DEARS Particulate Matter Relationships for Personal, Indoor, Outdoor, and Central Site Settings for a General Population

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ABSTRACT

This analysis provides the initial summary of $PM_{2.5}$ mass concentrations relationships for all seasons, and participants for a general population in the Detroit Exposure and Aerosol Research Study (DEARS). The summary presented highlighted the utility of the new methodologies applied in addition to summarizing the PM data.

Results include the requirement to adjust the exposure data for monitor wearing compliance and measured ETS levels, even though the study design specified a non-smoking household. A 40% wearing compliance acceptance level was suggested to be necessary to provide a balance between minimizing exposure misclassification (from poor compliance) and having sufficient data to conduct robust statistical analyses. An ETS threshold level equivalent to adding more than 1.5 μ g/m³ to the collected sample was found to be necessary to detect changes in the personal exposure factor (F_{pex}). It is not completely clear why such a large threshold level was necessary.

Statistically-significant spatial $PM_{2.5}$ gradients were identified in 3 of the 6 DEARS neighborhoods in Wayne County. These were expected, given the number of strong, localized PM sources in the Detroit metro area. Some residential outdoor bias levels compared with the central site at Allen Park exceeded 15%. After adjusting for ETS biases, the outdoor contributions to the personal exposure were typically larger by factors ranging from 1.75 to 2.2 compared with those of the non-outdoor sources. The outdoor contribution was larger in the summer than the winter, which is consistent with the fractions of time spent outdoors in the summer versus winter (6.7% versus 1.1% of the time).

Mean personal PM_{2.5} cloud levels for the general population DEARS cohort ranged from 1.5 to 3.8 (after ETS adjustment) and were comparable to those reported previously. The personal exposure collections indoors were typically at least a factor of 13 times greater than those contributed outdoors.

1. INTRODUCTION

Background - The National Academy of Science (NAS) continuing review of research priorities for particulate matter (PM) (NAS, 2001) concluded that substantial progress was being made in assessing "Outdoor Measures versus Actual Human Exposures". Key observations in NAS (2001) from recent literature were that a) significant advances had been made in the abilities to assess PM personal exposures for adults, b) that ambient concentration variability is indeed a key component of measured personal exposures, c) continued linkages of aerosol of ambient origin with personal exposures "is a relevant metric for public health", d) an incomplete understanding still exists between outdoor measures and personal exposures for a range of study populations. These findings suggested that research was still needed in several critical areas related to personal aerosol exposure characterization that addressed improving personal exposure sampling methodologies, utilizing these methods for both sensitive subpopulations as well as general populations to determine differences, and developing refined assessment and modeling tools to better relate

outdoor PM measures with actual personal exposures.

The Detroit Exposure and Aerosol Research Study (DEARS), (Williams et al., 2008) was specifically designed to address the NAS concerns for PM. and applying these technologies in a general population representing a major US metro area. The DEARS design filled key knowledge gaps by developing better exposure characterization procedures that could be directly linked to subsequent receptor modeling efforts,, applying the latest exposure technologies for PM to a general population in a major US metro area, and collecting exposure samples and applying constituent analyses that would allow robust associations across personal, indoor, residential outdoor, and central site locations. PM data analyses and modeling would develop predictive relationships spatially and temporally. Receptor samples and associated chemistry (Landis et al., 2001) will eventually allow robust association between PM exposure in all settings with Detroit metro area source categories. Conducting the DEARS in the Detroit (MI) metro area provided a wide range of source categories with high emission rates, and the potential for developing robust data bases to support concurrent future exposure and panel studies in the metro area (Williams et a., 2008).

Personal exposure characterizations are preferable to fixed-location measures since they are more representative of true breathing zone exposures (Rodes and Thornburg, 2005). The added complexity and cost in making personal exposure measurements provides benefit in that the data produced are less biased and more representative, resulting in expected strong associations between exposures and adverse health outcomes. But Rodes and Thornburg (2005) note that burdensome personal exposure monitors will not always be worn according to study protocols, with general population participants the most problematic. They also note that unworn personal monitors produce biased exposure assessments and can exhibit substantial exposure misclassification error.

The latest US EPA PM Criteria Document (USEPA, 2004) summarizes the relevant literature through 2003, with many additional adverse impact studies reported in the literature between 2003 and 2008. A large proportion of the cited epidemiologic and toxicity studies in USEPA (2004) address the PM_{2.5} size fraction, but recent emphases are now additionally reporting adverse outcomes from exposures to the coarse fraction (PM_{2.5-10}). The review by Brunkreef and Forsberg (2005) clearly showed that many of the adverse health outcomes previously attributed almost solely to fine particles, have also been reported for particles >2.5 μm. They also suggest that complex mechanisms exist for coarse particles, where inflammatory effects from constituents such as endotoxins may pre-dispose pulmonary response to other coarse particles. A heightened importance of personal exposure assessment for these complex mechanisms was reported by Rabinovitch et al. (2005) who found that statistically significant worsening of children's asthma symptoms by exposures to endotoxin could only be detected by personal exposures. Neither concurrent indoor or outdoor monitoring showed associations. The researchers concluded that the resuspended "personal cloud" (Rodes et al., 1991) from childrens activities contributed significantly to the observed associations.

But, key EPA regulatory issues are driven heavily by mortality and morbidity issues

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(USEPA, 2004) reported from a range of epidemiologic studies linking ambient aerosols to adverse cardiovascular and pulmonary health outcomes. This focus on the elevated toxicological influences of ambient aerosols is still consistent with the fact that personal exposures are the most representative of breathing zone exposures. But the interpretation of personal mass concentrations alone are now recognized as only part of the picture (Williams et al., 2003) in understanding the linkages between PM exposures and adverse outcomes. The application of receptor modeling (e.g. Landis et al., 2001) can address ambient aerosol contributions or estimating techniques using surrogate ambient markers that penetrate into residences such as sulfate (Wallace and Williams, 2005), can identify the contributions of ambient particles and all other sources to total personal PM exposure.

Previous personal aerosol exposure studies for elderly and/or compromised participants have focused on identification of the levels of individual personal clouds using combinations of fixed-location indoor and outdoor monitoring in parallel with the personal assessment to somewhat crudely identify the elevation in personal levels that can't be explained by the fixed metrics. Rodes et al. (2001) reported mean personal clouds of 3.0 $\mu g/m^3$ for PM $_{2.5}$ and 19.9 $\mu g/m^3$ for PM $_{10}$ for elderly in three non-smoking retirement center settings and attributed these excess exposures primarily to resuspension of dusts while walking along with other personal activity aerosol sources. Williams et al. (2003) reported a comparable mean personal cloud in an elderly cohort with 730 observations that included individuals with implanted defibrillators of 4.2 $\mu g/m^3$. These personal clouds represent the elevated total personal mass collections that would not be predicted by simply measuring the residential indoor and outdoor settings and weighting by the time spent in both settings.

Personal cloud assessment reflect particle contributions that were not necessarily composed of aerosol from the ambient air, but more likely come from indoor-generated PM sources. The potential that some of the aerosol may contain inflammatory co-factor constituents such as endotoxin (Brunkreef and Forsberg, 20054) warrants the characterizations of both ambient and non-ambient aerosols in PM exposure studies. Methodologies for doing this have been presented by a number of researchers including Wallace and Williams (2005) and Williams et al. (2003). On the premise that there are few significant indoor sources of sulfur, they utilized ratios of sulfur content (indoor to outdoor, personal to outdoor) to provide reasonable estimates of infiltration rates and exposure to aerosol of outdoor origin. However, they also noted that the distribution of sulfur within the 0 to 2.5 μm size range is not uniform, and some indoor sulfur sources do exist. Since the DEARS design placed strong importance on identifying, understanding, and modeling the sources of concurrent contributions from particles of outdoor and indoor origin, these latter exceptions need to be examined further.

Objectives - This paper is one of a planned series of analyses to examine, model, and report the key aspects of the DEARS PM data characterizing the exposures of a healthy, adult general study population and attempting to link them to sources. A key focus is characterizing the PM exposure levels in non-smoking households by scenario (personal versus indoor versus outdoor), by participant, by summer versus winter

season, by Detroit exposure monitoring area (EMA), and in a limited manner, relative to central site versus non-ambient source category contributions. While understanding the representativeness of central site monitoring is important for the current paper, it is the central focus and addressed in depth in a separate paper (Williams et al. 2008). The general population nature of the DEARS resulted in quantified levels of study protocol violations for both personal sampler wearing compliance as well as maintaining a non-smoking household during the sampling periods. Including measured compliance levels and environmental tobacco smoke (ETS) contributions to personal exposures allows for the first time an assessment of the impacts of these confounders on key exposure metrics. Prior studies addressing modeling of both personal exposure and residence infiltration factors (e.g. Wallace and Williams, 2005) and personal clouds (Rodes et al., 2001) did not consider the biases potentially imposed by these confounders. The present analysis examines the DEARS PM relationships considering these factors.

A wide range of supporting DEARS-related papers examining PM component areas will be reported separately, including identifying distributional statistics (e.g., medians, 99th percentiles) across all DEARS participants and seasons for PM_{2.5} and PM_{10-2.5} (Williams et al., 2008), summarizing PM_{10-2.5} sampling methodologies and spatial variability in Detroit (Thornburg et al., 2008), influence of personal PM sampler wearing compliance bias issues, and identification of ETS confounding levels for PM samples. While some overlaps in data analyses (e.g. spatial PM variability across the Detroit metro area) are included in the present paper for completeness, the modeling approaches applied are in general new.

