3 was seen during the afternoon pick-up time. The difference in peaks may be attributed to morning rush-hour traffic along the neighboring highway. The highway traffic may also play a role in the consistently higher average concentration of around 20,000 particles per cm^3 throughout the middle of the day. On the other hand, the proximity to a major highway per se had a limited utility for assessing the influence of the highway on the ambient concentration measured at schools. The schools differed not only by the proximity to highways but also by the traffic intensity on these highways. For example, school II is the closest one to a highway (243 m) but the average daily numbers of cars and trucks on this highway are as low as about 17,000 and 300, respectively; in contrast, school IV is located at more than 2000 m from highway but the traffic on this highway is very intense (about 137,000 cars and 19,700 trucks daily).

The changes in the P-Trak-measured outdoor concentration shown in Fig. 3, might have, in principle, been caused by changes in the ambient wind speed and direction. To test this possibility, the wind speed and direction information was incorporated in the t-test. No significant association ($p>0.05$) was observed for any monitoring day or any school. This suggests that meteorological factors had no major influence on the hourly values of the total ambient number concentration. This finding should not be generalized because of a limited number of data points available from this pilot study for analyzing the temporal variation of the particle number concentration.

Indoor particle number concentrations generally followed a similar trend as the one measured outdoors. The negative spike in indoor concentration occurred at school III on March 2, 2010 at about 11.45 AM (Fig. 3C) due to changing the alcohol cartridge in the P-Trak. The outdoor concentration increase, observed at school IV during morning hours (Fig. 3D) may not be solely attributed to the school bus traffic but also to substantial metropolitan bus traffic in the area. The building of school IV is also well sealed, which explains why the outdoor concentration changes did not always significantly contribute to the indoor concentration. Generally, increased highway and commuter traffic during the morning may have masked the actual affects of school buses at III and IV. The indoor and outdoor TWA total concentrations were calculated for each school day, except for the days when the student indoor activity caused irregular increases in indoor concentrations, which exceeded the outdoor levels by a factor of 2 to 5.

We compared the total concentrations recorded by P-Trak to those calculated for the same conditions and time periods by integrating the WPS measurement data. In principle, it was not expected that these values would be equal because the instruments utilize different particle measurement principles and consequently enumerate particles in different size ranges [the WPS was used in a range of 10 to 460 nm (DMA only); the P-Trak's lower limit was 20 nm but the upper limit significantly exceeded the one of the WPS]. In most cases, the

differences between the P-Trak- and the WPS-measured total concentrations was about or below 20%.

3.2. PM2.5 mass concentration of aerosol particles, EC and OC components of PM2.5

Outdoor PM2.5 mass concentrations measured on single days across all schools tested in this study ranged from 3.8 to 27.6 μg m^{-3}. An arithmetic average calculated from seven daily outdoor concentration values obtained at each school ranged from 11.7 μg m^{-3} (at school II) to 17.8 μg m^{-3} (at school I). High outdoor concentrations at school I were expected due to the highest bus and car counts and the close proximity to stationary air pollution sources (Table 1). The daily-measured indoor concentrations for the tested schools ranged from 6.9 μg m^{-3} to 28.3 μg m^{-3}. The seven-day averaged PM2.5 indoor values per school ranged from 12.4 μg m^{-3} (at III) to 18.8 μg m^{-3} (at II). It is evident that the school order (from lowest to highest with respect to the PM2.5 concentration) was different for indoor and outdoor average values.

The daily-measured outdoor mass concentrations of EC across the tested schools ranged from 0.06 to 2.7 μg m^{-3} with the highest school average value at school I (0.83 μg m^{-3}) and lowest at II (0.23 μg m^{-3}). This is consistent with PM2.5 average concentrations. Indoor EC mass concentrations determined on single days in all schools fell between 0.06 and 1.5 μg m^{-3} , with highest school average measured inside school IV (0.37 μg m^{-3}) and lowest measured inside school II (0.14 μg m^{-3}). T he high indoor EC concentrations observed at site IV may be partially attributed to the close proximity of the cafeteria (where cooking took place throughout the morning hours). The low indoor EC concentrations at school II are consistent with low outdoor EC values measured at this school. It is also important to note that 31 EC samples (18 indoor, 13 outdoor) were found to be below the uncertainty level.

Outdoor OC mass concentrations measured daily at all schools ranged from 3.1 to 13.2 μg m^{-3}. Similar to many other studies (HEI, 2010), we found that OC accounted for a large fraction of ambient PM2.5. The OC averages per school were in a tight range: from 5.4 μg m^{-3} at school IV to 7.1 μg m^{-3} at school I. Indoor OC mass concentrations ranged from 7.1 to 14.1 μg m^{-3}. School II had the highest indoor OC average value of 11.2 μg m^{-3} (which is consistent with the PM2.5 data) and school IV exhibited the lowest one of 8.4 μg m^{-3}.

