Development and evaluation of alternative approaches for exposure assessment of multiple air pollutants in Atlanta, Georgia

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Abstract

Measurements from central site (CS) monitors are often used as estimates of exposure in air pollution epidemiological studies. Since these measurements are typically limited in their spatiotemporal resolution, true exposure variability within a population is often obscured, leading to potential measurement errors. To fully examine this limitation, we developed a set of alternative daily exposure metrics for each of the 169 ZIP codes in the Atlanta, GA metropolitan area, from 1999-2002, for $PM_{2.5}$ and its components (EC, SO_4), O_3, CO, and NO_x. Metrics were applied in a study investigating the respiratory health effects of these pollutants. The metrics included: i) CS measurements (one CS per pollutant); ii) air quality model results for regional background pollution; iii) local-scale AERMOD air quality model results; iv) hybrid air quality model estimates (a combination of ii and iii); and iv) population exposure model predictions (SHEDS and APEX). Differences in estimated spatial and temporal variability were compared by exposure metric and pollutant. Comparisons showed that: 1) both hybrid and exposure model estimates exhibited high spatial variability for traffic-related pollutants (CO, NO_x, and EC), but little spatial variability among ZIP code centroids for regional pollutants ($PM_{2.5}$, SO_4, and O_3); 2) for all pollutants except NO_x, temporal variability was consistent across metrics; 3) daily hybrid-to-exposure model correlations were strong (r >0.82) for all pollutants, suggesting that when temporal variability of pollutant concentrations is of main interest in an epidemiological application, the use of estimates from either model may yield similar results; 4) exposure models incorporating infiltration parameters, time-location-activity budgets, and other exposure factors affect the magnitude and spatiotemporal distribution of exposure, especially for local pollutants. The results of this analysis can inform the development of more appropriate exposure metrics for future epidemiologic studies of the short-term effects of particulate and gaseous ambient pollutant exposure in a community.

Introduction

Measurements from central site (CS) monitors are often used as estimates of exposure in epidemiologic studies investigating the short-term health effects of air pollution (1-5). Fixed-site monitors may be sufficient for representing ambient concentrations for pollutants with limited spatial and temporal heterogeneity. For pollutants with local source impacts, the concentrations measured at CS monitors may not represent intra-urban variation in air pollution levels (6-8). This may lead to exposure misclassification in an epidemiology study, which can introduce statistical error that affects the strength and significance of estimated health effect associations (9).

Alternatives to exclusive reliance on ambient concentration data from central monitoring sites include various approaches, such as spatially dense sampling campaigns or modeling (e.g., air quality dispersion models, land use regression models) of pollutant concentrations, which may increase the spatial resolution of ambient pollutant concentrations (7, 10-17). Human exposure models (such as SHEDS and APEX) can provide spatiotemporally-refined ambient exposure estimates by incorporating factors such as human activity and behaviors of individuals as they move through space and time, in addition to relevant demographic and home environment characteristics (e.g., air exchange rate) that impact outdoor to indoor air pollutant infiltration (17-19). Where appropriate, the models could also be used to characterize the contribution of indoor sources of air pollution to total exposures.

While epidemiologic studies of the adverse health effects of exposure to ambient pollution have been conducted using modeled mid- to long-term exposure estimates (20-24), few studies of acute morbidity have used modeled daily, spatially-refined, estimates of ambient concentrations (25, 26). To our knowledge, no population-based studies of air pollution and acute morbidity are available where spatially-refined estimates of ambient population exposure have been applied, beyond a few feasibility studies (27, 28). Development and evaluation of alternative exposure assignment approaches, which provide information on spatiotemporally refined ambient concentrations and ambient population exposures, is needed for use in improved population-based acute health effects studies. The research presented here provides a unique comparison of alternative exposure estimates obtained from measurements, modeling of ambient pollution levels, and human exposure models in a single study.

Presented here is the development of a suite of alternative exposure metrics developed by the U.S. Environmental Protection Agency (EPA) in collaboration with Emory University and the Georgia Institute of Technology for use in a time-series study examining the relationships between ambient air pollution and acute morbidity outcomes (based on daily emergency department visits by ZIP code) in Atlanta, GA. The Atlanta study domain includes the city's downtown as well as surrounding suburban and rural areas, and has a wide range of air pollution emissions from a variety of point and mobile sources. The study examines a variety of pollutants with a range of spatial and temporal variability, including several that are highly influenced by local traffic (elemental carbon (EC), carbon monoxide (CO), and nitrogen oxides (NO_x))(8, 29-32) as well as pollutants more dominated by regional contributions (particulate matter with aerodynamic diameter less than 2.5 µm ($PM_{2.5}$), sulfate (SO_4), and secondary or regional ozone (O_3)) (31, 33-36). To provide spatially-refined ambient concentrations and exposures for this study, we applied a number of statistical, mechanistic and behavioral models (e.g., AERMOD, SHEDS and APEX) to develop five alternative exposure metrics for each of the six pollutants.

