AIR QUALITY MODELING NEEDS FOR EXPOSURE ASSESSMENT FROM THE SOURCE-TO-OUTCOME PERSPECTIVE

by

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All biological organisms, including humans, are exposed continuously to mixtures of air pollutants; the compositions of these mixtures vary with time and location and their components originate from many types of sources, both local and distant, including, among others, industrial facilities, vehicles, consumer products, etc. Exposure characterization is often the weakest link in the "source-to-outcome" sequence of processes and events (Figure 1) affecting human and ecological health risks from environmental pollutants. It is recognized[1] that it is generally easier to characterize exposures for ecosystems than for human populations, as in the latter case exposures can be particularly sensitive to high-resolution spatial and temporal variations in ambient concentrations and the "micoenvironmental" adjustments imposed by a variety of indoor and outdoor settings (occupational, residential, recreational, commuting, etc.). Ultimately, quantifying inhalation exposures of humans to atmospheric contaminants, such as criteria air pollutants and air toxics, requires characterization of the air flow that enters the human respiratory tract, i.e. "personal air." Assessing personal air concentrations, in turn, requires characterization of concentrations in residential and occupational microenvironments as well as at local (''neighborhood'') ambient scales. The constituents of the local outdoor air may originate, however, from a variety of distances, from regional to continental and beyond. Exposure modeling, therefore, involves processes spanning a wide range of spatial and temporal scales.

The last decade has seen an evolution in the practice of exposure assessment, with the focus changing from considering "potential exposures" to a single pollutant, that would occur outdoors at a given location or across an area of concern, to "person oriented" multipollutant exposure assessments. So, current assessments take into account the behavioral and physiological dynamics of contact with various contaminants, as individuals (actual or "virtual" in the case of computer simulations) "move" in different indoor and outdoor "microenvironments" while engaged in various activities that determine rates of contact and uptake of multiple pollutants. To characterize inhalation intakes, airborne concentrations of co-occurring pollutants have to be determined for each individual at the spatial and temporal scales defined by the microenvironments and exposure activity events. This progress was made possible by the availability of enhanced computational modeling resources, widespread GIS applications, new databases on human activities, demographics, microenvironmental attributes, emissions, etc.[2] However, the use of more comprehensive realistic frameworks for exposure assessment raises additional needs for air quality models. These needs are discussed here, following a brief summary of the current status of inhalation exposure models.

It should be pointed out that the focus of the present discussion is specifically on modeling needs related to *inhalation exposures of airborne contaminants*. Of course, there are situations where these contaminants affect other exposure pathways; for example, the dominant pathway of human exposure to methylmercury is fish consumption and, in this case, air quality modeling is

needed to characterize atmospheric deposition on watersheds etc. However, discussion of multipathway exposures from contaminants in multiple media is beyond the scope of this review.

1. INFORMATION NEEDS FOR EXPOSURE ASSESSMENT

Exposure is the "contact of a biological receptor with the contaminants of concern," resulting in the *intake* and subsequent (systemic) *uptake* of these contaminants; information necessary for calculating health-relevant metrics of exposure includes time-course profiles of concentration levels, frequency, duration, etc. of the contact. The environmental (physicochemical) and biological properties of the contaminants determine the relevant time scales and corresponding temporal and spatial averaging and sampling practices relevant to assessments of exposure and associated health risks. Potential health effects associated with the contaminants of concern determine the types of exposures that need to be considered (acute, subchronic, chronic) and subsequently the types of environmental, behavioral (activity patterns), and demographic information that needs to be collected. Spatial and temporal scales and resolutions can differ widely in exposure assessments, depending on the situation of concern; in general, the resolutions of the analysis should be consistent with the exposure events, while the time scale should be relevant to the health effects of concern.