Figure 4 presents the relationship between outdoor EC and OC quantified from the outdoor PM2.5 filter samples and the total PM2.5. EC and OC were both significantly associated with PM2.5 ($p<0.05$ and $p<0.01$, respectively); however, these associations are characterized by relatively low R^2 values (0.19 for EC and 0.28 for OC). This suggests that there must have been multiple factors responsible for the observed carbon levels. With respect to EC, low R^2 values are not surprising because EC is present primarily in small (submicrometer) particle sizes and therefore contributes very little to the total PM2.5 mass. In contrast, the contribution of OC to PM2.5 is much greater; in addition, OC is a prominent constituent of the regional background aerosol. Thus, one could have expected a somewhat less scattered relationship between OC and PM2.5.

The levels of outdoor PM2.5 and OC measured in this study were somewhat lower than, but generally comparable, to those reported by Martuzevicius et al. (2004) for the Cincinnati

metropolitan area while EC levels were notably lower. At this point, we have not identified the reason of the EC decrease. Additional studies are being initiated in the area to determine whether it reflects a general reduction trend for specific ambient air pollutants over the last 6 years or can be explained differently. It is also noted that EC levels obtained in this study were mostly lower than the mean values measured in major urban areas such as New York, Berlin, Munich, and Osaka, as summarized in HEI (2010), but in line with the data reported by Kim et al. (2004) for San Francisco metropolitan area schools. Similar to outdoor EC concentration, the indoor EC concentrations measured in this study were slightly lower compared to levels reported for similar microenvironments in HEI (2010).

3.3. Relationship between mass concentrations of different elements comprising PM2.5

The relationship between elemental components of PM2.5 in the outdoor air were quantified by Spearman coefficients of correlation. Results for selected elements are presented in Table 2. The elements were chosen due to their association with diesel exhaust (HEI, 2002), traffic, or the regional background. Arsenic and Vanadium were excluded from the list after the XRF analysis revealed no results above their uncertainty levels. Statistically significant correlations were found for 47 out of 55 relationships. Mn, Fe, Zn, Pb, and Ti were all significantly correlated with each other, with the exception of Fe versus Pb.

The considerable number of significant correlation and relatively high values of Spearman correlation coefficients between traffic-related elements provides a fair indication of traffic emission sources in the outdoor air. OC demonstrated a high correlation with Si, Mn, and Fe. The data should be interpreted with understanding that OC is a prominent constituent of the background aerosol.

PM2.5 and EC demonstrated significant correlation with each other, and most of the selected elements. However, the PM2.5-based correlation exhibited different Spearman coefficients – from 0.383 for EC to 0.636 for S and 0.678 for Mn. It is likely because different elements are present in ambient aerosol particles of different sizes and the smaller particles have vastly lower contribution to the aerosol mass concentration than the larger ones.

In the Greater Cincinnati area, S is present primarily due to regional coal combustion products and has less prominent link to diesel-engine exhaust (Martuzevicius et al., 2004). This explains the low to medium correlations of S with most other elements.

Generally, the correlations between elements determined in this investigation are similar to the results reported by Martuzevicius et al. (2008) from their six-site study carried out near major highways in Cincinnati. At the same time, the referred paper listed higher Spearman correlation coefficients for several elements, which is explainable given that all sites in Martuzevicius et al. study were close to powerful traffic emission sources.

3.4. Relationships between outdoor and indoor aerosols represented by different particle indicators

Relationships between outdoor and indoor aerosols were studied represented by the following particle indicators: PM2.5, EC, EC/OC, and the total number concentration of particles. The results are shown in Figure 5.

No significant association between indoor and outdoor PM2.5 was found for the tested group of schools ($R^2 = 0.08$, $p = 0.16$, Fig. 5A). This may suggest that the penetration of particles predominantly contributing to the PM2.5 mass (relatively large) into the building shell was limited and/or a considerable amount of these particles were produced by indoor sources. Relatively high mass concentrations of PM2.5 were indeed sometimes measured near the cafeterias and gymnasiums when the doors to these rooms were open. At the same time, we acknowledge that during eight out of 28 days, non-routine activities occurred typically in these areas (cafeteria and gym) led to extensive resuspension of particles indoors. When data points representing these days were excluded, R^2 increased to 0.34 and p-value to 0.07.

Indoor mass concentrations of EC and EC/OC showed statistically significant associations with outdoor mass concentrations for all schools combined (EC $R^2 = 0.66$, $p < 0.01$; EC/OC $R^2 = 0.43$, $p < 0.01$). The data are shown in Figs. 5B and 5C, respectively. The R^2 values indicate that the linear model with outdoor EC and EC/OC explains 66% and 43% of the variability of indoor EC concentration and EC/OC, respectively. Exclusion of the eight days featuring non-routine indoor resuspension, had no significant affect on the identified indoor-outdoor associations of either EC or EC/OC determined for all schools combined. Calculated separately for specific schools, indoor EC was also found to be strongly associated ($p < 0.01$) with outdoor EC at schools II, III, and IV. The fourth school – number I – demonstrated association with a borderline significance of $p = 0.06$. The EC/OC indoor-to-outdoor relationship was found to be significant ($p<0.05$) at sites II and IV. The strong indoor-to-outdoor association was expected at school II due to open windows and doors. At school IV, the outdoor-to-indoor aerosol transport might also have been enhanced on some days when the external door from cafeteria (to outside) was open after 11.30 AM.