In this paper, we outline the development of each exposure assignment approach and conduct a detailed characterization of how each alternative metric compares to CS monitor measurements. We also discuss implications for use of these alternative metrics in place of CS measurements in the Atlanta time-series epidemiologic study. We hypothesize that each increasingly complex exposure metric will show a greater degree of spatial and temporal variability in the exposure estimates, especially for traffic-related ambient pollutants. These refined exposure estimates may then provide greater power in detecting epidemiologic associations of interest for pollutants with heterogeneous or complex spatiotemporal patterns. Further details on the epidemiologic study design and the results from the related epidemiologic analyses using the various exposure metrics are described in two related companion papers (37, 38).

Materials and Methods

Study Design

The Atlanta study area encompassed 169 ZIP codes and extended about 70 km in each direction from the Atlanta city center. This analysis was performed on a subset of the 225 ZIP codes included in the larger Study of Particles and Health in Atlanta (SOPHIA) study, for the years 1999-2002. The ZIP codes selected were based on availability of data for all exposure estimation approaches, availability of census data for each ZIP code, and presence of the ZIP code during this study period (certain ZIP codes included in the original SOPHIA study were discontinued prior to 1999). Ambient pollutant data were measured and modeled for $PM_{2.5}$ and two of its components (EC and SO_4), and gaseous pollutants (O_3, CO and NO_x), on an hourly or 24-hr basis from 1999-2002. We developed the five metrics of exposure described below to characterize spatiotemporal patterns of ambient concentrations and population exposures to these six pollutants within the Atlanta study area. The similarities and differences in pollutant concentrations between exposure metrics were compared. We examined the spatial variability of exposure metrics across days and between exposure metrics, the temporal variability of exposure metrics, including: seasonal variability, level of temporal variability across ZIP codes and between exposure metrics, and daily correlations between exposure metrics for the six pollutants. General classes of exposure metrics are discussed in Özkaynak et al., 2012 (10). Each of these pollutant-specific alternative exposure metrics were subsequently applied in an epidemiologic analysis of daily emergency department visit data from each of the ZIP codes in the Atlanta study area. Results from the related health effects analyses are reported elsewhere as companion papers (37, 38).

Exposure metric i: Central site monitor measurements (CS)

Pollutant measurements from central monitoring sites in the study area comprise the primary exposure metric. Metric i included monitoring sites from the Southeastern Aerosol Research Characterization (SEARCH) network, the Assessment of Spatial Aerosol Composition in Atlanta (ASACA) network, and the EPA's Air Quality System (AQS) monitoring network. Details regarding the central sites selected for each pollutant, including their location, measurement methods and imputations done to fill in for missing data can be found elsewhere (39-41). In brief, hourly measurements for CO were from the Dekalb Tech AQS site, and hourly NO_x measurements were from the Georgia Tech AQS site. Hourly O_3 measurements for March/April – October were largely from the Confederate Ave AQS site; the Jefferson Street SEARCH site provided O_3 measurements for November – February. Daily 24-hr average $PM_{2.5}$, EC, and SO_4 concentrations were all from the Jefferson Street SEARCH site and have been detailed previously (39, 42) (Figure 1). Hourly data for CO and NO_x were aggregated to daily 24-hr average values; hourly data for O_3 were aggregated to daily 8-hr maximum values.

Exposure metric ii: Regional background (BG)

To create metric ii, we modified an earlier approach for creating population-weighted daily averages of ambient pollution concentrations to create spatially resolved hourly estimates of regional background pollution by removing local source impacts modeled by hour-of-day and day-of-week (12). The modified approach took ambient CS monitor hourly measurements for each pollutant and removed local source contributions as modeled by AERMOD (see exposure metric iii below) to infer hourly estimates of regional background pollution at each monitoring site, later interpolated to ZIP code centroids as described below. Hourly measurement data from six NO_x monitors, four CO monitors, 14 O_3 monitors, and five $PM_{2.5}$ monitors were used in this study; two $PM_{2.5}$ composition monitors provided 24-hr measurements of EC and SO_4. For details of locations of monitors used for creating BG estimates see Figure 1. Regression models were developed to predict hourly EC and SO_4 from 24-hr measurements (for additional details see Supplemental Text 1).