This paper presents a summary of the DEARS personal, residential indoor, residential outdoor, and central site community PM data by season and metro area location within Detroit (MI). Data presented are limited to the PM_{2.5} fraction. Relationships between settings and components are provided to illustrate the levels of associations observed, with comparisons made to a similar study in the Research Triangle Park (RTP), NC area reported by Williams et al. (2003) and Wallace and Williams (2005). The present analyses, similar to these prior analyses, will focus on understanding the contributions of outdoor aerosol to personal exposures, compared with all other sources. The methodologies of Rodes et al. (2001) to examine the relative contributions of indoor and outdoor sources relative to the personal cloud will also be applied.

The DEARS design recognized the potential for excessive ETS occurrence in the general population, and applied the methodology of Lawless et al. (2004) to identify spurious contributions of ETS to each collected sample to allow adjusting for these contributions in the data analyses. An important objective of this paper is to expand the modeling of Wallace and Williams (2005) in defining the contributions of ambient aerosols to personal and indoor exposures, but allowing adjustments for the potential confounding from ETS.

2. METHODS

General - The DEARS study design and the personal PM exposure sampling and

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analyses methods are generally described by Williams et al (2008). Much of the DEARS PM methodology evolved from the earlier Research Triangle Park (RTP) Panel study described by Williams et al (2003). Importantly, Williams et al. (2008) provided that the PM data quality objectives (DQO's) and noted that these were all achieved for the DEARS, including data capture rates, precision accuracy. The DEARS DQO's were very comparable to those of the RTP Panel study, readily facilitating the intercomparisons of data between studies.

The RTP Panel study (Williams et al., 2003) was a much smaller effort by comparison to the much larger and more complex DEARS. For example, the total number of PM samples deployed/collected in RTP Panel was nominally 750, while for the DEARS it exceeded 9,600. In addition to the DEARS cohort being three times as large (36 for RTP Panel, 143 for DEARS), the DEARS sampling was conducted over 3 calendar years rather than the one year for RTP Panel. Another key difference was that the RTP (NC) metro area contained few strong PM sources, while the Detroit (MI) area of Wayne County contains many heavy manufacturing sources, spatially-heavy distributions of heavy duty diesel vehicles, and numerous high-traffic count freeways). This increased the probability dramatically that spatial gradients would more likely to be apparent in the DEARS data, with some households showing much stronger influences from selected source categories. While the residence selections for RTP Panel were primarily purposeful, the DEARS residences were randomly selected to represent specific Enumeration Monitoring Areas (EMAs) within Wayne County that tended to contain high proportions of specific source categories (see Williams et al., 2008). This added an important stratification level difference between the DEARS and the RTP Panel effort, such that comparing PM characteristics between and among EMAs, provides defacto relationships driven by the expected presence and absence of certain source influences. The primary (a priori) source categories for the DEARS were: EMA 1 -industrial, EMA 3 - heavy duty diesel, EMA 4 - traffic & industrial, EMA 5 - industrial, EMA 6 - traffic, and EMA 7 - regional background. The EMA 2 area was proposed, but never used. Subset residence selections, especially in EMA 6, attempted to select residences that were well-within 300 m of a roadway compared to those that were much further away.

DEARS PM Methodologies - The basic personal sampling approach for the DEARS utilized a versatile low-lint, nylon vest worn by each participant to which the PM samplers were attached or placed in pockets (see Figure 2-1). Example photos of the indoor and outdoor sampling systems are described elsewhere by Williams et al. (2008). The design included special shoulder padding and adjustments to allow the fit to be reasonably-tailored to each individual in distributing the weight across the shoulders. However, while effective in collecting sampling personal exposures in both summer and winter seasons (worn over coats), the vest weight at just over 2,000g (4.5 pounds) was sometimes unpopular with participants.

The PM methodologies utilized in the DEARS were described by Williams et al. (2008) and were generally-comparable to those used in the RTP Panel Study and described in Williams et al. (2003). Both studies utilized parallel personal, residence

indoor, residence outdoor (typically backyard), and ambient monitoring site locations to allow developing relationships across scenarios. The primary personal aerosol sizing inlet employed was the PM_{2.5} MSP Model 200 (MSP Corporation) low-flowrate sampler operating at either 2 or 4 lpm. Battery-operated personal sampling pumps allowed 24-hour collections at each location. Each participant also carried an active flow, PM_{2.5} pDR-1200 MIE (Thermo Electron Corporation) personal nephelometry system to provide real-time indications of peak levels that could be compared with the integrated PM_{2.5} collections. The nephelometry data will be reported in a subsequent paper. Parallel indoor PM collections were made in the most utilized room in the residence (as identified by the participant). The indoor, residential outdoor (backyard), and central site (ambient) collections reported here were made using 10 lpm PM_{2.5} Harvard Impactors (Air Diagnostics Inc.). Personal sampler wearing compliance was monitored with an activity sensor which provided fraction daily compliance levels for each personal sample, representing the proportion of time adhering to the waking hour wearing requirements (Williams et al., 2008).

While $PM_{2.5}$ personal exposures were conducted across all six DEARS summer and winter seasons, the application of new indoor and ambient monitoring technologies to additionally allow PM_{coarse} ($PM_{10-2.5}$) became available in seasons 4, 5 and 6, as reported separately by Thornburg et al. (2009). However, resources permitted deployment only at indoor and outdoor location, even though the inlet methodology is only slightly larger than the MSP 200 inlet.

PM Spatial Variability - A key objective of the DEARS was assessing whether the central (Allen Park) monitoring site in Wayne County provided accurate estimates of PM_{2.5} exposures for the Detroit metro area. The ability of central sites to represent population PM_{2.5} exposures has been examined in many prior analyses and was summarized in detail in the USEPA particulate matter (PM) criteria document (USEPA, 2004). The present analysis addresses these spatial associations in a very limited way for mass concentrations (only), as a separate focused paper planned to address both PM_{2.5} mass and species spatiality is being prepared to follow. The current analyses will apply simple statistics and linear models to relate overall EMA mass concentrations (personal, indoor, and residential outdoor) to the concurrent central site PM_{2.5} measures. This is the first PM exposure study that also allows adjusting the personal and indoor exposure levels for the presence of unexpected ETS, and biases inherent in not wearing the personal monitors faithfully according to the study protocol.

ETS-Corrected PM Infiltration Modeling - The DEARS integrated the non-destructive optical absorbance method of Lawless et al. (2004) to provide estimates of the contributions of environmental tobacco smoke (ETS) to each integrated filter collection. This added exposure characterization to the DEARS study design proved extremely important from the outset of field activities in Detroit, as the levels of compliance with the non-smoking household protocol requirement appeared substantial at times. The potential confounding of the DEARS study objectives by non-compliance

violations, if not detected and adjustments made, could be substantial. In general, the participants were cautioned when inappropriate activities were observed by the study field technician, but these cautions were sometimes not heeded. In these cases, the personal vest and the indoor monitoring stations were pulled from the participant and residence, leaving only the residential outdoor monitors. In several cases, the violations were not identified until after the participant had completed the sampling period. In several cases, the household was indeed non-smoking, but the participant was significantly exposed to passive smoking in other locations.

Paoletti et al. (2006) describes some of the contributions that ETS provides to exposure characterization during controlled room experiments. Of particular importance to the DEARS, they noted significant contributions to PM_{2.5} and PM_{10-2.5} mass, sulfur, and elemental and organic carbon. Typically, the confounding is eliminated by simply excluding data from filters observed to contain ETS aerosol. But this was only possible in gross cases (yellow filters) until the Lawless et al. (2004) method provided actual mass contribution estimates. But can some level of quantitative compensation be applied and modeled?

As a starting point, the compartmental models described by Wallace and Williams (2005) utilized the ratios of the sulfur contents of personal-to-outdoor (ambient) and personal-to-indoor $PM_{2.5}$ collections to define an "outdoor exposure factor", or F_{pex} .

$$F_{pex} = S_{pers} / S_{out}$$
 (1)

This model was applied to the RTP Panel study data on the premises that a) no strong sources of sulfur were present near participants or indoors, and b) that sulfur was uniformly founding across the $PM_{2.5}$ size fraction. These were reasonable assumptions for RTP Panel, but defining the validity for the DEARS study design proved to be challenging.

The Wallace and Williams model then defined the total PM exposure to be composed of ambient aerosol and a non-ambient aerosol components, or:

$$\mathsf{E} = \mathsf{E}_o + \mathsf{E}_{no} \tag{2}$$

where: ${\sf E}$ is the total personal mass exposure, ${\sf E_o}$ is the contribution of outdoor (ambient) sources, and ${\sf E_{no}}$ is the contribution of all other, non-ambient sources. They then defined ${\sf E_o}$ to equal the product of the measured total outdoor PM concentration, or ${\sf C_{out}}$, with the outdoor exposure factor, to result in:

$$E = C_{out} F_{pex} + E_{no}$$
 (3)

The non-outdoor portion, E_{no} in equation (2) is comprised of the sum of indoor generated aerosol with all other non-outdoor sources, and contributes to the personal

exposures only during the periods at home. The portion from indoor-generated is heavily influenced by cooking, while all other sources includes personal cloud aerosol, predominantly from resuspension while walking (Rodes et all, 2001). With smoking households or simple passive ETS exposures, E_{no} includes an E_{ETS} component that is was quantified during the DEARS, but often goes unmeasured. We additionally define an E_{pers} term that is the sum of $E_0 + E_{no}$ shown in equation (2), but that is adjusted to exclude E_{ETS} . The value of E_{no} adjusted for ETS is described here as E^*_{no} . An important consideration is how much mass E_{ETS} contributes to E, given the known toxicity of the ETS component and it ability to confound relationships between exposures and adverse health outcomes. Prior personal exposure studies not quantifying E_{ETS} may have inadvertently included ETS contributions such that $E \neq E_{pers}$.