The indoor-to-outdoor relationship in terms of the total number concentration measured with P-Trak is shown in Fig. 5D. Unlike the filter samples, which were analyzed for PM2.5, EC, and OC, the particle counts recorded by P-Trak are predominantly influenced by the ultrafine fraction. Consequently, the particle number concentration is especially sensitive to activities generating aerosol in the area of monitoring. Therefore, when testing the indoor-outdoor association based on the number concentration data, we included in the analysis only 20 “routine” days (leaving out the eight days of extensive particle generating indoor activities. The above analysis demonstrated a strong association ($R^2 = 0.72$, $p < 0.01$).

Indoor-to-outdoor (I/O) ratios were calculated for PM2.5, EC, EC/OC and the total particle number concentration. PM2.5 I/O ratios ranged from 0.5 to 2.8. At school II, PM2.5 I/O ratio exceeded 1 on six out of seven days. This possibly suggests considerable contribution of indoor sources and efficient penetration of relatively large particles (prominently contributing to PM2.5) from outdoors. The latter is consistent with the observation that windows and doors were open at that school throughout most of the monitoring days, which enhanced the particle penetration. Two days of the fall sampling campaign at school III were also characterized by I/O>1. As the windows and doors were not routinely open at this school, high I/O values may have been due to maintenance and painting activities at the school site or cafeteria operations. Only one day at school I and two days at school IV produced I/O ratios above 1. Building IV had tighter structures as compared to II and III but

(as indicated above) the aerosol transport from outdoors might have been enhanced on the days when the external door from cafeteria was open.

The EC-based I/O ratios were mostly below 1. Only one day at school III and one day at IV produced EC I/O>1. Both cases are believed to be less than representative because the EC concentrations fell below the uncertainty level. The lower levels of indoor EC compared to outdoors is consistent with its outdoor source.

The EC/OC-based I/O ratio was below 1 in all cases. Given that EC/OC has been used as a marker of diesel emission (Burch and Cary, 1996), this is consistent with the fact that outdoor air was a primary source of diesel particles. Additionally, indoor EC and OC may have been produced in the cafeteria during cooking while OC is also pronouncedly present in the background aerosol.

The I/O ranges determined in this study for PM2.5, EC and EC/OC are generally similar to the corresponding ranges reported by Martuzevisius et al. (2008) for residential homes located in the Cincinnati metropolitan area. At the same time, the EC- and EC/OC-based I/O ratios appear to be lower. The building envelopes of the schools and homes tested in the present and quoted studies, respectively, as well as the local aerosol backgrounds at the monitoring sites were different, which limits the validity of the above comparison.

The I/O ratios in terms of the total particle number concentration were mostly below 1. Exceptions occurred on seven out of 28 days when monitoring stations operated in close proximity to the doors opened to either the cafeteria (school IV), or gymnasium (III), or outdoors (II). In cases when the indoor sampling results were affected by aerosol generated by high student traffic during recess and activities in the gym, the particle count indoors exceeded the outdoor counts. It has been acknowledged in the literature that the aerosol concentration at schools increases due to the student physical movement (Roorda-Knape et al., 1998; Stranger et al., 2008). Morawska et al. (2009) reported that various school activities, which were not associated with any outdoor concentration patterns, elevated indoor particle number concentrations, at times, to the levels up to 2 orders of magnitude above the outdoor levels. Blondeau et al. (2005), who studied the relationship between indoor and outdoor air quality in eight French schools, also found that aerosol generation inside occupied schools may have a major effect on the I/O ratio and the effect is especially pronounced for smaller airborne particles.

Overall, the indoor-outdoor relationships obtained in this study for different particle indicators suggest notable infiltration of aerosol particles originated outdoors to the school indoor microenvironments. At the same time, the I/O ratio is an overestimation of the infiltration factor of an air pollutant from outdoors to indoors (Bennet and Koutrakis, 2006) leaving a space for indoor sources, which may contribute significantly as discussed above. A comprehensive modeling involving the source emission rates, the air exchange rate and other factors may offer a more accurate estimation of the particle infiltration than a crude I/O ratio.

The relatively small sample size of this study posed limitation on utilizing the reported associations. If possible, a larger study should have a higher number of both the monitoring

days and tested schools given the unavoidable differences in sampling locations, locations and configurations of the bus and car drop-off and pick-up areas, as well as the building characteristics of the schools. Another limitation of this study is that the data were analyzed and interpreted using the wind speed and direction information available on a zip-code-specific basis, which might not always accurately represent the conditions at specific school locations. The air temperature may also play a role especially considering a particularly wide range – between 0°C at school IV in late February and 22°C at schools II and I in late April.