In situations involving *acute inhalation exposures*, as for example, in cases of accidental releases of chemicals, the criteria for limiting potential outdoor contacts are provided through the "Acute Exposure Guideline Levels" (AEGLs), which combine concentration levels and contact durations to relate exposures to different harmful effects[3]. The National Advisory Committee for AEGLs has been developing these guidelines to help both federal and local authorities, as well as private companies, deal with emergencies involving spills, or other accidental releases of hazardous chemicals. The AEGL values are intended to serve for alert and emergency response planning as well as for disaster control. AEGL values represent toxicologically relevant "ceiling" exposure levels for different relevant exposure periods (10 min, 30 min, 1 hr, 4 hr, 8 hr) and for three different degrees of severity of toxic effects, i.e. a threshold for notable discomfort (AEGL-1), a threshold for serious, long-lasting effects or an impaired ability to escape the area of exposure (AEGL-2), and a threshold for lethal effects (AEGL-3).

In situations involving *sub-chronic and chronic inhalation exposures*, it was common practice -- up to the recent past -- to use ambient levels of concentrations of criteria pollutants and air toxics as inputs to epidemiological studies addressing questions on human health risks. Specifically, outdoor concentrations, either from ambient "central" fixed monitors or from numerical simulation models have been used as surrogates for personal exposure. However, *"A key problem in using modeled or monitored ambient concentration data to estimate exposures is the fact that people in most societies spend most of their time indoors"*[4]. The fact that actual human exposures are dominated by personal activities and the attributes of microenvironments (see, e.g., Figure 2) is now gaining universal recognition. Figure 3 further illustrates this point by presenting the distributions of simultaneous measurements of background, local outdoor, indoor, and personal air concentrations for three pollutants with different physicochemical properties (benzene, formaldehyde, and $PM_{2.5}$) collected for over 300 homes of non-smokers in three US cities representing different climatological and demographic settings (Elizabeth, NJ, Houston, TX, and Los Angeles, CA). It is clear that personal exposures cannot be adequately represented by ambient concentration levels. Figure 4, which presents predicted concentrations *vis a vis*

inhaled doses of $PM_{2.5}$ in Philadelphia, PA, provides an example of a modeling analysis consistent with this fact.

It should be noted that exposure science is a rapidly evolving field and the development of a "standard" and commonly accepted terminology is an ongoing process. Very often, procedures that are called "exposure modeling", "exposure estimation", "exposure assessment", etc. may in fact refer to only a sub-set of the components required for exposure characterization, e.g., modeling local dispersion patterns of a contaminant. Though not complete exposure studies per se, such efforts have value, as they potentially improve individual components of a comprehensive assessment. Nevertheless, in the following, the terms "exposure model" or "modeling" will refer specifically to formulations that explicitly describe contact with contaminants by considering (a) microenvironmental attributes such as concentration levels, (b) behavioral attributes such as activities of individuals in a given microenvironment, and (c) biological attributes such as gender, age, weight, body mass index, etc.

2. EVOLUTION AND CURRENT PRACTICE OF EXPOSURE MODELING

Existing comprehensive inhalation exposure models consider the "movement" of individual human subjects (actual or "virtual"), or of appropriately defined cohorts, in space and time, as sequences of "activity" or "exposure events". In these sequences, each event is defined by time, a geographic location, a microenvironment and the activity of the subject. USEPA has supported comprehensive efforts in developing models implementing this general concept, and these efforts have resulted in the NEM/pNEM (National Exposure Model and Probabilistic National Exposure Model[5]), HAPEM (Hazardous Air Pollutant Exposure Model[6]), SHEDS (Simulation of Human Exposure and Dose System[7]), APEX (Air Pollutants Exposure model[8]), and MENTOR (Modeling Environment for Total Risk studies[9]) families of models. Table 1 summarizes essential attributes of these inhalation exposure models that currently also represent the state-of-the-art in the field. The overall approach followed in these models in general consists of the following components:

1. Estimation of the *background or ambient levels* of the pollutants of concern via (a) spatio-temporal analysis of fixed monitor data, (b) emissions-based air quality modeling applied in a coarse resolution mode at the regional-to-urban scales, or (c) a combination of model output and observations.
2. Estimation of *local outdoor levels* of the pollutants of concern. These levels would typically characterize the ambient air of either an administrative unit (such as a census tract) or a conveniently defined high resolution grid cell of an urban-scale air quality model. This component can also involve (a) spatio-temporal statistical analysis of local monitor data, (b) application of grid-based air quality models at their highest resolution (typically around 2-4 km), (c) application of a "sub-grid" scale modeling employing local-scale dispersion models, or (d) "refinement" of the estimates of a regional model using schemes that take into account subgrid variation in topography and land-use combined with considerations of sub-grid transformation and mixing processes.
3. Characterization of relevant *attributes of the individuals or populations* under study (residence and work locations, occupation, housing data, income, education, age, gender, race, weight, body mass index, and other physiological characteristics). These factors affect not only the physical activities, but also the intake rates and subsequent systemic uptakes of