The Outdoor Exposure Factor is related to the fraction of aerosol that infiltrates into a residence, or \mathbf{F}_{inf} ., by knowing the fractions of time spent indoors and outdoors:

$$F_{pex.} = f_{in} F_{inf.} + f_{out}$$
 (4)

This assumes that the participant's indoor exposures can be described by those that occur in the personal residence and are described by \mathbf{F}_{inf} . Wallace and Williams showed that while \mathbf{F}_{pex} . is expected to always be greater than \mathbf{F}_{inf} , across all RTP Panel study participants, the two terms were methodologically-indistinguishable, or \mathbf{F}_{pex} . $\approx \mathbf{F}_{inf}$. Since most individuals spend the great majority of their time indoors, the median Outdoor Exposure Factor based on the ratio of personal-to-outdoor sulfur for RTP Panel (0.54), was very close to the median ratio based on indoor-to-outdoor sulfur (0.58). They also expected \mathbf{F}_{pex} to vary similarly to the Air Exchange Rate (AER).

Applying these equations to estimating the contributions of outdoor aerosol to the total personal collections is a reasonable assumption when there are no strong sulfur or PM sources confounding either the sulfur composition measures that comprise F_{pex} , or add unexpectedly to the total personal PM mass collection, E. When significant passive ETS exposures confounds either the personal or indoor concentration measures, the amount of sulfur contributed is not completely clear (e.g. from literature data such as that by Hecht, 1999), but some sulfur contributions could be assumed. The sulfur addition would be an additive term in the determination of F_{pex} . In addition, the balance of the ETS contributes substantially to the total personal mass, such that the measured total personal, E, is actually composed of ETS and non-ETS components:

$$E = E_{pers.} + E_{ETS}$$
 (5)

with both E and E_{ETS} as measured quantities, and E_{pers} . computed by difference.

Thus, for significant ETS exposures such as would be expected in general population studies like the DEARS, equation (3) could be rewritten as:

$$E_{pers.} + E_{ETS} = C_{out} [S_{pers.} + S_{ETS})/(S_{out}] + E_{no}$$
 (6)

Regressing C_{out} against E with and without ETS correction would provide slope terms (F_{pex} and F^*_{pex} , respectively, where the star, *, denotes the ETS-corrected value) that differ by the addition of the ETS sulfur component to the numerator.

Since, ETS is not expected to confound outdoor levels, the ratio of personal-to-outdoor sulfur could only be expected to be biased high, over-estimating the fraction of aerosol attributed to outdoor sources. If the fraction of non-outdoor aerosol is computed by subtracting the estimated outdoor contribution from the measured total personal exposure, it should additionally be adjusted by deducting the ETS contribution (by the method of Lawless et al. (2004) to minimize misinterpreting the Eno portion as coming from expected indoor and resuspension sources. Studies without significant ETS confounding (including all of our previous exposure panel studies for elderly cohorts (Williams et al., 2003; Evans et al., 2000) could utilize equation (3). Studies such as the DEARS with significant levels of observed ETS confounding of both personal and indoor exposure collections, should consider equation (5).

Some compromises are required in incorporating compliance screening, since eliminating exposure data can quickly reduce the amount of available data and significantly reduce the number of data points. For the present analyses, it was decided to utilize a reasonable compliance level that still retained at least 50 to 60% of the personal exposure data. A compliance level of 0.40 (exposure samplers worn at least 40% of the time during waking hours) was selected as reasonable. Accordingly, all computations utilizing personal samples required a compliance level of at least 0.40 to be included. This addition to the procedures described by Wallace and Williams (2005) for compromised cohorts, was considered important to provide representative \mathbf{F}_{pex} data.

ETS- and Wearing Compliance-Corrected PM Personal Cloud Modeling - The computation of personal clouds (Rodes et al., 2001) provides an additional level of understanding as to how and where participants become exposed. The personal cloud (PC) is defined as the portion of the mass collection by a personal exposure monitor (E) that would not be predicted simplistically from the sum of indoor residence (Ein) and outdoor (Eout) contributions combined with data on the fractions of time spent at home (fin) and outdoors (fout). The basic model is shown in equation (x):

 $PC = E - (E_{out} + E_{in})$ (7)

The indoor and outdoor contributions are computed from the respective mean concentrations as:

 $E_{out} = C_{out} \times f_{out}$ $E_{in} = C_{in} \times f_{in}$ (8)

Several researchers have summarized the levels of personal clouds including those for $PM_{2.5}$ mass by Wallace and Williams (2005) describing the RTP Panel Study conducted in North Carolina for two compromised, non-smoking cohorts.

Confounding from ETS was not expected in that study design and parallel assessments of ETS contributions to the personal exposures was not considered. The general population nature of the DEARS study population suggested that even with a protocol requiring non-smoking household, the levels of ETS contribution to each personal and indoor exposure sample would be characterized by the method of Lawless et al. (2004). This would additionally allow the contributions of ETS (EETS) to the personal clouds be determined to place it into perspective with the expected indoor and outdoor contributions. Thus, the personal cloud including all contributions (PC) would be recomputed without the ETS addition as (PC*):

$$PC^* = E - (E_{out} + E_{in}) - E_{ETS}$$
(10)

An additional step taken to assure the most representative data, was incorporation of fractional wearing data during the computation of Equation (10). Selecting an acceptable compliance level for all personal exposure data (E) applied assures that the personal clouds are the most representative of periods when the monitors are actually being worn.

3. RESULTS

DEARS Participant Recruitment- Table 3-1 shows the numbers of participants recruited for each season by DEARS neighborhood and monitored for PM (and other contaminants). The overall target was 120 participants, with 40 to be recruited in seasons 1, 3, and 5 and enrolled to participate across two seasons longitudinally. A total of 38 of the nominal 40 were enrolled for seasons 1 and 2, with 40 of 40 enrolled for seasons 2 and 3. As noted previously (Williams et al., 2008), a total of 137 participants were ultimately recruited to adjust for second season dropouts. Phillips et al. (2008) describe that the DEARS recruitment process was very successful, given the inherent difficulties in recruiting and enrolling a general adult population with a complex and relatively burdensome personal exposure component. The rolling selection plan provided reasonable representation of the \underline{E} numeration \underline{M} onitoring \underline{A} reas (EMA's). All EMA's were active across all seasons, except for EMA 7 which was utilized for only the initial two seasons. Participants in this far upwind location (Belleville, MI) were sampled only in the initial two seasons to better focus the available resources on more sourceimpacted locations in Wayne County. Although 40 were enrolled in season 5, resource availability necessarily limited season 6 to only 28 participants. Two seasons were not monitored in EMA's 3 and 5 in favor of sampling additional residences that were within

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300m of a major roadway in other EMA's. Phillips et al. (2008) describes the DEARS study population in detail as well as the success and shortcomings of the recruitment and retention efforts for a general population combined with a relatively complex compliance protocol. The DEARS recruits were all adults ((>18 years of age), with women making up 77% of those actually enrolled (Williams et al., 2008).

Fractional Times by Microenvironment - Tables 3-2 and 3-3 summarize the arithmetic means and standard deviations for the fractions of time spent in selected microenvironments, by EMA and by season, respectively, for all participants. These results were summarized from the validated participant time-activity diaries. The mean fraction of time spent at home in the primary residence across all EMA's (Table 3-2) was 77.4% (range of 66.4% to 83.5%), with a fairly large relative standard deviation of 24.5%. Table 3-3 shows that the overall means for time spent at home for all summer and all winter seasons were nearly identical. The overall mean times spent outdoors were also consistent across EMA's, varying from 3.4% to 5.5%. Table 3-3 shows that, as expected, the fraction outdoors in the summer (6.7%) was substantially greater than the time during the winter seasons of 1.1%.

The fractions of time spent in transit were very similar across either EMA's or seasons for all participants (1.6 to 4.3%), with the mean summer and winter fractions being identical. The fractional time spent away from home was quite variable across EMA's, with the far upwind EMA 7 being substantially larger at 20.8% than the other EMA's (5.8 to 13.1%). This was attributed to the longer commuting times from Belleville required to work at locations in the Detroit metro area. Only 2 seasons of data were available for this EMA. The fractions of time spent in all other miscellaneous or mixed microenvironments (all indoors), whether by EMA or season were very similar and fairly small for all participants (2.6 to 5.3%).

DEARS PM Concentrations Across EMAs and Microenvironments - The mean PM_{2.5} mass concentrations for each DEARS EMA are shown in Table 3-4 for personal, indoor, residential outdoor (backyard), and central site locations. The ability to characterize both personal and indoor filter collections for the mass contributions expected from ETS also allows separate ETS-adjusted personal and indoor categories. The same categories are summarized for all EMA's by DEARS season in Table 3-5.