The local source contribution at each monitor location for each pollutant of mainly primary source origin (CO, NO_x, $PM_{2.5}$, and EC) was modeled as a function of hour-of-day, day-of-week, month-of-year, and year using AERMOD. These modeled contributions were then removed from the hourly regulatory ambient CS measurements to yield regional background estimates. For the remaining two pollutants that are almost entirely of secondary origin (O_3 and SO_4), the regional background was assumed to be the same as measured by the ambient monitors. Having estimated hourly regional background pollution levels at central monitoring sites, these estimates were spatially translated across the study domain as described in Supplemental Text 1.

Exposure metric iii: AERMOD

Local-scale hourly pollutant concentrations for $PM_{2.5}$, EC, SO_4, CO, and NO_x at each ZIP code centroid were also modeled using the AERMOD dispersion model version 09292 (43). AERMOD simulates concentrations of pollutants directly emitted into the atmosphere. Because O_3 is formed by photochemical processes and has no direct emissions, O_3 concentrations were not modeled with AERMOD. SO_4 concentrations output from AERMOD are from direct vehicle exhaust emissions, and do not include the secondary SO_4 contribution due to photochemical transformations in the atmosphere. The AERMOD model provides near-source pollutant contributions from each stationary source at receptors on a designated spatial scale by using emission source coordinates and stack parameters. To estimate mobile source contributions to roadway concentrations, we treated individual road links as elongated area sources in AERMOD. After modeling, the contributions to air quality from all sources were added together at each receptor (located at each ZIP code centroid).

Local emissions source data and meteorological data were input into the AERMOD model, with major stationary source emissions data (including airport sources at Hartsfield-Jackson Atlanta International Airport) coming from the EPA's National Emissions Inventory (NEI) from 2002. Roadway emissions were estimated using detailed road network locations from an improved methodology developed by the authors for a previous study (11), with link-specific highway vehicle emission rates estimated as the product of traffic activity by vehicle class on individual road links and running emission factors by vehicle class. Non-running vehicle emissions (e.g. idling emissions) were treated as part of background. Meteorological data came from the National Weather Service site at the Hartsfield-Jackson Atlanta International Airport and the Jefferson Street SEARCH site. For detailed specifications of the AERMOD model, see Supplemental Text 2. For details on model evaluation, see Supplemental Text 3.

Exposure metric iv: Hybrid

As part of metric iv, we used a combination of local- and regional-scale modeling to account for all major atmospheric processes, including local contributions (driven by local-scale variation in pollutant emissions and meteorology) and regional contributions (background levels associated with large-scale synoptic patterns), to provide spatially- and temporally-resolved concentration surfaces in Atlanta. The sum of the regional background contribution (metric ii) and the local contribution from AERMOD (metric iii) was computed hourly to obtain total ambient air concentrations for the each pollutant being studied, at each ZIP code centroid. As AERMOD does not estimate O_3 concentrations, the hybrid exposure metric for O_3 was identical to the regional background.

Exposure metric v: APEX and SHEDS exposure models

Models were used to estimate population exposures to ambient pollution, rather than approximating exposure using outdoor ambient pollutant concentrations (as in metrics i-iv), at each ZIP code and for each pollutant. As part of metric v, we used the U.S. EPA's Stochastic Human Exposure and Dose Simulation (SHEDS) model (19, 44, 45) to estimate 24-hr $PM_{2.5}$, SO_4, and EC exposures, and 8-hr maximum O_3 exposures. The U.S. EPA's Air Pollutants Exposure Model (APEX) (46, 47) outputs hourly estimates of exposure to CO and NO_x, which were aggregated to 24-hr average exposures (APEX estimates were used for CO and NO_x as model runs for the Atlanta study area had previously been completed). The SHEDS and APEX models estimate population exposure distributions by accounting for both the spatial variability in pollutant concentrations in locations where people are exposed (outdoor, indoor, and in-vehicle), and person-level variability in locations visited and time spent in each microenvironment, as the simulated individuals move about the study domain. Key input to the models were the hybrid pollutant concentrations from metric iv, described above, time-location-activity data from the U.S. EPA's Consolidated Human Activity Database (CHAD) (48), spatially varying local air exchange rates calculated as described in Sarnat et al. 2013 (38), and census tract-level home-to-work commuting data (47, 49). Penetration and decay parameters used in the models were specific to each pollutant, but did not vary spatially or temporally. The exposure estimates represent exposure of individuals to ambient pollution resulting from time spent in outdoor, indoor, or vehicular microenvironments; the APEX and SHEDS models included infiltration of ambient pollution to indoor microenvironments, but for this application did not include the contribution from indoor source emissions due to the intended subsequent application of the exposure estimates in an epidemiological analysis of health effects of air pollution due to ambient sources. For detailed SHEDS and APEX modeling specifications including penetration and decay parameters, see Supplemental Text 4-5, and Tables S2-S5; for details on model validation, see Supplemental Text 3.