chemicals. This component of the exposure analysis can be pursued by (a) assembling the required information for each actual person to be considered in the study, (b) selecting a fixed-size sample population of "virtual individuals" in a way that statistically reproduces essential demographics of the administrative population "unit" used in the assessment (e.g. a sample of 500 people can be used to represent the demographics of a given census tract), or (c) dividing the population-of interest into a set of cohorts representing selected subpopulations and defining one or more "representative individuals" for each cohort.

4. Development of *activity event (or exposure event) sequences* for each member of the sample population (actual or virtual), or for each cohort for the exposure period using (a) study-specific information, or (b) "sampling" from available time-activity databases (such as USEPA's Consolidated Human Activity Database[10]) using appropriate matching criteria for the virtual subjects.
5. Estimation of levels and temporal profiles of the pollutants in various specific *outdoor and indoor microenvironments* such as street canyons, roadway intersections, gas stations, school yards, parks, residences, offices, schools, restaurants, vehicles, etc. This is typically done through either (a) statistical analyses of observational datasets (typically for indoor/outdoor relationships or for vehicle/outdoor relationships) that produce microenvironmental factors, (b) simple microenvironmental mass balance models with or without atmospheric chemistry, (c) simplified microenvironment-specific dispersion models, such as street canyon models, (d) detailed Computational Fluid Dynamics models, possibly combined with chemistry, for indoor and outdoor microenvironments.
6. Calculation of appropriate *inhalation rates* for the members of the sample population, combining the physiological attributes of the (actual or virtual) study subjects and the activities pursued during the individual exposure events.
7. Calculation of *target tissue dose* through respiratory dosimetry modeling, if sufficient information is available.

The above components can be implemented in a "nested manner" in order to characterize both uncertainty and variability (intra-individual and inter-individual) involved in the exposure and dose assessment. In order to characterize the uncertainty in estimates of distributions of exposures, these calculations can be performed multiple times by sampling from distributions of corresponding inputs and parameters representing the uncertainties.

In practice, the majority of past exposure modeling studies have either incorporated only subsets of these components or treated some of them in a simplified manner, often focusing on selected factors affecting exposure. Of course, depending on the objective of a particular modeling study, implementation of a selected subset may in fact be adequate. For example, outdoor levels of pollutants, in conjunction with basic demographic information such as residential and occupational locations and commuting patterns of individuals, can be used to calculate metrics of potential population exposures associated with ambient air (as opposed to total exposures that would include microenvironmental adjustments and contributions from indoor sources); such metrics can be useful in comparing alternative scenarios related to different meteorology, emissions, etc. Indeed, though metrics thus derived would not be quantitative indicators of total human exposures, they can serve as surrogates of population exposures associated with outdoor air, and thus aid in regulatory decision making concerning pollutant standards and in studying the efficacy of emission control strategies. This approach has been used in comparative evaluations of regional and local emissions reduction strategies in the United States[11].

Air quality modeling plays a critical role in the steps associated with characterizing background, neighborhood, and microenvironmental concentration levels. Table 2 summarizes essential attributes of widely used air quality models in relation to different types of exposure characterizations. The next three sections identify specific air quality modeling needs, from an exposure assessment perspective, for regional, urban, local and microenvironmental scales.

3. AIR QUALITY MODELING NEEDS: REGIONAL AND URBAN SCALES

Model/Data Assimilation

Ambient (outdoor) concentrations of pollutants over a regional domain may be estimated either through (a) emissions-based mechanistic modeling, (b) through ambient-data-based modeling, or (c) a combination of both. Monitored ambient air pollutant levels generally reflect point measurements of atmospheric concentrations at individual locations near the ground. Air quality models provide spatially-resolved descriptions of pollutant concentrations across a geographical domain, but the model outputs reflect volume-average concentrations at each grid cell and can be inaccurate due to inadequate physics/chemistry of the model and errors in input data. In practice, the availability and accuracy of emissions inventories are often the limiting factors in applying mechanistic atmospheric fate and transport models on regional levels. In addition, for pollutants capable of long-range transport, either a global simulation must be done or boundary conditions must be supplied to account for the effects of sources outside of the region. Depending on the pollutant of interest, uncertainties in atmospheric chemistry, phase partitioning, and/or deposition phenomena can also limit the accuracy of regional/urban models.