As expected and as reported for previous studies, for a given cohort (EMA), the relative magnitude of unadjusted (for ETS) levels in Table 3-4 for all EMA's are personal (P) > indoor (I) > Outdoor or Central site. Unadjusted P averaged 20.3 $\mu g/m^3$ compared with 18.7 $\mu g/m^3$ for indoor, and 16.4/16.6 $\mu g/m^3$ for outdoor/central site. While this is also consistent for the ETS-adjusted means for all EMA's, at least one EMA (4) shows that the ETS-adjusted personal levels are slightly less than those measured simultaneously indoors.

Across EMA's, the mean concentrations in Table 3-4 are variable as might be expected from the diverse general populations in each location. The far-upwind EMA 7 personal and indoor concentrations were significantly lower than any of those in the Wayne County EMA (1, 3, 4,5, or 6).

Across seasons, the mean concentration in Table 3-5 for Season 2 (winter) was significantly lower for personal and indoor than all other seasons. However, all other seasons are reasonably similar, with the composite summer and winter personal means nearly identical over the entire 3 year study period. The overall indoor winter means were significantly smaller than those for summer, with the ETS-adjusted indoor at 19.2 $\mu g/m^3$ in the summer, but only 14.4 $\mu g/m^3$ in the winter.

PM_{2.5} Exposures and Concentrations - The data in Tables 3-4 and 3-5 summarizing the mean PM_{2.5} concentrations by EMA's and seasons, respectively, did not provide any real surprises, except perhaps for the importance of adjusting for the potential confounding of ETS. The previously held notion that requiring non-smoking households was a sufficient requirement to limit excessive ETS confounding is not consistent with the findings here for the DEARS general population. The data summarized represent a concerted effort to stress to participants the importance of this requirement, as well as early terminations of personal and indoor components when repeated violations occurred - and confounding still happened. These violations were not frequent during the DEARS but occurred with enough regularity to strongly suggest that routine ETS personal and indoor monitoring for ETS levels should be considered mandatory for future exposure studies. Even with non-smoking household compliance, the levels of passive exposures outside the home were apparent, as a linear regression of indoor ETS levels against personal ETS exposures for all participants and study days showed a 30.2% bias (R² = 0.71), personal levels exceeding those indoors.

Figures 3-2 and 3-3 show the personal $PM_{2.5}$ exposure distributions for all seasons and participants, both uncorrected (E) and corrected (E_{pers}) for ETS, and applying two different levels of required wearing compliance - 0% and 60%. Included on all graphs is the same linear (log-normal) fit line that shows that the central portions of data on all graphs exhibit the same slope, deviating below ~ the 5th percentile, and above ~the 95th percentile. Importantly, as the required level of wearing compliance increases, the portion of the distribution above the 95th percentile increases, while the median values at all compliance levels are the same. Included on Figures 3-2 and 3-3 are distributions for the personal exposures to $PM_{2.5}$ aerosol of outdoor (E_O) and indoor (En=no) origin , which do not exhibit the tendency to deviate from log-normality above the 95th percentile.

DEARS Central Site Versus Residential Outdoor Concentrations - The comparability of the Allen Park central site in Wayne County to represent PM_{2.5} mass concentrations across the country were evaluated by comparison of the outdoor data for each EMA with concurrent central site values. An example plot for EMA 1 for all seasons is shown in Figure 3-1, along with the linear regression statistics. Table 3-6 provides the regressions statistics for all DEARS EMA's, with statistical slope differences from unity and intercept differences from zero at the 95% confidence level marked. Three of the EMA's exhibited significantly differing slopes (from unity), with two exhibiting significant intercept differences. Thus, the PM_{2.5} at outdoor and central site

locations were essentially identical for EMA's 3, 5, and 6, while the other EMA regression slopes showed significant biases. The slopes for EMA's 4 and 7 show biases from unity exceeding 15%. The significant intercept differences for EMA's 1 and 5 are thought to be associated with nearby localized strong sources. The subsequent paper analyzing both mass and constituents for PM_{2.5} should provide this confirmation.

Infiltration Modeling - The mean computed factors F_{inf} and F_{pex} for DEARS participant-days by season (for all EMAs) are provided in Table 3-7. As noted previously, the F_{pex} factors are computed using personal data with a wearing compliance level of 0.40 or greater. Additionally, the impact of ETS levels on the computation of F_{pex} considering four levels: <1.5, 1.5 to 3.0, 3.0 to 5.0, and > 5.0 $\mu g/m^3$ mass contributions from ETS to the total mass for both personal and indoor samples. Elemental sulfur data for DEARS Season 6 samples were not available during these data analyses, and no computations were possible for this season.

The values of F_{inf} were nearly identical for the three summer seasons, averaging 0.81. The mean F_{inf} level for the two winter seasons was lower at 0.59. The F_{pex} data for ETS levels <1.5 $\mu g/m^3$ were similarly high during the summer (mean of 0.78), and lower during the winter (0.59). The season mean F_{pex} levels for the 1.5 to 3.0 and 3.0 to 5.0 $\mu g/m^3$ ETS mass contributions were not different from the <1.5 $\mu g/m^3$ level.

However, when the ETS contributions exceeded 5.0 $\mu g/m^3$, the F_{pex} levels increased significantly, with a summer mean of 0.94 and a winter mean of 0.81. These increased F_{pex} levels undoubtedly reflect sulfur contributions from the ETS deposits as suggested by Equation (6).

The various mean exposure component fractions for all participants (and EMA's) by season is provided in Table 3-8. The mean value for E in Table 3-8 is comparable to the Personal column given in Table 3-5, but those in Table 3-8 consider only personal sampler data with wearing compliance levels >0.40. Comparison between tables suggests that stratifying for wearing compliance had no statistically-significant effect on the summer means (p=0.16), but that the winter mean with compliance was significantly lower (p=0.015) by 1.0 $\mu g/m^3$.

As expected, the mean contributions from ETS were highly variable across seasons, ranging from 0.8 $\mu g/m^3$ in Season 5 to 6.9 $\mu g/m^3$ in Season 6. The mean ETS contribution was greater than 5 $\mu g/m^3$ for two of the seasons. The mean ETS contribution for season 4 is larger than the contribution for all other non-outdoor sources.

The contributions from outdoor sources (E_O) was always at least 50% larger than contributions from (ETS-adjusted) non-outdoor sources. During the winter seasons, E_O was consistently a factor of 1.7 to 1.8 larger (than E^*_{NO} (8.7 versus 5.0) but was larger during the summer seasons (13.8 versus 6.4), ranging from 1.8 to 2.6 times larger than the non-outdoor contributions. This is consistent with the fraction of time spent outdoors in Table 3-3, where the summer season fraction was much greater than that for the

winter.

1 2 3

Personal Cloud Modeling - The expected contributions to the personal exposures from being indoors and outdoors for know fractions of time are provided in Table 3-9 for each DEARS seasons (all EMAs). These personal cloud contributions considered only data with wearing compliance >40% of the time. As would be expected from the substantially greater fractions of time spent indoors (Table 3-5), the expected contributions from outdoor particles while outside were dramatically smaller than those from the indoor concentrations. The mean ETS-adjusted indoor contributions (E* $_{\rm in}$) were 62 times greater (12.4 versus 0.2) than those from being outdoors in the winter seasons, but only 13 times greater in the summer seasons.

The computed total summer personal clouds (PC_{tot}) were greater than the ETS-adjusted personal clouds, but were not statistically different, 4.2 vs 1.5 $\mu g/m^3$ (p=0.13), while those for the winter seasons were statistically different (p=0.0.002) from the ETS-adjust values. The total personal clouds were slightly larger in the summer seasons but not statistically so (p=0.21), while the ETS-adjusted personal clouds were significantly smaller in winter compared to summer periods (p=0.005).

4. DISCUSSION

Recruitment/Enrollment - Effective recruitment and retention efforts as reported by Phillips et al. (2008) successfully facilitated exposure data collection for this general population cohort. The relatively heavy personal sampling hardware vest combined with the daily activity logging requirements by the participants were reasonably tolerated. Such burden undoubtedly contributed to an excessive level of protocol wearing compliance violations which were monitored for each exposure period.

Successfully meeting the DEARS recruitment and retention targets was the most important requirement for the PM data to reasonably represent the general population of Wayne County. This also facilitates future DEARS analyses and papers which will address linking the cohort exposures more closely with selected source categories within and upwind of the population.

Fractional Times in Microenvironments - The fractions of time DEARS participants self-reported being in various microenvironments can be compared against prior exposure studies (e.g. Burke et al, 2001; Klepeis et al., 2001; Kruize et al, 2003; Schweizer et al., 2007), with Klepeis et al. (2001) providing a very complete assessment representing a US composite. The comparison of the DEARS times [Klepeis et al. data] include: home: 77.4% [68.7%]; outdoors: 4.3% [7.6%]; in transit: 3.3% [5.5%]; away: 12.6% [20.3%]; other: 3.5% [11%]. The DEARS and Klepeis et al. don't always agree closely - e.g. time spent away, but it was difficult to establish the comparability of the characteristics of the DEARS cohort with other general metro area populations. As noted by Phillips et al. (2008), the probable lower-socioeconomic status of many of the participants undoubtedly is a factor in their activities and exposures. Philips et al. reported that on average, 33% of the cohort worked (away from home) during the

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previous week. But the high unemployment levels in the metro area may have resulted in few commutes and less time spent away from home.

All three winter seasons for the DEARS were typically cold and snowy, providing reasonable explanations for the relatively small fractions of time spent outdoors compared with the summer seasons. No real differences were observed across seasons for time spent in transit with the overall average of 0.033 translating to an average daily commuting time of 47.5 minutes.