Statistical methods

Summary statistics and Pearson correlations between exposure metrics are described below. The coefficient of variation (CV) for each pollutant was calculated to allow for comparison of the differences in spatial and temporal variability across pollutants. Defined as CV = σ/μ, where σ = standard deviation and μ = mean, the CV is a dimensionless index which allows for a normalized way to compare variability across pollutants with different units. A higher CV indicates a greater degree of dispersion of the variable. We define the "spatial" CV as the CV calculated across ZIP codes over the study domain, thus resulting in one spatial CV for each day (n~1461 for each pollutant, and each metric), and quantifying the amount of spatial variation in daily pollutant concentrations. The "temporal" CV was calculated as the CV across days, with one temporal CV for each ZIP code (n=169 for each pollutant, and each metric), and representing the degree of temporal variability of pollutant concentrations over the entire domain. GIS mapping was used to visually depict spatial variation. Pearson correlations were used to compare temporal correlations for each pollutant between exposure tiers.

All statistical analyses were completed in R version 2.13.2 (R Foundation for Statistical Computing, Vienna, Austria). All mapping was done in ArcGIS 10 (Esri, Redlands, CA).

Results

Summary statistics (Table S1) and comparison between exposure metrics (Figure 2) for each pollutant show that for all pollutants, the magnitude of 4 year annual mean hybrid estimates across all ZIP codes approximate the magnitude of CS monitor measurements well. In comparison, exposure model estimates are lower than ambient concentrations for $PM_{2.5}$, SO_4, EC, and O_3 due to reduced residential infiltration and removal of these pollutants indoors, and higher than ambient concentrations for CO and NO_x due to inclusion of a roadway proximity factor in the APEX model. For detailed discussion of the comparison between metrics for each pollutant, and of the seasonal variability for each pollutant, see Supplemental Text 6.

Spatial variability

As noted above and seen in the spread of the boxplots in Figure 2, there are differences in the amount of spatial variation in annual mean concentrations between ZIP codes for the various pollutants and metrics. Figure 3 displays the spatial CVs that quantify this spatial variation. Metrics iv and v have varying degrees of spatial variability for each pollutant, evidenced by the range of spatial CVs (Figure 3). While mean ambient concentrations from the hybrid estimates (metric iv) agree with those from CS measurements (metric i), hybrid estimates vary spatially, particularly for pollutants with predominantly local sources (EC, CO, and NO_x). These results are consistent with findings from previous work conducted in Atlanta showing increased spatiotemporal variability for traffic related pollutants (31).

The varying degrees of spatial variability can also be seen visually in the selection of maps presented in Figure 4. The spatial variability in ambient concentrations and larger spatial CVs observed for metric iv for EC, CO, and NO_x, is also reflected within the exposure modeling (metric v) (Figures 3, S1), partially due to the fact that metric iv concentrations are used as inputs in calculating the metric v estimates, but also suggesting spatial variability in population exposures. We also observe noticeable differences in both the magnitude and structure of the variability in distributions between metrics iv and v, likely due to space and time dependent mobility and infiltration factors incorporated in the exposure models.

There is little difference in the spatial CVs for $PM_{2.5}$, SO_4, and O_3, both between metrics iv and v and within metric iv or v for each pollutant, with little to no spatial variation in either metric for these three pollutants (mean spatial CV < 0.16) (Figures 3, S1). The low spatial CV for $PM_{2.5}$ and SO_4 may be because $PM_{2.5}$ and SO_4 are largely derived from regional sources, as seen in Figure 2 where the BG contribution dominates over AERMOD. Thus we do not expect to see spatial variability in the concentrations of these pollutants at the ZIP code level, and over the geographic scale of our study area. O_3 concentrations are likely spatially homogeneous in our study area as they are mostly driven by regional photochemistry at the ZIP code level. Note that these pollutants may have increasing degrees of spatial variability if more enhanced modeling or measurement data in fine-scale microenvironments (e.g. near roadway, and at varying distance from roadway) were being analyzed. PM composition, especially the ultrafine component for example, varies considerably when a finer spatial scale than ZIP code level is examined (29, 50). In addition, the fine-scale variation in O_3 photochemistry, particularly near busy roadways, was not considered in our local emissions model (AERMOD).