4. Summary and Conclusions

The indoor and outdoor aerosol at four urban elementary schools that use diesel-powered school buses was characterized with respect to the PM2.5 mass concentrations and elemental compositions as well as the particle number concentrations and size distributions. It was determined that presence of school buses affected the outdoor particle size distribution, specifically in the ultrafine fraction. The total number concentration of the outdoor aerosol at schools was significantly associated with both the bus and the car counts. The concentration increase was consistently observed during the morning drop-off hours and in most of the days during the afternoon pick-up period (although at a lower degree). In spite of the similarities, the time series recorded for outdoor aerosols were different at different sites on different days, which could be attributed to the variations in school bus traffic as well as the influence of other PM sources and the meteorological conditions. At the same time, no statistically significant associations between the total outdoor number concentration of aerosol particles and the wind speed and direction were found in this study. Day-to-day and school-to-school differences were also observed in terms of how the indoor number concentration followed the outdoor changes.

Outdoor PM2.5 mass concentrations at schools ranged from 3.8 to 27.6 μg m^{-3}. The highest average concentration was recorded at the school with the highest number of operating buses. Outdoor EC and OC mass concentrations were also the highest at that school. Although the EC-PM2.5 and OC-PM2.5 associations were both significant ($p<0.05$ and $p<0.01$, respectively), these relationships were characterized by relatively low R^2 (0.19 for EC and 0.28 for OC). The latter can be explained given that carbon is present primarily in the submicrometer particle size range, which provides a relatively small contribution to the total PM2.5 mass concentration.

Among elements chosen due to their relevance to diesel exhaust, traffic, or the regional background, statistically significant correlations were found for 47 out of 55 relationships. The considerable number of significant correlation and moderate-to-high values of correlation between traffic-related elements indicate a prominent presence of traffic emission sources in outdoor air at schools.

While no significant association was found between indoor and outdoor PM2.5 concentrations for the tested group of schools, significant relationships were identified between indoor and outdoor aerosols in terms of EC, EC/OC, and the particle number concentration. These results, as well as analysis of data on I/O ratio obtained in this study,

are consistent with the fact that diesel-powered school bus engines emit mostly ultrafine particles, which can efficiently penetrate indoors.

We believe that the data generated in this investigation will be found useful for epidemiological studies designed to estimate the aerosol exposure of school children and develop appropriate air quality control strategies in schools.

Acknowledgement

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> In this study, the indoor and outdoor aerosol at four urban elementary schools serviced by diesel-powered school buses was characterized with respect to the particle number concentrations and size distributions as well as the PM2.5 mass concentrations and elemental compositions. > The presence of school buses significantly affected the outdoor particle size distribution, specifically in the ultrafine fraction. > Significant associations were observed between indoor and outdoor aerosols for EC, EC/OC, and the total particle number concentration.