PM_{2.5} **Exposure Characteristics by EMA and Season** - The DEARS PM exposures and concentration data by microenvironment in Tables 3-4 and 3-5 - uncorrected for ETS - can be compared against numerous prior studies including Williams et al. (2000); Williams et al. (2003), Weisel et al. (2005), and Johannesson et al. (2007). While the Weisel et al. data are might be the closest in PM source contributions and having a general population cohort to the DEARS study, the personal, indoor, and outdoor level for PM_{2.5} reported are substantially higher than DEARS levels. PM_{2.5} levels in μ g/m³ for DEARS [RIOPA/Houston] include: personal: 20.3 [73], indoor: 18.7 [69], and outdoor: 16.4 [58]. Similar divergent data to the DEARS are reported by Weisel et al. for Elizabeth (NJ) and Los Angeles. Thus, attempts to intercompare absolute PM_{2.5} levels by microenvironment with other literature data are not very meaningful without extensive consideration of many co-factors including meteorology and source contributions. These are well beyond the scope of the current paper.

While the relationships between residential outdoor and indoor PM_{2.5} levels can be inferred from simplistically computing indoor/outdoor ratios, this aspect is better described by the infiltration modeling approach of Wallace and Williams (2005), where sulfur is used as an infiltration marker. This modeling and its relationship to the personal exposure fraction are addressed in a subsequent section. Of importance in Tables 3-4 and 3-5 is the recognition of the impacts on personal and indoor levels of ETS confounding. The observed differences for the DEARS were not large overall, but at times, provided differences that exceeded computed personal cloud computations in Table 3-9.

Johannesson et al (2007) reported PM $_{2.5}$ personal, indoor, and outdoor concentrations for a general population study in Gothenborg, Sweden. They reported mean levels substantially lower than the DEARS data: personal: 11.0. indoor: 9.7; and outdoor: 7.8 μ g/m 3 . They did observe differences caused by passive ETS exposures, but defined the presence/absence of ETS by self-reported questionnaire. This resulted in somewhat inconsistent data with some ETS-present categories lower than those without ETS.

The DEARS distributional personal exposure data shown in Figures 3-2 through 3-5 illustrate the influences that violations to the study protocol requirements for wearing compliance and maintaining a non-smoking household. These data are important since they represent the first data of this type reported for a general population metro area cohort. Some biases were still observed in median and 95th percentile levels, even with

very aggressive efforts to eliminate the confounding effects of ETS on the study mass concentration data. Study designs without such aggressive protocol compliance coaching and quantification for the ETS contributions would undoubtedly result in far more serious - and unknown - bias levels. Wearing compliance monitoring should be considered mandatory for all personal exposure studies. Without including this simple aspect in the study design, the most exposed portions of the distributions will be significantly biased low. For example, while the median differences between ETS-corrected and uncorrected personal exposure levels were essentially identical at 1.6 and 1.9 $\mu g/m^3$ for 0% and 60% compliance, respectively, the differences at the 98th percentile were 10.9 compared with 22.8 $\mu g/m^3$ for these same compliance levels. Clearly, not wearing the personal monitors faithfully can result in extremely biased data for the most-exposed. These exposure biases could be expected to seriously weaken associations with observed adverse health outcomes, given the impact of extremes in the distributions on the statistical correlations.

Providing a thorough understanding of co-factors influencing these differences is beyond the scope of the current analyses, but as noted earlier, will be part of a much more in depth paper applying mixed-models to the potential influencing variables identified here. The substantial amount of data available by season, approach n=500 in most cases, should provide a robust data set for this modeling.

Spatial Uniformity for PM_{2.5} Mass - The PM_{2.5} concentrations at outdoor EMA locations was observed to be statistically different from the central site data for three of the metro areas - EMA's 1, 4, and 7, but the same for EMA's 3, 5, and 6. The biases for EMA's 4 and 7 exceeded 15%. The biases for EMA were attributed to nearby PM_{2.5} sources, while the bias for EMA 7 (the far upwind site) was attributed to the distance between sites (>30 miles). Similar PM_{2.5} mass (and species) gradients in metro areas have been reported previously (e.g. Williams et al., 2003 for RTP, NC; Krudysz et al., 2008 for Long Beach, CA). Further analyses will be conducted in a subsequent paper that considers both mass and species gradients.

A modeling approach suggested by Strand et al. (2006) to calibrate personal and indoor exposures against central site data is potentially relevant for subsequent analyses, if modified to include ETS adjustments as suggested here. This model presumes that central site monitoring is indeed representative of the outdoor across the region of interest. Including regression fits such as those described here would sharpen the accuracy of the approach.

Infiltration and Exposure Factor Modeling - The computation of infiltration and personal exposure factors and described by Wallace and Williams (2005) to understand PM exposure relationships for the RTP Panel Study, significantly advanced the science. The addition of new technologies applied during the DEARS to identify both ETS confounding and personal sampler wearing compliance provides another step forward in minimizing personal exposure biases. The procedures suggested here should be considered for future studies that examine both PM mass relationships and those for the constituent fractions. The technologies required to accomplish these additional

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measures are relatively low burden and inexpensive, transparent to the participants, and are non-destructive for the collected filter samples.

The apparent contribution of sulfur compounds from ETS to the collected indoor and personal exposure samples appears to definitely confound the computations of both F_{inf} and F_{pex} . Literature on ETS chemistry does not clearly identify the total sulfur contributions from ETS by PM size fraction, suggesting that focused data analyses such as these will help identify these contributions. A simplistic estimate of the sulfur contributed to the personal exposures by elevated ETS levels can be made by multiplying the F_{pex} increases for summer and winter seasons (0.16 and 0.23, respectively) times their mean personal exposure sulfur levels (1.44 $\mu g/m^3$ and 0.65 $\mu g/m^3$), respectively. These computations suggest that significant ETS smoking contributes between 0.15 and 0.24 $\mu g/m^3$ (summer and winter, respectively) or more of sulfur to the personal exposure filters. Since ETS contributions less than 5 $\mu g/m^3$ did not show significant changes in F_{pex} , perhaps these small increments are apparently easily masked at lower levels. This sulfur level suggests that the mass fraction of sulfur in (passive) tobacco smoke is roughly 4% (0.2 $\mu g/m^3$ ETS/5 $\mu g/m^3$ mass).

Further analysis of the acceptable threshold level applying the ETS method is warranted, since allowing mass concentration additions as large as $5 \,\mu g/m^3$ to the collected personal and indoor samplers obviously provides excessive mass and other ETS component (aldehydes, VOC's, carcinogens, etc.) biases. The methodology applied to compute F_{pex} appears to become confounded when the sulfur content added by the ETS aerosol becomes a significant part of the total sulfur. This aspect will be examined in more detail in the subsequent paper that includes the PM exposure constituent chemistry along with the mass analyses.

Even though the DEARS ETS confounding was considered quite small, given the non-smoking protocol compliance requirement, the percentage of personal PM exposure data affected was substantial. Table 4-1 provides the percentage of personal samples with ETS levels exceeding 1.5, 3.0 and 5.0 $\mu g/m^3$ of ETS mass for each DEARS season. If the 5 $\mu g/m^3$ ETS is the threshold for significant biases to F_{pex} estimates, this would suggest that 3.4 to 18.1% of the DEARS personal data would have been impacted.

The stratification of exposure fraction computations in Table 3-8 and personal clouds in Table 3-9 at a wearing compliance level of 40% necessarily reduces the amount of data available for these computations. Table 4-2 indicates the percentage of personal data by season that remain after applying compliance levels of 20, 40, and 60%. While a higher level of 60% is shown in Figures 3-5 to more clearly identify the most exposed, a lower compliance level of 40% retains far more of the data in the core of the exposure distribution. This provides greater confidence in the mean and median data due to the nearly doubling of the data available with a 20% lower required compliance limit with virtually no impact on the observed means.

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The distributions of PM_{2.5} exposure shown in Figures 3-2 and 3-3 highlight features and findings for the DEARS, program and suggest how prior similar exposure data are likely to compare. For example, the median (and mean) levels were statistically identical no matter which wearing compliance level was utilized. This suggests that the median levels reported for prior residential PM personal exposure studies would likely have been the same whether or not compliance was monitored. Only in personal exposure scenarios with very strong localized PM_{2.5} sources would median differences be detected due to poor wearing compliance. The data 0% compliance level of Figure 3-2 would be the most comparable to prior personal exposure studies not incorporating actual wearing compliance or adjustment for ETS confounding. Ott et al. (2000) reported very similar exposure distribution shapes for the data from three different studies (although for PM₁₀), and highlighted significant departures from log-normality for the Toronto data taken from the personal PM_{2.5} exposure study reported by Pellizzari et al. (1999). The most exposed in the latter cohort often deviated from the median levels by factors of five or more. Closer scrutiny showed that even after stratifying for ETS confounding, proximity to strong combustion source during commuting and hobby events (e.g. riding the subway, auto repair, welding) could result in dramatically higher exposures. These atypical levels can be extremely difficult to predict and as noted by Burke et al. (2001) induce significantly higher uncertainties into models such as the EPA SHEDS-PM at levels above the 90th percentile. Similarly, Hanninen et al. (2005) observed that models consistently underestimated the exposures of those above the 95th percentile for the EXPOLIS cohort. Edward and Juntenen (2009) expanded upon the latter observation, concluding that exposure models based on ambient concentrations do not adequately account for the sources of PM influencing the "high end" of personal $PM_{2.5}$ exposure distributions.