Local pollutants show a different pattern, with moderate spatial variation for EC (Figure 4) (mean spatial CV of ~0.5 for metrics iv and v). While CO has low-moderate and NO_x has moderate-high spatial variability for metrics iv and v, for both pollutants the spatial CV of the exposure model estimates is lower than the spatial CV of the hybrid estimates (Figure 3). EC, CO, and NO_x all exhibit a range of spatial CVs across the days covered by the study period, evidenced by the wider boxplots (Figure 3). As shown previously, spatial variability in ambient concentrations of these pollutants is expected since their main source is local traffic emissions (31). The lower degree of spatial variability across days of exposure model estimates (metric v) compared to hybrid estimates (metric iv) for CO and NO_x is potentially due to the relatively uniform air exchange rates used as input for the exposure model, or to the influence of mobility and commuting related exposure factors in these models that are accounting for the movement of individuals between high and low concentration areas (18). As a result of movement of commuters between multiple ZIP codes on a given day, the daily average exposure concentrations for commuters may be more similar than ambient concentrations of each ZIP code individually.

Temporal variability

Temporal CV

For all pollutants except NO_x, there is no substantial difference in the mean temporal CV across ZIP codes when comparing the three main exposure metrics (metrics i, iv and v) (Figure 5). This indicates that the overall degree of temporal variability of $PM_{2.5}$, SO_4, EC, CO, and O_3 is similar across the exposure metrics. For $PM_{2.5}$, SO_4, and O_3, the narrow boxplots indicate little difference in the degree of temporal variability across ZIP codes for metrics iv and v compared to EC, CO, and NO_x, where wider boxplots indicate differences in the degree of temporal variability across ZIP codes not represented in CS measurements. NO_x exhibits a unique pattern, with the overall degree of temporal variability (i.e. the mean temporal CV) decreasing as the complexity of the exposure metric increases (highest mean temporal CV for the CS measurements (metric i), lowest mean temporal CV for the exposure model estimates (metric v)) (Figure 5). This result is potentially due to regularizing effects of commuting and related exposure factors varying across the study domain.

In breaking down the hybrid estimates into the two component parts (metric iii: AERMOD and metric ii: BG), we see that for $PM_{2.5}$ and SO_4, AERMOD does provide added temporal variability compared to the CS measurements (metric i), evidenced by the wider boxplots for metric iii (Figure 2). However as described above, the magnitude of the AERMOD contributions from local primary emissions for $PM_{2.5}$ and SO_4 are so low compared to the regional contributions that this added temporal variability is lost when the AERMOD and regional components are combined.

Correlation between metrics

The regional pollutants ($PM_{2.5}$, SO_4, and O_3) all exhibit a strong daily correlation between the CS measurements (metric i) and the hybrid estimates (metric iv) (r=0.90, 0.95, and 0.97 respectively), and between the CS measurements (metric i) and the exposure model estimates (metric v) (r=0.84, 0.93, and 0.93 respectively), indicating that the CS measurements co-vary over time with both the hybrid estimates and the exposure model estimates (Table 1). In studies where the day-to-day variability of ambient concentrations is desired, hybrid estimates obtained using the methods presented here may not provide greater temporal resolution as compared to CS measurements for these pollutants. It is important to note that in this case, the strong correlation between CS measurements and hybrid estimates are partially due to the CS measurements being used as input for the BG estimates (metric ii), which in turn are used in calculating the hybrid estimates. The strong correlation between CS measurements and exposure model estimates is consistent with previous findings of a strong correlation between ambient concentrations and personal exposures for $PM_{2.5}$ and SO_4 (51-54). Previous studies have typically found a weak correlation between ambient concentrations and exposure estimates for O_3 (51, 52, 55) with the main assumption that low infiltration and high removal rates indoors may have contributed to this weak correlation. In this instance, stronger correlations between CS measurements and exposure model estimates for O_3 may be due to O_3 exposures being based on BG estimates only (i.e. no metric iii: AERMOD modeling was done for O_3), and due to the SHEDS model predictions including O_3 infiltration and decay parameters which do not vary temporally.

Pollutants dominated by local emissions sources (EC, CO, and NO_x) exhibit a moderate daily correlation between the CS measurements (metric i) and the hybrid estimates (metric iv) (r=0.70, 0.54, and 0.58 respectively), and between the CS measurements (metric i) and the exposure model estimates (metric v) (r=0.64, 0.63, and 0.73 respectively) (Table 1). This moderate correlation indicates that day-to-day variability at the CS is different than that represented by the hybrid estimates or the exposure model estimates, which may influence epidemiological study results depending on the co-variance between exposure and health outcome data at the ZIP code level. The decrease in the metric i – metric iv and metric i-metric v correlations for local pollutants as compared to regional pollutants may be a result of the hybrid and exposure model estimates accounting for local traffic related sources of emissions which may vary on a day-to-day basis (8, 29, 30). This temporal variability in local source emissions may not be captured by CS measurements. Previous studies have found weak correlations between ambient concentrations and personal exposures for NO_2 (51, 52, 56), potentially for the same reasons as explained above and potential NO_x exposures from indoor combustion sources.