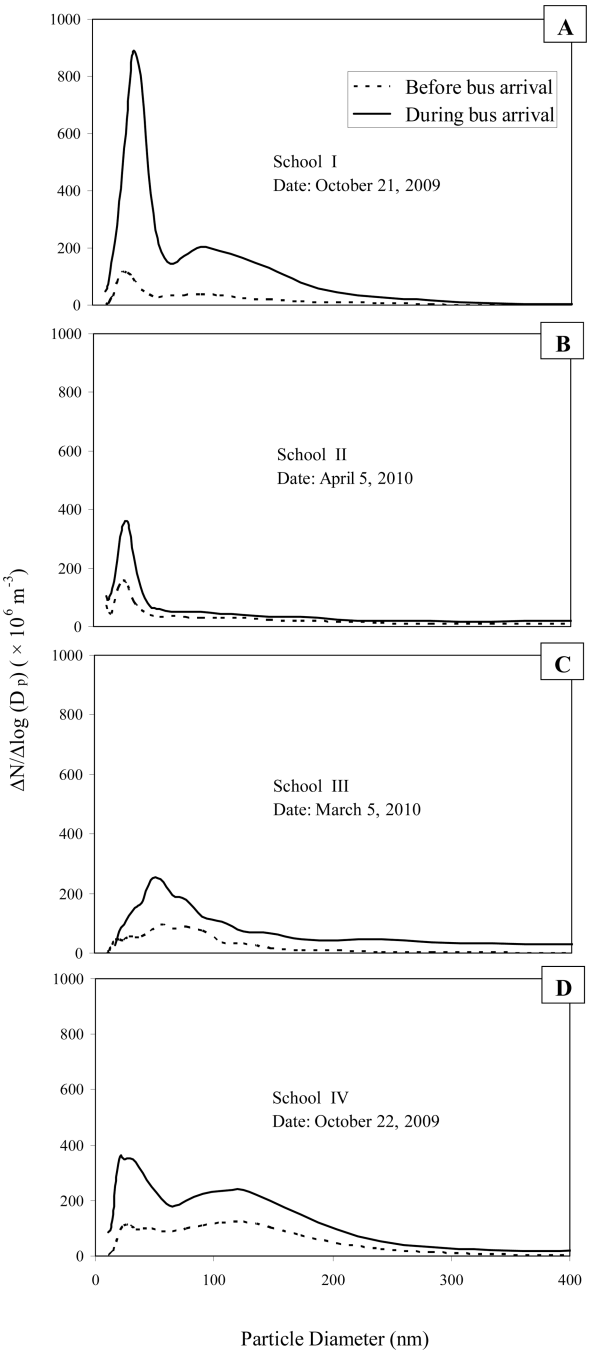

Figure 1.

Representative examples of the particle size distributions of outdoor aerosol before and during an afternoon pick-up at four schools: I (A), II B), III (C), and IV (D). Each curve represents a 2.5-min integrated snapshot measured with a WPS operating in the DMA mode.

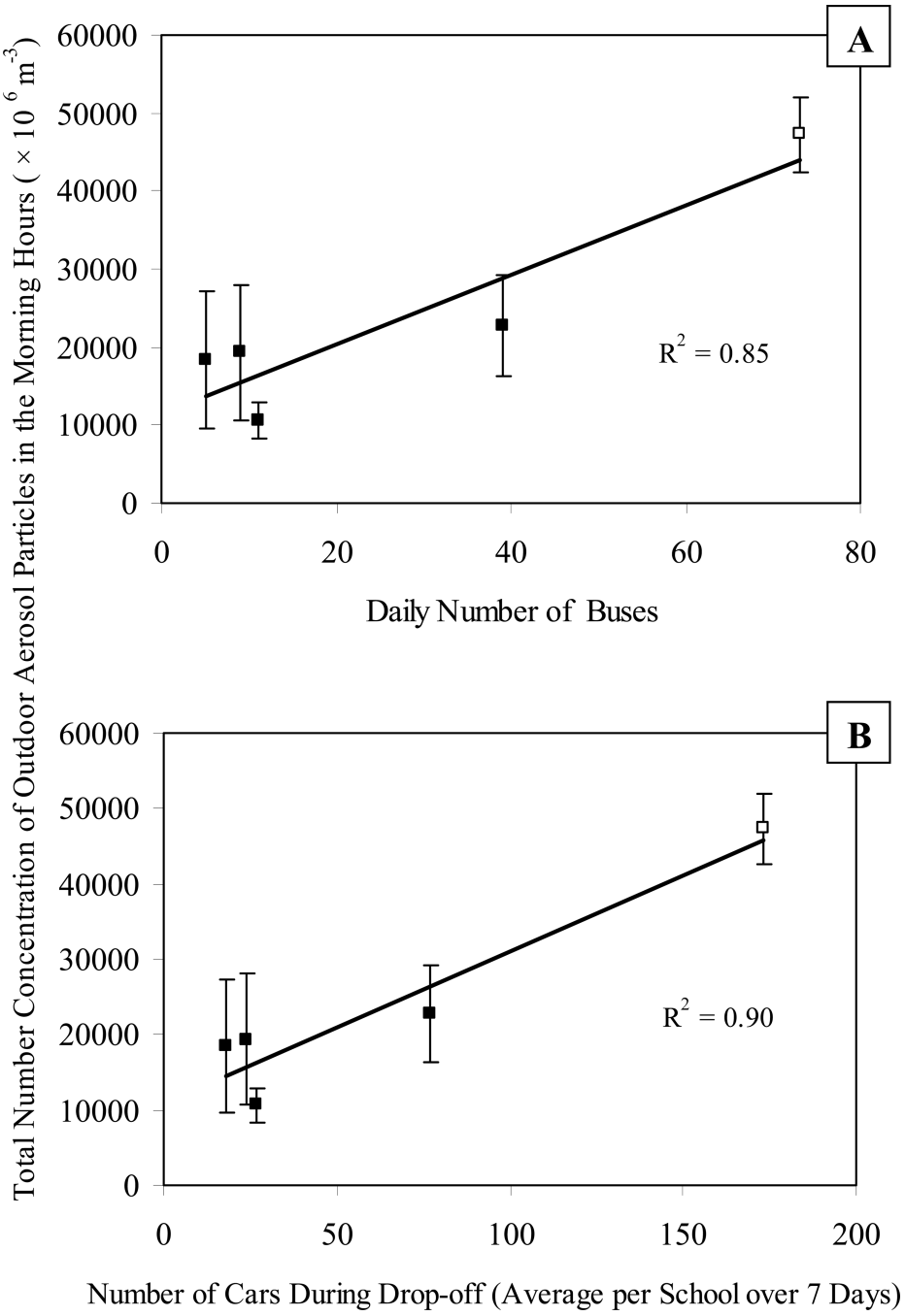

Figure 2.