Importantly, the exposures comprising the >90th percentile levels define the range of the data for associative statistics in epidemiologic studies. Errors in either measuring or predicting these exposures could be expected to substantially weaken epidemiologic relationships. Providing the most accurate and representative exposure data for the elevated levels is then critical to establishing the most robust associations. Actual, rather than modeled, personal PM exposure monitoring provides this level of robustness, but only if wearing compliance levels are verified and adjustments made for ETS confounding.

The personal clouds, unadjusted and ETS-adjusted in Table 3-9 for the DEARS seasons, are similar to data reported previously, but importantly the data reported here reflect a minimum 40% wearing compliance and could be expected to reflect less exposure misclassification bias. The mean PM_{2.5} personal clouds for three different compromised cohorts (mostly elderly)was reported to be 3.0 μ g/m³ by Rodes et al. (2001). While ETS characterizations were not made for these studies, careful review suggested minimal smoking was present in or around these elderly participants. The DEARS personal PM_{2.5} cloud levels of 3.8 μ g/m³ in the summer and 1.5 μ g/m³ (ETS adjusted) for the winter are reasonably consistent with these prior data, but some level of uncharacterized passive ETS exposures may indeed be reflected in the prior studies.

5. CONCLUSIONS AND IMPLICATIONS

contributions for EXPOLIS study.

These analyses provide the initial summary of PM_{2.5} mass concentrations relationships for all seasons, participants, and EMA's for the DEARS. They are intended to highlight the utility of the new methodologies applied in addition to summarizing the data. The general population nature of the DEARS cohort as described by Phillips et al. (2008) required more aggressive recruiting and retention efforts to successfully select the target numbers of participants during each season and from each EMA. The fractions of time spent in various microenvironments for the DEARS cohort was not substantially different from those reported in other studies and survey. Local factors such as the high unemployment rate in the metro area, especially for lower socioeconomic level participants, undoubtedly played some role in factors such as the fractions of time spent away from home and commuting.

The personal cloud level for the most exposed DEARS participants (90th

and sources may be disproportionately contributing to the upper portion of the exposure

Juntenen (2009) who studied the distributions of elemental analyses and reported that

the most and least exposed individuals reflect substantially differing source category

percentile) for all participant days was 10.7 μg/m³, highlighting that differing activities

distributions. This point was reinforced for PM2.5 exposure data by Edwards and

Unlike studies of compromised individuals, the Detroit metro area general population cohort had poorer study protocol compliance in two key areas - personal sampler wearing compliance, and maintaining a non-smoking household. The application of real-time wearing compliance monitoring and the ability to non-destructively characterize collected exposure samples for ETS equivalent mass contributions allowed these violations to be addressed in a quantitative manner. A 40% wearing compliance acceptance level was determined to be necessary to provide a balance between minimizing exposure misclassification (from unworn monitoring) and having sufficient data to conduct robust statistical analyses. An ETS threshold level equivalent to adding more than 5 $\mu g/m^3$ to the collected sample was found to be necessary to detect changes in the personal exposure factor (Fpex).

Statistically-significant spatial PM_{2.5} gradients were identified in 3 of the 6 EMA's in Wayne County. These were expected, given the number of strong, localized PM sources in the Detroit metro area. The EMA 4 residential outdoor bias levels compared with the central site at Allen Park exceeded 15%. More extensive analyses in a followup paper will also examine the gradients for PM constituents.

The overall personal exposure levels unadjusted for ETS compared with indoor and outdoor levels were similar to prior PM personal exposure studies where the personal levels were significantly larger than either the indoor or outdoor concentrations. Breaking this total personal exposure into components suggested that after isolating the ETS component, the relative contributions from outdoor sources was very similar to other metro area studies. After adjusting for ETS biases, the outdoor contributions to the personal exposure were typically larger by factors ranging from 1.75 to 2.2

compared with those of the non-outdoor sources. The outdoor contribution was larger in the summer than the winter, which is consistent with the fractions of time spent outdoors summer versus winter (6.7% versus 1.1% of the time).

It is critically-important to recognize that the impact of acceptable personal PM monitor wearing compliance was determined from a contaminant category ($PM_{2.5}$) previously identified as reasonably uniformly spatially-distributed. Some constituents of $PM_{2.5}$ and the mass and constituents in larger (PM_{coarse}) and smaller ($PM_{ultrafine}$) size fractions with stronger spatial gradients are expected to show substantially-biased distributions for the most-exposed. This contention also extends to personal exposures for gas-phase contaminants.

The importance of characterizing personal wearing compliance is demonstrated, given that various threshold levels can then be applied, depending on the requirements of the data analyses and the expected sensitivity of the analyses to compliance. While a 60% threshold should provide more representative personal exposures than 40% or below, compromises are required when the burden of the sampling systems results in significant levels of protocol violation, even with very aggressive technician participant coaching. Smaller, lighter, less-obtrusive exposure systems should a critical key goal of future sampling and sensor developmental efforts, with the ability to quantify the levels of compliance no matter what the burden level. Participants can also simply forget to wear even the smallest, lightest monitor, providing exposure misclassification error in the best designs.

Personal PM_{2.5} cloud levels for the general population DEARS cohort were surprisingly comparable to those reported previously for compromised cohorts. While this will be examined more fully in a subsequent paper, it the contributions from typical activities such as cooking are fairly universal in the levels added due to source proximity and unmonitored microenvironments. The mass contributions to the personal exposures from outdoor air while the participants were actually outside for all DEARS seasons was quite small compared to the contributions from all sources while indoors. The personal exposure collections indoors were typically at least a factor of 13 times greater than those contributed outdoors.

Overall, the DEARS PM personal, indoor, and outdoor exposure methodologies provided new insights and new methodologies for assessing ETS and wearing compliance that should seriously be considered for all future aerosol exposure studies. These methods provided high quality data with high data capture rates. The primary drawback to the PM methodologies was the high level of participant burden due to the weight and bulkiness of the sampling systems. Reducing the burden of these technologies while retaining the current features would greatly advance exposure sciences, while facilitating their application for an even broader range of cohorts - general population adults and children (Weis et al., 2005; Nieuwenhuijsen et al., 2006).

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Table 3-1. DEARS participants recruited by season and across EMA's. Since participants recruited in seasons 1, 3, and 5 were expected to participate for two successive seasons, those recruited in seasons 2, 4, and 6 were replacements for dropouts

Season	EMA 1	EMA 3	EMA 4	EMA 5	EMA 6	EMA 7	totals
1	8	8	7	none	9	6	38
2	6	11	5	none	7	7	36
3	9	5	8	8	10	none	40
4	9	5	8	8	10	none	40
5	9	none	11	9	11	none	40
6	8	none	8	5	7	none	28
totals	49	29	47	30	54	13	20

Table Note: 1) Those recruited for Seasons 1, 3, and 5 were expected to participate for two consecutive seasons; see Williams et al., 2008

DEARS Season identification: 1 - Summer, 2004; 2 - Winter, 2004/2005; 3 - Summer, 2005; 4 - Winter, 2005/2006; 5 - Summer, 2006; 6 - Winter, 2006/2007.

Table 3-2. DEARS mean fractional times (x) spent each day in microenvironments by EMA across all seasons

DEARS EMA				1	
	Home	Outdoors	Transit	Away	Other
	x = 0.825	x = 0.055	x = 0.024	x = 0.058	x = 0.033
1	s = 0.135	s = 0.073	s = 0.045	s = 0.087	s = 0.041
	n = 150				
	x = 0.754	x = 0.036	x = 0.043	x = 0.122	x = 0.04
3	s = 0.180	s = 0.056	s = 0.055	s = 0.133	s = 0.041
	n = 112				
	x = 0.764	x = 0.041	x = 0.041	x = 0.103	x = 0.04
4	s = 0.190	s = 0.062	s = 0.053	s = 0.134	s = 0.046
	n = 142				
	x = 0.835	x = 0.055	x = 0.016	x = 0.062	x = 0.02
5	s = 0.148	s = 0.073	s = 0.032	s = 0.106	s = 0.036
	n = 98				
	x = 0.751	x = 0.034	x = 0.037	x = 0.131	x = 0.04
6	s = 0.201	s = 0.060	s = 0.047	s = 0.167	s = 0.04
	n = 172				
	x = 0.664	x = 0.042	x = 0.035	x = 0.208	x = 0.05
7 ¹	s = 0.285	s = 0.087	s = 0.052	s = 0.254	s = 0.05
	n = 54				
A.,	x = 0.774	x = 0.043	x = 0.033	x = 0.106	x = 0.03
ALL	s = 0.190	s = 0.067	s = 0.049	s = 0.149	s = 0.04
EMAs	n = 728				

Table Notes: 1) EMA 7 site operated for Seasons 1 & 2 only 2) x=mean; s=arith std dev.; n=#

Table 3-3. DEARS mean fractional times (x) spent each day in microenvironments by Season across all EMAs