All pollutants exhibited strong correlations between hybrid estimates (metric iv) and exposure model estimates (metric v), with correlations ranging from 0.82 to 0.98 for the six pollutants. The strength of these correlations indicate that the day-to-day variability of hybrid estimates as compared to exposure model estimates is quite similar, most likely because exposure models used the hybrid estimates as a main input. If temporal variability alone is of interest (e.g., in a time-series study in a geographically small study area) for these six pollutants, it may not be necessary to consider both hybrid and exposure modeling to obtain adequate estimates of temporal variability for these pollutants.

Influence of spatial and temporal variability on exposure metrics for EC

In Figure 6, we have highlighted EC as an example of a spatially and temporally varying pollutant dominated by local emissions sources which benefits from the modeling approaches presented here. The hybrid estimates (Figure 6c) provide added spatial variability which is not present in the CS measurements (Figure 6b), highlighting the differential exposure misclassification which could occur on the spatial scale for EC if CS measurements were used as the exposure estimate in epidemiologic analyses with geographically-defined subpopulations. The exposure model estimates (Figure 6d) provide an additional benefit for epidemiology studies as these estimates take into account the added spatial variability of hybrid estimates (due to hybrid estimates being used as input to the exposure model estimates), yet also account for the spatially varying air exchange rates in the study area (Figure 6a). Additional exposure factors such as commuting patterns and differences in time-location-activity budgets included in the exposure model result in a reduced magnitude of the exposure model estimates (Figure 6d) compared to hybrid estimates (Figure 6c). The slight reduction in the spatial CV of EC (Figure 3) when comparing exposure model and hybrid estimates may be a result of commuting patterns, whereby commuting between ZIP codes in a given day will result in less spatial variability in exposure model estimates as compared to hybrid estimates, which are inherently modeled for each ZIP code individually. In addition, Figures 6e and 6f demonstrate that the degree of temporal variability present in the EC concentrations changes across the study domain, and is dependent on the spatially varying EC concentrations, highlighting the potential for introducing differential exposure misclassification on the temporal scale if the non-spatially varying CS measurements were used as the exposure estimate in epidemiologic analyses.

Discussion

Though previous related epidemiology studies of spatially variable pollutants in this same study area of Atlanta have reported associations with cardiovascular and respiratory outcomes using CS monitor measurements (40, 41, 57-59), simulation studies have suggested that error due to spatial variability in ambient pollution may result in reductions in observed relative risks by 43-68% for spatially heterogeneous pollutants such as CO, NO_x, and EC (60). This finding further motivated us to assess the potential for exposure misclassification when using CS measurements in a health study (60, 61) so that error might be minimized in future epidemiological studies.

To summarize our findings, air quality modeling of ambient concentrations (metric iv) approximates mean CS measurements (metric i) well, but includes a degree of spatial variability of ambient concentrations that CS monitor measurements do not capture, especially for local pollutants (EC, CO, and NO_x). Human exposure models incorporating infiltration parameters, time-location-activity budgets, and other exposure factors also introduce a certain level of spatial variability for local pollutants. The mean level of temporal variability across ZIP codes for all pollutants except NO_x is represented well by CS measurements, however for local pollutants there is a range of temporal variability across ZIP codes that is not represented in CS measurements.

In applying the results of this analysis to exposure metrics for future epidemiologic studies, there are a few key points to consider. First, exposure models not only introduce variability in predicted exposures, but may impact the magnitude and distribution of the predicted exposure concentrations both within and across ZIP codes. Second, exposure misclassification on both the spatial and temporal scales may be introduced for local pollutants if CS measurements are used as the estimate of exposure, due to the spatial and temporal variability of local pollutant concentrations or spatially varying exposure factors (especially infiltration and commuting patterns) which are not accounted for in CS measurements (37). Though air quality and exposure models have the ability to introduce variability not present in CS measurements, the potential to introduce greater uncertainty in the resultant health effect estimates due to modeling error must be considered (4).