Ambient-data-based models typically calculate spatial or spatiotemporal distributions of the pollutant through the use of interpolation schemes, that include various Bayesian approaches. These schemes employ deterministic or stochastic models for allocating monitor observations to the nodes of a virtual regular grid covering the region of interest. Geostatistical techniques such as Kriging provide various procedures for generating an interpolated spatial distribution for a given time, from data at a set of discrete points. However, Kriging techniques are severely limited by the nonstationary character of the atmosphere and the resulting concentration patterns of air pollutants; this lack of stationarity requires semivariograms that change with time[12]. Alternative approaches that interpolate monitor data simultaneously in space and time should be more applicable to air quality assessments.[13, 14] Fusion of model outputs and ambient measurements can be performed to improve the estimates of the spatial distribution of pollutant concentrations[15]. Such approaches include data assimilation methods[16] or simpler hybridization methods, using for instance Kriging of the model error[17].

Expansion of the One-Atmosphere Approach

As human and ecological exposure studies further recognize the need for multipollutant assessments that take into account synergistic effects of co-occurring contaminants, air quality models need to provide information on an increasing number of air pollutants, accounting for their emissions, transport and fate, as well as their patterns of co-occurrence and interactions ("one-atmosphere" approach). Various steps have taken place towards this objective, including for example, the incorporation of new versions of atmospheric chemistry mechanisms in models such as CMAQ[18], to consider a larger number of organic compounds in addition to those affecting the levels of criteria air pollutants. Furthermore, as biogenic emissions of allergens

such as pollen, fungal spores, etc., (see, e.g., Figure 5) are associated with similar endpoints (such as asthma) as photochemical oxidants[19], it is necessary to include them in future versions of regional air quality models.

Expansion of Multimedia Linkages of Air Quality Models

The role of air quality modeling has been expanding to support combined assessment of ecological and human health risks. A representative example is the application of models such as CMAQ and HYSPLIT to study regional-scale transport of mercury and its contribution to watershed loading[20, 21]. An example of a tool linking CMAQ, a grid-based photochemical air quality model (PAQM), with a watershed model is provided in Schwede et al.[22]. Linkages may be dynamic (on-line) or static (off-line) depending on the pollutant, the phenomena of interest, the different media involved, etc. The relevant spatial and temporal scales in different media can vary dramatically and the linking of models must consider these differences; the linkage of models for various media is in fact an emerging science. Lammel et al.[23] compared the predictions of multimedia models of different levels of detail for a set of non-ionic and moderately polar organic chemicals and found that simpler mass balance models tend to overestimate substance sinks in air and to underestimate atmospheric transport velocity due to neglect of spatiotemporal variability. The need for spatially resolved models is especially important for semi-volatile chemicals that have intermediate lifetimes and are therefore distributed regionally rather than locally or globally while cycling among various environmental media.

Implementation of Diagnostic Tools

In an accountability framework, assessment and management of exposures should be ultimately related to source activities, whether the sources are proximal or remote. Regional/urban air quality models employed in this context can be enhanced by diagnostic methods that allow computationally efficient characterizations of source-receptor relationships. Several techniques exist for developing source attribution for modeled concentrations and deposition, including approaches based on linear superposition[20, 24], source elimination[25], and emissions tagging[26]. The suitability, efficiency and inter-comparability of different approaches depend on the pollutant involved. Furthermore, several methods for "inverse modeling" and diagnostic sensitivity analysis can be applied to regional air quality models; these include direct techniques such as the Direct Decoupled Method (DDM[27]) and model adjoints[28], and surrogate techniques such as the High Dimensional Model Representation (HDMR[29]). These provide alternative means for assessing model response without requiring simulations for each combination of different variables. The DDM requires the addition of equations for sensitivity calculations to the original model source code, and calculates local derivatives with respect to perturbations in input parameters such as emissions, chemical reaction rates, and initial/boundary conditions. Similarly, model adjoints propagate perturbations, but for receptor-based metrics and backward in time. HDMR techniques are applied without alteration to the computational code model and rely on a "global" response of the model to changes in inputs.