Relationships between the total outdoor number concentration (a time-weighted average, TWA, over three morning hours) at each school and two independent traffic counts: the number of buses (A) and cars (B) dropping off students. Each point represents an arithmetic average of the TWA values determined from the data recorded by a P-Trak on different days, and the error bars represent the standard deviation. The fifth point (hollow) on each graph represents the measurements reported in the study by Li et al. (2009) conducted at a different school in the Cincinnati area.

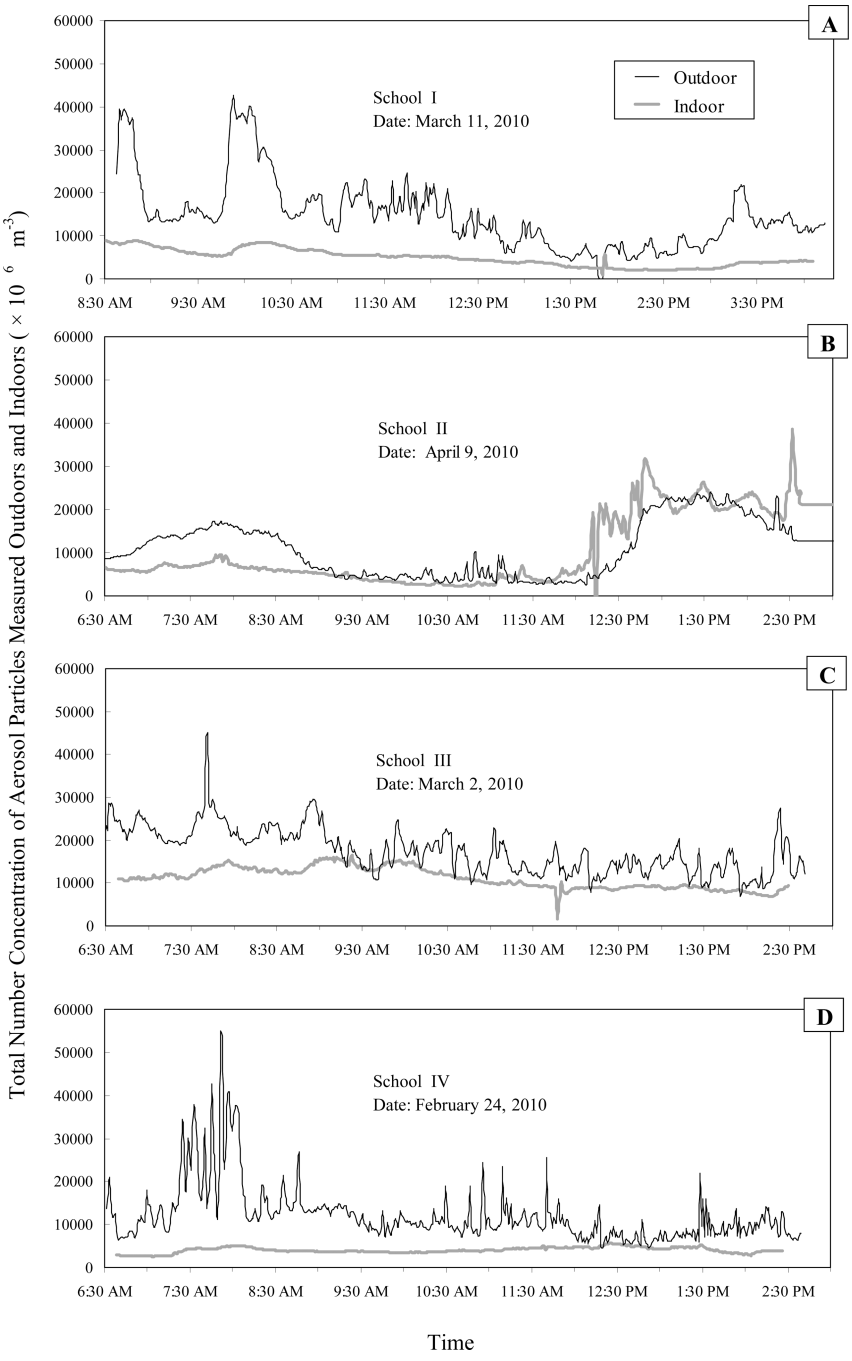

Figure 3.

Typical time-series representing the total particle number concentration outdoors and indoors at four schools: I (A), II (B), III (C), and IV (D) recorded with P-Traks.