When regional pollutants ($PM_{2.5}$, SO_4, O_3) are of interest, CS measurements may be sufficient to reflect spatial variability, especially for time-series or case-crossover studies over large urban or metropolitan scales, due to the limited local-scale spatial variability of these pollutants at the ZIP code level, and due to the strong temporal correlation between CS measurements and either hybrid or exposure model estimates. However, in studies of local pollutants (EC, CO, and NO_x), both air quality modeling and exposure modeling may need to be considered in order to represent spatial variability adequately. In addition, air quality and exposure modeling represent different levels of temporal variability for local pollutants compared to CS measurements. While the strong correlation between hybrid and exposure model estimates for local pollutants suggests that hybrid and exposure models comparably represent day-to-day variability, it is important to remember that exposure modeling estimates may represent differences in the magnitude and spatial variability of pollutants that the hybrid estimates do not. For all pollutants, if the appropriate magnitude of exposure is desired at a fine spatial and temporal resolution (i.e., when fine scale spatiotemporal health data are available), exposure models may be necessary to better represent levels of human exposure because of the variety of human exposure factors which they incorporate.

To our knowledge, no other single study has developed the diverse range of spatial and temporal refinement in exposure metrics presented here, compared both air quality and exposure model results to central site monitor measurements, and further done so for multiple pollutants. Including alternative exposure assessment approaches in one study allows for a direct comparison of how different methods perform relative to each other. This study provides support for the development of alternative approaches for specific epidemiologic applications (9, 59, 62-65). While the study aimed to reduce exposure misclassification for traffic-related pollutants (CO, NO_x, and EC), pollutants of secondary origin (O_3 and SO_4), which previous work has shown have little spatiotemporal variation in Atlanta were included for comparison (31). The inclusion of six pollutants allowed for comparison of how the alternative approaches perform when applied to pollutants with varying spatial and temporal patterns. In addition, the estimates developed allowed for the investigation of how these exposure surrogates might improve health effect estimates in a time-series study (37, 38).

Results from this study should be followed with additional studies analyzing exposure estimates from multiple alternative exposure estimation approaches at different geographic locations. Studies in locations with different meteorological conditions (e.g., the North-East where the residential air exchange rates may be more variable), or with different emissions profiles (e.g., greater quantity of industrial sources, less traffic, or a more concentrated city center), may yield different results. Conclusions regarding spatial variability may also vary when modeling is conducted at finer spatial scales (e.g., $PM_{2.5}$ or ultrafine PM may show increased spatial variability in near-road environments). Further, the resources required for completing local-scale modeling for $PM_{2.5}$ and SO_4 in the future should be weighed against the local versus regional contribution for these pollutants, keeping in mind that while $PM_{2.5}$ concentrations measured at central sites within an urban area may be highly correlated, some variation in their concentrations can occur spatially on any given day, especially when analyzed at a finer spatial scale (66). Temporal variability may differ in areas where there are greater variations in meteorology from day-to-day. Lastly, patterns of exposure model estimates may change in areas where air exchange rates are higher than those in Atlanta. For certain pollutants, the spatial and temporal variability added when using air quality and exposure models demonstrates the potential for exposure misclassification when using CS measurements as estimates of exposure in an epidemiologic study. Keeping the results of this analysis in mind, there must be careful consideration in future epidemiologic studies of the choice of the exposure assignment approach, with consideration given to the epidemiological study design, pollutant of interest, and temporal and spatial scales of both exposure and health data.

Supplemental Information: Supplemental information is available at the Journal of Exposure Science and Environmental Epidemiology's website at: http://www.nature.com/jes/index.html.

Conflict of Interest:

The authors declare no conflict of interest.

Acknowledgments & Disclaimer:

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Figures

Figure 1: Map of metropolitan Atlanta with monitoring site locations and population density. Letters reference monitor locations. The table identifies station name, network, and air pollutants monitored, with air pollutants indicated by numbers (1=NO_2/NO_x, 2=CO, 3=O_3, 4=$PM_{2.5}$ mass, 5=$PM_{2.5}$ composition (SO_4, EC). Population density is from 2000 Census data.

Figure 2: Mean annual pollutant concentrations for each study ZIP code in Atlanta, GA (1999-2002). Note metric i: CS, metric ii: Regional Background, metric iii: AERMOD, metric iv: Hybrid, and metric v: APEX or SHEDS. Bottom and top of box represent 25th and 75th percentiles, the band near the middle of the box is the median, and the ends of the whiskers are the 5th and 95th percentiles. (Note n=169 for each pollutant, for each metric.)

Figure 3: Mean spatial CV for each day (1999-2002). Note metric i: CS, metric iv: Hybrid, and metric v: APEX or SHEDS. Bottom and top of box represent 25th and 75th percentiles, the band near the middle of the box is the median, and the ends of the whiskers represent the 5th and 95th percentiles. The spatial CV = 0 for CS monitor measurements because the same CS measurement was applied to each ZIP code.