4. AIR QUALITY MODELING NEEDS: LOCAL AND NEIGHBORHOOD SCALES

Local ("neighborhood" or "subgrid") spatial variability is a major issue with respect to characterizing local concentrations of contaminants in most exposure-relevant settings. For example, the fast rates of reactions in photochemical systems result in significant concentration

gradients in the vicinity of sources (e.g. titration of ozone by NOx emissions in the immediate vicinity of roadways). These gradients are not resolved directly by currently available grid-based PAQMs such as CMAQ and CAMx. "Plume-in-Grid" options have been developed for both these models and they can be used for large point sources (such as smokestacks) that exist within a grid cell. Nevertheless, plume-in-grid formulations will mostly resolve gradients in upper atmospheric layers and thus are not necessarily relevant to human exposure calculations, which are affected by gradients caused by a multiplicity of smaller ground-level or near-ground-level combustion sources.

Currently PAQMs are typically applied with horizontal resolutions ranging from 36 km to 2 km and a surface layer thickness that is of the order of 30 m. Though, computationally, it is possible to increase further the resolution of these modeling grids, there are theoretical limits imposed by assumptions inherent in the formulation of governing equations for transport and transformation in these models, therefore, a resolution of the order of 2 km is the highest allowable by current grid-based PAQMs[30]. Application of PAQMs to urban domains is further complicated by urban topography, the urban heat island, etc. It is beyond the scope of the present discussion, to overview the various issues relevant to urban fluid dynamics and related transport/fate processes of contaminants; various reviews of these issues and of available approaches for modeling urban fluid mechanics and dispersion are available[31,32].

One way of accounting for subgrid concentration gradients is the so-called "hybrid modeling" approach[33, 34], where concentrations from a grid-based PAQM (such as CMAQ) and a local plume dispersion model (such as AERMOD) are added to provide total contributions from regional transport/chemistry and from local-scale dispersion. In such an approach it is important to avoid "double counting" sources in the two models[35]. Furthermore, since local dispersion models such as AERMOD cannot account for photochemical transformation of contaminants, the hybrid approach is expected to be more appropriate for less reactive (e.g. benzene) rather than highly reactive (e.g. formaldehyde) components of the urban air pollution mix.

5. AIR QUALITY MODELING NEEDS: MICROENVIRONMENTAL SCALE

In the context of modeling, exposure occurs in "microenvironments" - the outdoor, indoor, or in-vehicle locations that individuals spend time in - where the "ambient" atmospheric concentration value (monitored or modeled) is modified by the "immediate surroundings" of the individual person/receptor (e.g., residential indoor: different rooms in a house; occupational indoor: different types of workplaces; public indoor: schools, restaurants, theaters, shopping malls; vehicular: car, bus, train; outdoor: urban street canyons, suburban streets, roadway intersections, gas stations, parks and recreation areas, schoolyards, etc.)

Characterizing microenvironments can involve modeling of various local sources and sinks and transport/fate processes, and interrelationships between ambient and microenvironmental concentration levels. Three general approaches have been used in the past to model microenvironmental concentrations: empirical (typically linear regression) fitting of data; parameterized mass balance modeling; and detailed Computational Fluid Dynamics (CFD) modeling.

Characterization of outdoor microenvironments

Empirical regression analyses have been used in various studies to relate specific outdoor "locales" defined by land use (that can be interpreted as generalized types of exposure microenvironments) to spatial variability of pollutant concentrations. For example, some studies[36] use regression analysis to assess the associations between airborne concentrations and land-use variables such as the area of open space, traffic count on nearest highway, the length of highways and major roads within a certain radius, population density, industrial land-use, the length of minor roads, distance from the nearest highway, etc. However, various arguments have been made regarding the value of using land-use regression modeling to assign exposure classifications in large-scale epidemiologic studies.