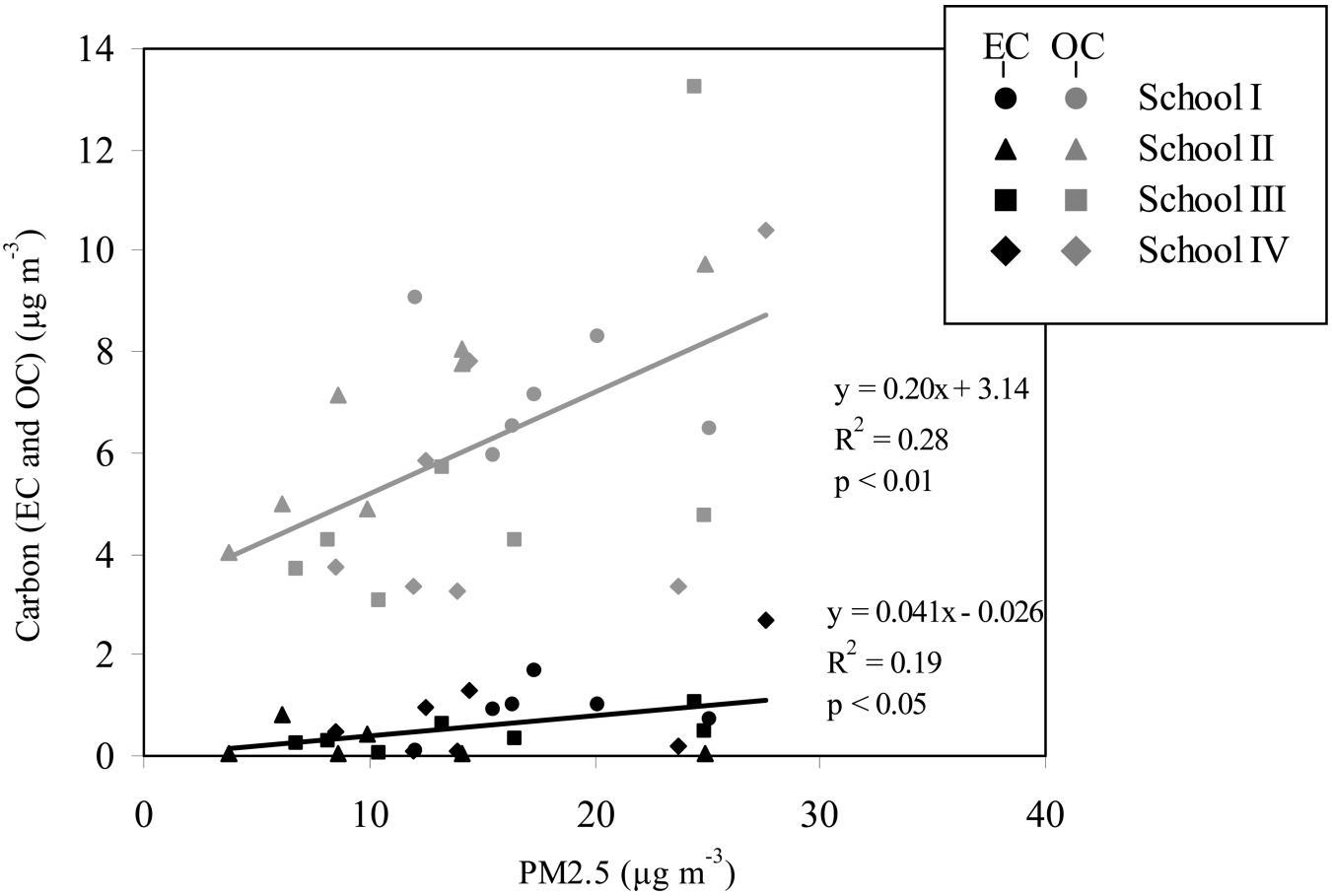

Figure 4.

Relationships between the mass concentration of carbon (EC and OC) quantified from the PM2.5 fraction and the total PM2.5 concentration. Each point represents a sample collected with a Harvard impactor during a school day (approximately 9 hours).