DEARS Season				T	
	Home	Outdoors	Transit	Away	Other
1 - sum	x = 0.714	x = 0.061	x = 0.042	x = 0.135	x = 0.048
	s = 0.191	s = 0.086	s = 0.052	s = 0.151	s = 0.046
	n = 144				
2 - win	x = 0.741	x = 0.011	x = 0.041	x = 0.170	x = 0.037
	s = 0.239	s = 0.017	s = 0.054	s = 0.200	s = 0.041
	n = 138				
3 - sum	x = 0.779	x = 0.078	x = 0.035	x = 0.068	x = 0.026
	s = 0.151	s = 0.076	s = 0.047	s = 0.105	s = 0.038
	n = 149				
4 - win	x = 0.830	x = 0.014	x = 0.026	x = 0.096	x = 0.029
	s = 0.167	s = 0.032	s = 0.047	s = 0.1364	s = 0.034
	n = 120				
5 - sum	x = 0.793	x = 0.060	x = 0.020	x = 0.068	x = 0.053
	s = 0.169	s = 0.069	s = 0.035	s = 0.120	s = 0.052
	n = 127				
6 - win	x = 0.844	x = 0.003	x = 0.028	x = 0.076	x = 0.045
	s = 0.179	s = 0.011	s = 0.54	s = 0.110	s = 0.049
	n = 50				
Summer Seasons	x = 0.761 s = 0.174 n = 420	x = 0.067 s = 0.078 n = 420	x = 0.033 s = 0.046 n = 420	x = 0.091 s = 0.130 n = 420	x = 0.042 s = 0.046 n = 420
Winter Seasons	x = 0.792 s = 0.209 n = 308	x = 0.011 s = 0.024 n = 308	x = 0.033 s = 0.052 n = 308	x = 0.126 s = 0.169 n = 308	x = 0.035 s = 0.040 n = 308

Table Notes: 1) EMA 7 site operated for Seasons 1 & 2 only 2) sum = summer; win = winter

³⁾ x=mean; s=arith std dev.; n=#

Table 3-4. DEARS PM overall arithmetic mean concentrations by size fraction for each EMA, averaged across all participants and seasons

DEARS						
EMA	Personal ¹	Personal ²	Indoor ¹	Indoor ²	Outdoor	Central ³
-	18.2 (10.8)	16.6 (10.4	17.6 (11.8)	16.0 (10.5)	16.8 (9.1)	15.8 (9.1)
က	15.1 (10.0)	14.3 (9.7)	12.6 (8.3)	12.2 (8.4)	17.3 (12.7)	17.5 (13.0)
4	29.0 (32.6)	22.4 (24.2)	26.8 (29.5)	23.0 (23.9)	15.3 (8.9)	16.9 (9.4)
2	23.7 (18.5)	21.4 (18.3)	20.5 (13.2)	18.7 (13.4)	18.4 (8.3)	15.3 (7.4)
9	18.4 (20.9)	16.2 (18.0)	17.6 (23.2)	15.8 (20.5)	15.9 (10.2)	16.7 (9.7)
73	12.1 (8.6)	11.3 (8.1)	10.5 (9.2)	9.2 (8.0)	14.5 (11.5)	18.4 (14.3)
all EMA's mean [median]	20.3 [15.5] (n=874)	17.6 [13.7] (n=874)	18.7 [14.3] (n=973)	16.8 [13.2] (n=973)	16.4 [14.5] (n=1,049)	16.6 [13.9] (n=1,044)

Table Notes: 1) <u>Not</u> adjusted for ETS mass confounding
2) Adjusted for ETS mass
3) Central and Dichot samplers at Allen Park
4) EMA 7 sampling only conducted during Seasons 1 & 2

Table 3-5. DEARS PM arithmetic mean levels for each season, averaged across all participants and EMA's, and including all levels of wearing compliance

DEARS							
Season #	Season	Personal	Personal ²	Indoor	Indoor ²	Outdoor	Central ³
1	summer	18.7 (15.5)	17.1 (15.2)	17.6 (15.9)	17.5 (16.2)	16.0 (10.3)	17.6 (11.8)
2	winter	12.6 (12.3)	11.7 (11.1)	10.7 (9.5)	10.1 (8.7)	17.1 (14.7)	17.8 (14.5)
က	summer	22.5 (14.4)	21.0 (14.0)	20.6 (12.0)	18.8 (11.6)	19.1 (10.0)	18.6 (8.9)
4	winter	23.9 (32.5)	18.8 (23.6)	23.1 (32.9)	18.4 (18.0)	14.2 (6.7)	13.1 (6.1)
2	summer	21.4 (16.9)	20.7 (15.8)	21.5 (20.8)	21.0 (25.7)	17.0 (8.2)	16.98 (8.0)
9	winter	25.8 (30.8)	17.4 (22.2)	17.9 (16.7)	16.7 (19.3)	14.8 (8.0)	15.6 (7.9)
1, 3, 5	summer mean [median]	20.9 [17.5] (n=502)	19.6 [16.1] (n=502)	20.0 [16.2] (n=539)	19.2 [14.9] (n=539)	17.4 [15.1] (n=498)	17.7 [15.2] (n=546)
2, 4, 6	winter mean [median]	19.6 [11.7] (n=372)	15.1 [10.0] (n=372)	17.2 [10.3] (n=434)	14.4 [9.9] (n=434)	15.3 [13.8] (n=489)	15.4 [12.7] (n=498)

Table Notes: 1) <u>Not</u> adjusted for ETS mass confounding
2) Adjusted for ETS mass
3) Central and Dichot samplers at Allen Park
4) DEARS Season identification: 1 - Summer, 2004; 2 - Winter, 2004/2005; 3 - Summer, 2005; 4 - Winter, 2006/2007.

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Table 3-6. Linear regression statistics for each DEARS EMA personal exposure PM_{2.5} mass on the fixed-location central site (Allen Park) data

EMA	Slope	Intercept	R²	n
1	0.9279*	2.0251*	0.8818	214
3	1.0396	0.0064	0.9055	132
4	0.8259*	1.0380	0.7976	209
5	0.9398	3.4925*	0.8019	136
6	1.0076	-0.7794	0.8849	242
7	0.8302*	0.0131	0.9385	60

Table Notes: 1) EMA 7 site operated for Seasons 1 & 2 only
2) * slope statistically differs from unity or intercept statistically differs from zero at CI=95%

Table 3-7. Mean (x) personal exposure (F_{pex}) and infiltration (F_{inf}) factors for PM_{2.5} across all participants and seasons, for wearing compliance levels >0.40. Personal exposure factors stratified at four levels of ETS contributions (in $\mu g/m^3$) to show ETS impacts on F_{pex} .

DEARS Season #	Season	F _{inf}	F _{pex} (<1.5)	F _{pex} (1.5 - 3.0)	F _{pex} (3.0 - 5.0)	F _{pex} (> 5.0)
1	summer	x = 0.81 s = 0.23 n = 66	x = 0.74 s = 0.23 n = 77	x = 0.76 s = 0.18 n = 42	x = 0.79 s = 0.16 n = 6	x = 0.98 s = 0.38 n = 4
2	winter	x = 0.58 s = 0.14 n = 93	x = 0.56 s = 0.15 n = 95	x = 0.55 s = 0.13 n = 7	x = 0.71 s = n = 1	x = 0.78 s = 0.17 n = 6
3	summer	x = 0.82 s = 0.17 n = 70	x = 0.78 s = 0.21 n = 72	x = 0.81 s = 0.15 n = 28	x = 0.79 s = 0.12 n = 3	x = 0.79 s = 0.41 n = 3
4	winter	x = 0.64 s = 0.17 n = 36	x = 0.63 s = 0.22 n = 38	x = 0.62 s = 0.15 n = 25	x = 0.64 s = 0.11 n = 12	x = 0.82 s = 0.34 n = 12
5	summer	x = 0.81 s = 0.22 n = 91	x = 0.80 s = 0.20 n = 94	x = 0.80 s = 0.10 n = 9	x = 0.79 s = 0.17 n = 5	x = 1.08 s = 0.34 n = 2
6	winter	x = na s = na n = na	x = na s = na n = na	x = na s = na n = na	x = na s = na n = na	x = na s = na n = na
1, 3, 5	summer mean	x = 0.81 s = 0.21 n = 227	x = 0.78 s = 0.21 n = 243	x = 0.78 s = 0.16 n = 79	x = 0.79 s = 0.14 n = 14	x = 0.94 s = 0.35 n = 9
2, 4	winter mean	x = 0.59 s = 0.15 n = 129	x = 0.59 s = 0.17 n = 133	x = 0.61 s = 0.15 n = 32	x = 0.64 s = 0.11 n = 13	x = 0.81 s = 0.29 n = 18

Table Notes: 1) Not adjusted for ETS confounding

DEARS Season identification: 1 - Summer, 2004; 2 - Winter, 2004/2005; 3 - Summer, 2005; 4 - Winter, 2005/2006; 5 - Summer, 2006; 6 - Winter, 2006/2007.

³⁾ x=mean; s=arith std dev.; n=#

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Table 3-8. Mean (x) total personal mass exposure (E) and component exposure fractions across all participants and seasons for $PM_{2.5}$, adjusted for wearing compliance levels >0.40. Personal exposure factors stratified at four levels of ETS contributions (in $\mu g/m^3$) to show ETS impacts on F_{pex} .