Parameterized mass balance models for outdoor microenvironments include various local roadway, intersection, and street canyon models[37]. Near-highway pollutant dispersion models consider vehicle wake parameterizations derived from canopy flow theory and wind tunnel measurements to adjust the atmospheric velocity and turbulence fields. In parameterized street canyon models, typically, concentrations of exhaust gases are calculated using a combination of a plume model for the direct contribution and a box model for the recirculating part of the pollutants in the street. Parameterization of flow and dispersion conditions in these models is usually deduced from analysis of experimental data and model tests that considered different street configurations and various meteorological conditions.

Various CFD-based street canyon models have also been developed in recent years (see, e.g., the series of International Conferences on Harmonization - http://www.harmo.org), employing various alternatives for local closure of the turbulent transport equations. Reviews and intercomparisons of such models *vis-a-vis* field data are available[38].

Characterization of indoor microenvironments

Numerous indoor air quality modeling studies have been reported in the literature; however, depending on the modeling scenario, only few of them address - typically a limited subset of - physical and chemical processes that affect complex air pollution mixtures (e.g. photochemical oxidants) indoors[39-42]. It is beyond the scope of the present discussion to review in detail the current status of indoor air modeling. Existing indoor air concentration models indeed are available as a wide range of (a) empirical regression relationships, (b) parameterized mass balance models (that can be either "single-zone" - that is, single well-mixed room - or "multi-zone" models), and (c) CFD-based models. Various studies have compared the different formulations of zonal models and of more complex, CFD, models[43].

Some indoor air models have considered atmospheric chemistry, that can be especially important in the presence of indoor sources such as gas stoves, etc.[44], while others considered potential limitations of uniform mixing assumptions[45]. These can be important issues when calculating personal exposures and need to be addressed in conjunction with improving indoor emission inventories.

6. SUMMARY

Though existing inhalation exposure modeling systems have evolved considerably in recent years, the air quality models that provide the inputs to these systems can be substantially

improved from the perspective of providing *exposure-relevant* estimates of air quality metrics. Deriving from the discussion in the previous sections, various specific and evolving needs are summarized here:

Ambient photochemical modeling systems are not currently optimized for estimating pollutants at the "neighborhood" scale. Therefore, practical methods are needed for "downscaling" regional/urban modeling estimates to neighborhood and microenvironmental scales, with an emphasis on consistency in linking and coupling models at different scales.

Microenvironmental modeling efforts need to balance mechanistic detail and usability by developing "simplified but adequate" models that take into account mixing, "local" (indoor or outdoor) chemistry, etc. These models can be developed either directly or as simplifications of detailed Computational Fluid Dynamics methods. For population exposure assessment, there is a need for computationally efficient methods for modeling air quality dynamics in representative realistic distributions of (outdoor and indoor) microenvironments, in ways that allow aggregation and statistical extrapolations of results across the range of such microenvironments within a regional/urban model cell. These approaches should utilize high-resolution information in urban and suburban topography, combined with detailed "microinventories" of local emission sources.

Comprehensive air quality models such as CMAQ can be enhanced through incorporation of, or linking with additional modules for dynamic soil, water, and other compartments. In the future, exposure assessments can be substantially improved through the development and application of comprehensive multimedia models that address multiple scales ranging from local to regional to global and can be coupled with multi-pathway human exposure models.

Also, in light of the synergistic effects of co-occuring pollutants, there is a need for expanding the range of airborne contaminants included in the "one-atmosphere" approach to include allergens and other biological agents (see Figure 5).

Modeling frameworks for exposure assessment in the past have typically focused on individual contaminants and on subsets of their pathways/routes and sources of exposure, potentially neglecting significant contributions from remaining pathways/routes or sources. However, in recent years, the focus of environmental human and ecological health risk analyses, pursued by both the research community and regulatory agencies, has been gradually shifting from considerations of single to multiple contaminants and pathways; in the future, integrative analyses that link environmental, behavioral and biological considerations[2] will allow increased accountability and more realistic and accurate risk assessments.