Figure 4: Selection of GIS maps of Atlanta metropolitan area showing spatial variability for annual means of $PM_{2.5}$, EC, and NO_x for metric iv: Hybrid. Boundaries delimited on maps are ZIP code boundaries. GIS maps for the full set of metrics, for all pollutants, all seasons, can be found in Figure S1.

Figure 5: Mean temporal CV for each study ZIP code in Atlanta, GA (1999-2002). Note metric i: CS, metric iv: Hybrid, and metric v: APEX or SHEDS. Bottom and top of box represent 25th and 75th percentiles, the band near the middle of the box is the median, and the ends of the whiskers represent the 5th and 95th percentiles. The temporal CV for metric i is the same for each ZIP code because the same CS measurement was used for each ZIP code.

Figure 6: Influence of spatial and temporal variability on exposure metrics for EC.

a) GIS map of annual mean air exchange rate for each ZIP code.

b) GIS map of annual mean EC concentration (metric i: CS) for each ZIP code.

c) GIS map of annual mean EC concentration (metric iv: Hybrid) for each ZIP code.

d) GIS map of annual mean EC concentration (metric v: SHEDS) for each ZIP code.

e) GIS map of annual mean temporal CV for each ZIP code for EC (metric iv: Hybrid).

f) Temporal CV vs. annual mean concentration for each ZIP code (metric iv: Hybrid).

Table 1: Mean and standard deviation of ZIP code-specific Pearson correlations between exposure metrics in Atlanta, GA: 1999-2002. (Correlations between metrics for each ZIP code were calculated separately – the mean and standard deviation of the correlations across all ZIP codes are presented here.)

		$PM_{2.5}$	SO_4	EC	O_3	CO	NO_x
Metric i – Metric iv (CS – Hybrid)	Mean ± SD	0.90 ± 0.03	0.95 ± 0.01	0.70 ± 0.08	0.97 ± 0.02	0.54 ± 0.07	0.58 ± 0.08
	Winter	0.84 ± 0.04	0.94 ± 0.02	0.63 ± 0.11	0.92 ± 0.07	0.44 ± 0.08	0.48 ± 0.08
	Spring	0.88 ± 0.04	0.91 ± 0.02	0.69 ± 0.09	0.96 ± 0.03	0.52 ± 0.10	0.46 ± 0.09
	Summer	0.90 ± 0.04	0.94 ± 0.01	0.61 ± 0.08	0.94 ± 0.04	0.47 ± 0.11	0.42 ± 0.11
	Fall	0.93 ± 0.02	0.93 ± 0.01	0.78 ± 0.07	0.97 ± 0.02	0.62 ± 0.07	0.66 ± 0.12
Metric i – Metric v (CS – APEX/SHEDS)	Mean ± SD	0.84 ± 0.02	0.93 ± 0.01	0.64 ± 0.07	0.93 ± 0.02	0.63 ± 0.04	0.73 ± 0.04
	Winter	0.80 ± 0.02	0.90 ± 0.02	0.58 ± 0.07	0.85 ± 0.06	0.57 ± 0.06	0.68 ± 0.05
	Spring	0.82 ± 0.04	0.88 ± 0.01	0.62 ± 0.07	0.88 ± 0.03	0.63 ± 0.05	0.63 ± 0.07
	Summer	0.84 ± 0.03	0.92 ± 0.01	0.57 ± 0.08	0.89 ± 0.04	0.54 ± 0.08	0.55 ± 0.06
	Fall	0.86 ± 0.02	0.91 ± 0.01	0.73 ± 0.08	0.92 ± 0.02	0.69 ± 0.03	0.79 ± 0.03
Metric iv – Metric v (Hybrid – APEX/SHEDS)	Mean ± SD	0.93 ± 0.01	0.98 ± 0.01	0.94 ± 0.01	0.96 ± 0.01	0.83 ± 0.07	0.82 ± 0.09
	Winter	0.89 ± 0.02	0.96 ± 0.01	0.88 ± 0.03	0.93 ± 0.01	0.76 ± 0.06	0.73 ± 0.07
	Spring	0.92 ± 0.01	0.97 ± 0.00	0.94 ± 0.02	0.90 ± 0.02	0.80 ± 0.07	0.78 ± 0.09
	Summer	0.94 ± 0.01	0.98 ± 0.01	0.98 ± 0.01	0.94 ± 0.01	0.82 ± 0.09	0.81 ± 0.17
	Fall	0.93 ± 0.01	0.97 ± 0.00	0.97 ± 0.01	0.95 ± 0.01	0.89 ± 0.08	0.86 ± 0.12

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