DEARS Season #	Season	E	E _O	E* _{NO}	E _{pers}	E _{ETS}
1	summer	x = 19.3 s = 13.3 n = 108	x = 12.2 s = 9.5 n = 70	x =5.2 s = 7.9 n = 53	x = 17.7 s = 16.4 n = 6	x = 1.7 s = 1.5 n = 70
2	winter	x = 12.31 s = 13.3 n = 108	x = 9.0 s = 7.8 n = 89	x = 5.2 s = 8.7 n = 89	x = 11.5 s = -12.0 n = 108	x = 0.9 s = 2.9 n = 152
3	summer	x = 24.0 s = 16.3 n = 112	x = 16.4 s = 10.0 n = 64	x = 6.4 s = 7.1 n = 55	x = 22.6 s = 15.6 n = 64	x = 1.5 s = 1.7 n =175
4	winter	x = 22.5 s = 25.9 n = 93	x = 8.1 s = 4.4 n = 34	x = 4.6 s = 6.5 n = 24	x = 18.2 s = 16.1 n = 92	x = 5.0 s = 13.8 n = 155
5	summer	x = 20.9 s = 17.5 n = 112	x = 13.1 s = 6.2 n = 80	x = 7.2 s = 7.0 n = 70	x = 20.3 s = 16.3 n = 112	x = 0.8 s = 1.9 n = 162
6	winter	x = 23.3 s = 27.3 n = 84	x = 15.4 s = 15.1 n = 25	x = 9.3 s = 14.0 n = 24	x = 17.2 s = 20.1 n = 85	x = 6.9 s = 19.4 n = 114
1, 3, 5	summer mean	x = 21.3 s = 16.7 n = 358	x = 13.8 s = 8.7 n = 214	x = 6.4 s = 7.3 n = 178	x = 20.1 s = 16.2 n = 358	x = 1.3 s = 1.8 n = 504
2, 4	winter mean	x = 18.6 s = 23.6 n = 249	x = 8.7 s = 7.1 n = 123	x = 5.0 s = 7.9 n = 65	x = 14.6 s = 16.0 n = 249	x = 4.1 s = 11.6 n = 370

Table Notes: 1) Not adjusted for ETS confounding

DEARS Season identification: 1 - Summer, 2004; 2 - Winter, 2004/2005; 3 - Summer, 2005; 4 - Winter, 2005/2006; 5 - Summer, 2006; 6 - Winter, 2006/2007.

³⁾ x=mean; s=arith std dev.; n=#

Table 3-9. Mean (x) total and ETS-adjusted personal exposure cloud computations (PCtot, PC*tot, respectively) across participants and seasons, adjusted for wearing compliance levels >0.40.

	T	7			
DEARS Season #	Season	E _{out}	E*in	PC _{tot}	PC*tot
1	summer	x = 1.0 s = 1.8 n = 138	x =13.5 s = 14.5 n = 126	x = 5.7 s = 16.4 n = 90	x = 4.2 s = 16.1 n = 90
2	winter	x = 0.2 s = 0.33 n = 126	x = 7.9 s = 8.8 n = 133	x = 4.1 s = -5.5 n = 79	x = 3.5 s = 4.8 n = 79
3	summer	x = 1.5 s = 1.6 n = 139	x = 17.1 s = 10.5 n = 148	x = 6.0 s = 10.3 n = 83	x = 4.6 s = 10.3 n =83
4	winter	x = 0.2 s = 0.5 n = 116	x = 15.8 s = 14.4 n = 116	x = 2.7 s = 5.1 n = 68	x = 0.5 s = 5.4 n = 68
5	summer	x = 1.1 s = 1.7 n = 124	x = 16.6 s = 13.0 n = 122	x = 3.1 s = 5.9 n = 74	x = 2.6 s = 5.9 n = 74
6	winter	x = 2.0 s = 14.1 n = 78	x = 18.0 s = 17.7 n = 83	x = 6.8 s = 17.4 n = 55	x = 1.3 s = 14.0 n = 55
1, 3, 5	summer mean	x = 1.2 s = 1.7 n = 401	x = 15.8 s = 12.7 n = 396	x = 5.0 s = 12.0 n = 247	x = 3.8 s = 11.9 n = 247
2, 4	winter mean	x = 0.2 s = 0.4 n = 288	x = 12.4 s = 13.0 n = 299	x = 4.2 s = 9.3 n = 182	x = 1.5 s = 7.2 n = 182

Table Notes: 1) Not adjusted for ETS confounding

DEARS Season identification: 1 - Summer, 2004; 2 - Winter, 2004/2005; 3 - Summer, 2005; 4 - Winter, 2005/2006; 5 - Summer, 2006; 6 - Winter, 2006/2007.

³⁾ x=mean; s=arith std dev.; n=#

Table 4-1. Percentage of DEARS personal exposure data with ETS mass contributions that exceeded selected ETS threshold percentage levels

	ETS person	nal mass thres	shold, μg/m
Season	1.5	3.0	5.0
1	42.5	10.2	3.6
2	13.2	7.9	6.6
3	33.1	8.0	3.4
4	55.5	30.3	18.1
5	17.5	8.8	3.8
6	na	na	na

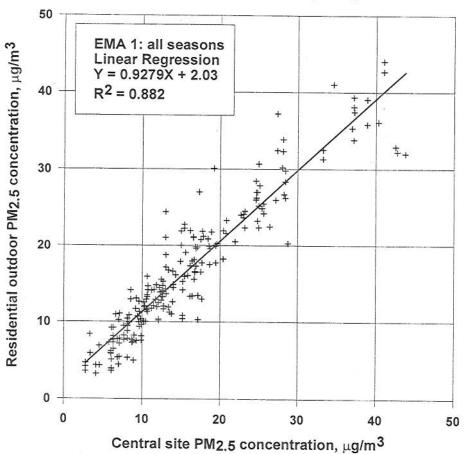
Table 4-2. Percentage of DEARS personal exposure data with wearing compliance levels less than 20, 40, and 60%

	Wearin	g compliance	level, %
Season	20%	40%	60%
1	88.2	79.7	38.0
2	82.1	70.9	29.6
3	80.2	61.4	18.6
4	64.8	51.3	12.1
5	80.5	65.0	17.5
6	na	na	na



Figure 2-1. DEARS personal sampling vest showing size-specific sampling inlets near the breathing zone and pumps located in pockets near the waist.

Figure 3-1. Example regression of DEARS Central site versus residential outdoor $PM_{2.5}$ for EMA 1 for all seasons and participants



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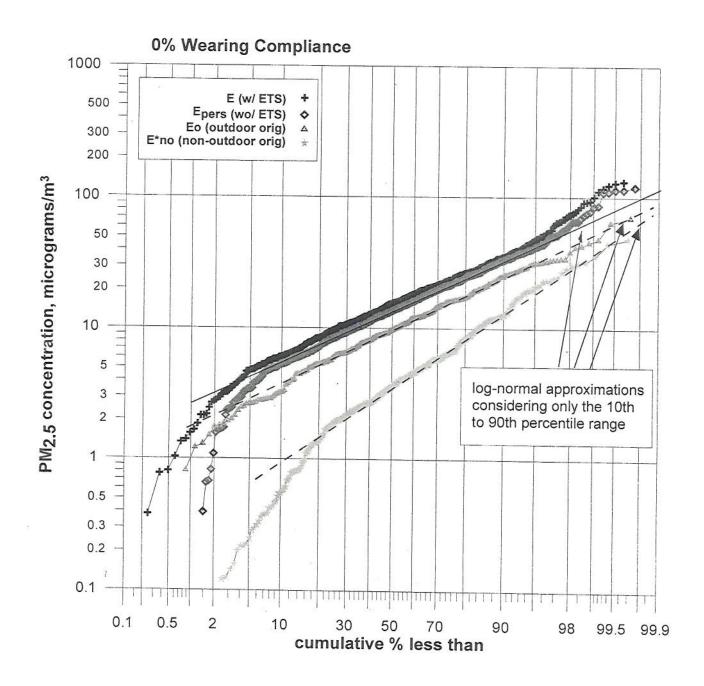
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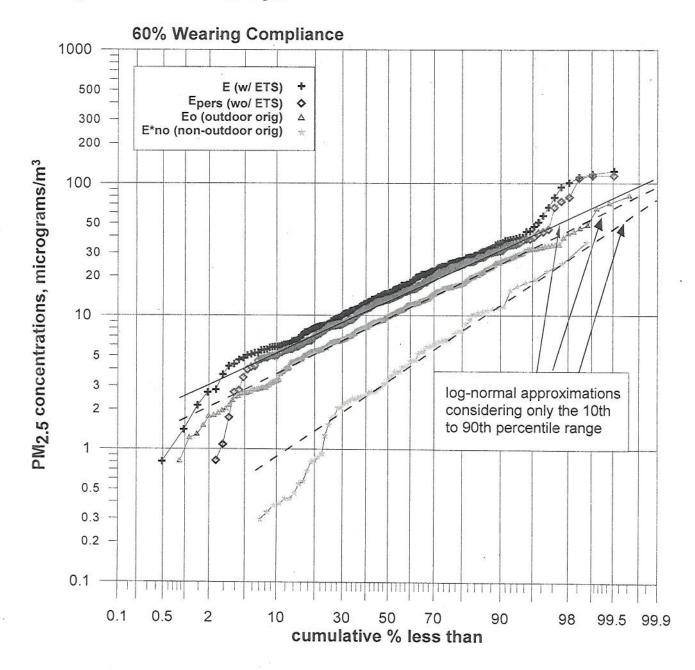
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Figure 3-2. ETS-corrected and uncorrected personal PM_{2.5} exposure distributions for all DEARS seasons and participants, applying no (0.0%) wearing compliance threshold requirement, including a comparison of the distributions of personal PM_{2.5} of outdoor and indoor origin.



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Figure 3-3. ETS-corrected and uncorrected personal $PM_{2.5}$ exposure distributions for all DEARS seasons and participants, applying a 60.0% wearing compliance threshold requirement, including a comparison of the distributions of personal $PM_{2.5}$ of outdoor and indoor origin.



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