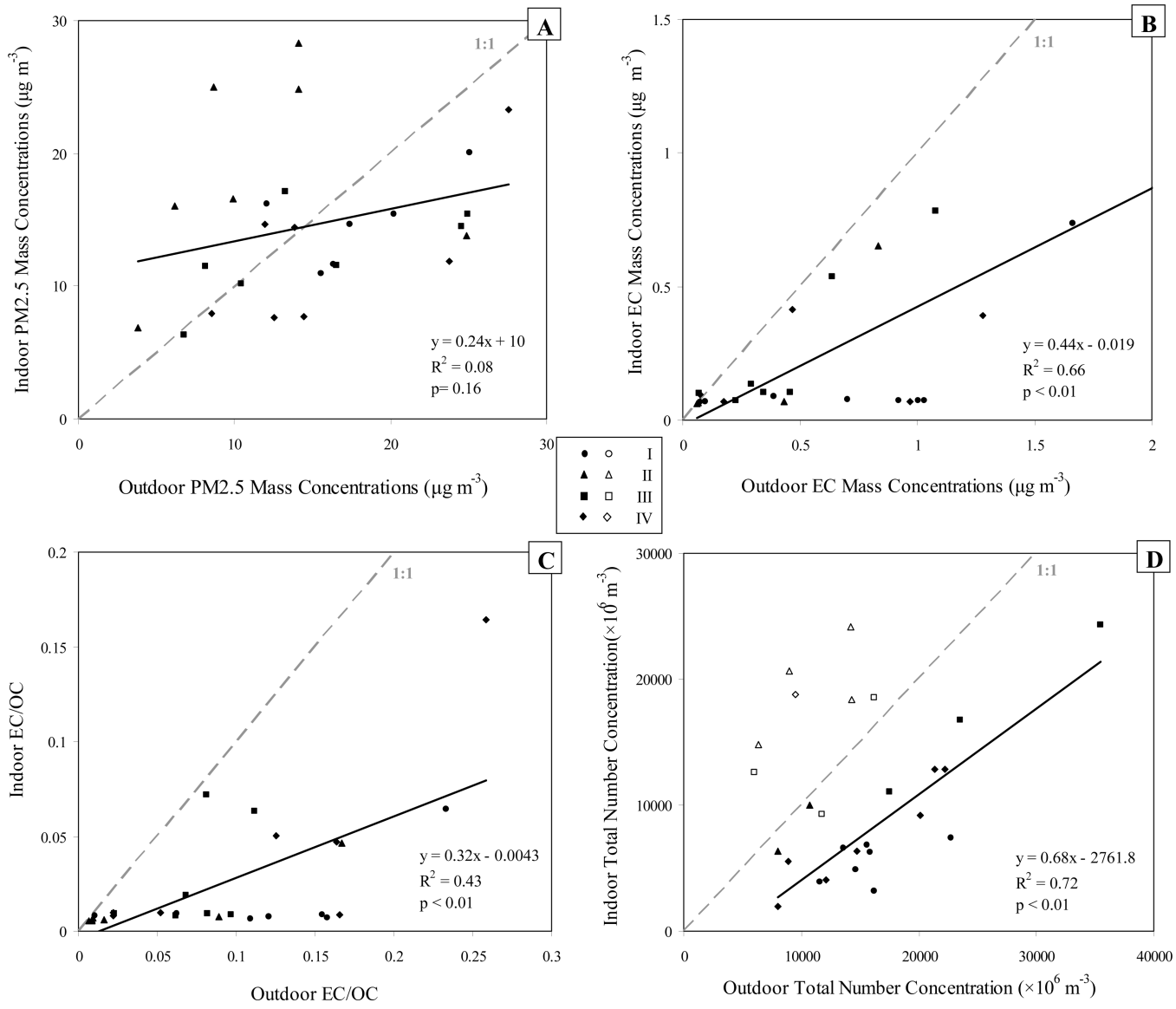

Figure 5.

Relationships between the indoor and outdoor aerosols in terms of PM2.5, EC, EC/OC, and the total number concentration of particles. Each point represents a daily TWA measurement. Linear regression and p-values were calculated for all monitoring days, except in figure D, where days with non-routine activities (hollow symbols) were excluded from the regression analysis. A dotted line corresponds to 1:1 ratio.

Table 1

School characteristics and location of sampling stations

School Characteristics	School I	School II	School III	School IV
Year built	2007	1962 (renovated 1967)	2007	2005
Distance to highway (m)	526	243	303	2083
Proximity to major stationary air pollution sources	Close	Far	Far	Close
Number of buses operating	39	11	5	9
Average number of cars during drop-off hours	77	27	18	24
Windows/doors mostly open	No	Yes	No	No
Location of Sampling Stations				
Outdoor: proximity to car drop off area (m)	20–50	75–100	50–200	25–40
Outdoor: proximity to bus drop off area (m)	40–100	10–30	50–200	25–40
Indoor: proximity to cafeteria/gymnasium (m)	75	100	< 10	< 10

Table 2

Correlations between PM2.5 elemental components in outdoor air

	EC	OC	Si	S	Mn	Fe	Zn	Br	Pb	Ti	Cu
PM2.5	**0.383**	**0.481**	**0.559**	**0.636**	**0.678**	**0.634**	**0.479**	**0.619**	0.257	**0.451**	**0.459**
EC		0.311*	**0.431**	0.128	**0.683**	**0.573**	**0.602**	**0.719**	**0.714**	**0.791**	**0.806**
OC			**0.772**	0.281	**0.550**	**0.716**	0.321	0.167	0.056	0.378	0.269
Si				0.257	**0.656**	**0.794**	**0.495**	0.250	0.177	**0.583**	**0.451**
S					0.213	0.160	0.023	**0.444**	0.031	0.000	0.071
Mn						**0.838**	**0.616**	**0.531**	**0.482**	**0.714**	**0.780**
Fe							**0.659**	**0.393**	0.374	**0.748**	**0.741**
Zn								**0.526**	**0.731**	**0.576**	**0.690**
Br									**0.576**	**0.544**	**0.617**
Pb										**0.530**	**0.698**
Ti											**0.762**

The values denote Spearman coefficients of correlation (n=27). Significant correlations ($p<0.05$) are marked bold.

*n=28