7. ABOUT THE AUTHORS

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Table 1: Comparison of typical features/attributes of comprehensive inhalation exposure models

	pNEM	HAPEM	REHEX	APEX	SHEDS	MENTOR
Typical Spatial Extent/Resolution	Urban areas; Census tract	Urban/Regional; Census tract	Urban; Census tract	Urban areas; Census tract	Urban areas; Census tract	Urban areas; Census tract
Typical Temporal Extent/Resolution	Year: Hour	Season; 3 Hours	Season; Hour	Weeks to Year; Hour/Minutes	Weeks to Year; Hour/Minutes	Weeks to Year; Hour/Minutes
Population Activity Pattern Simulation	Top-down analysis	Top-down analysis	Top-down analysis	Bottom-up analysis	Bottom-up analysis	Bottom-up "person-oriented"
Estimation of Concentrations in Microenvironments (μE's)	Mass-balance	Empirical I/O (input/output) relationships (eIOR)	Mass balance (steady-state only); dynamically generated μE's	Mass balance and eIOR; fixed μE's	Mass balance (steady-state only), and eIOR; dynamically generated μE's	Mass balance, eIOR and indoor chemistry; dynamically generated μE's
Indoor Source Emissions & Micro-environmental factors	Limited sources; Probability distributions	Additive terms for sources; Probability distributions	Additive terms for sources; Probability distributions	User-defined sources; Probability distributions	Limited sources; Probability distributions	User-defined sources; Probability distributions
Uptake and Doses	Potential doses	NA	NA	Potential doses and uptake	Potential doses and uptake	Physiologically based doses
Exposure Characterization; Resolution	Population distributions; Hourly	Limited population distributions; Annual	Population distributions; Hourly	Population distributions; Hourly/Minutes	Population distributions; Hourly/Minutes	Population distributions; Hourly/Minutes
Notes	Cohort-based model; precursor to APEX	Has been employed in developing NATA estimates	Proprietary implementation	Follows "One Atmosphere" approach for mixtures of chemicals	Different implementations for individual groups of chemicals	Follows "One Atmosphere" approach for mixtures of chemicals

Table 2: Essential attributes of various air quality models, including Eulerian grid-based and Lagrangian/Gaussian dispersion models

	AERMOD	HYSPLIT	Calpuff	HYPACT	HPAC	CAMx	CMAQ
Developer	USEPA	NOAA-ARL	EarthTech	Alliant Inc	DTRA	Environ	USEPA
Model Type and Solution Scheme	Gaussian plume; Analytical	Lagrangian puff/particle dispersion; Discrete	Lagrangian puff dispersion; Discrete	Lagrangian particle dispersion; Discrete	Lagrangian puff dispersion; Discrete	Eulerian grid; 3D Finite Difference	Eulerian grid; 3D Finite Difference
Chemical Species/ Contaminants	Individual chemicals	Individual chemicals†	Individual chemicals†	Individual chemicals	Individual chemicals†	Mixtures of chemicals	Mixtures of chemicals
Chemical Interactions	Linear decay of single chemical	Simplified CB4 chemical mechanism	Simple chemistry modules	NA	Simple chemistry modules	Complex mechanisms (CB4, RADM, SAPRC)	Complex mechanisms (CB4, RADM, SAPRC)
Scenarios where Model is most Applicable	Longer-term averages; local scale	Multiple spatial scales with relatively few sources and receptors	Urban to regional scales; time scales of hours/days	High resolution spatial scales; computationally demanding at large spatial scales	Very high resolution spatial and temporal scales; Acute exposures	Urban/regional scales; hourly temporal resolution; secondary pollutants	Urban/regional scales; hourly temporal resolution; secondary pollutants
Use of Model Outputs as Inputs for Exposure Modeling	Hourly-annual assessments at local scale for non-reactive chemicals	Screening level analysis with few sources and receptors	Short- to intermediate term exposures	Dynamic, short-term exposure assessments	Dynamic, short-term exposure assessments	Hours-months environmental exposures to multiple chemicals	Hours-months environmental exposures to multiple chemicals

† custom modules exist for simplified chemistry involving a limited set of chemicals; Terminology for spatial scales: local (0-10 km), urban (10 km to 100 km), and regional (> 100 km); Acronyms: CB4, Carbon Bond IV; RADM, Regional Acid Deposition Model; SAPRC: Statewide Air Pollution Research Center chemical mechanism.

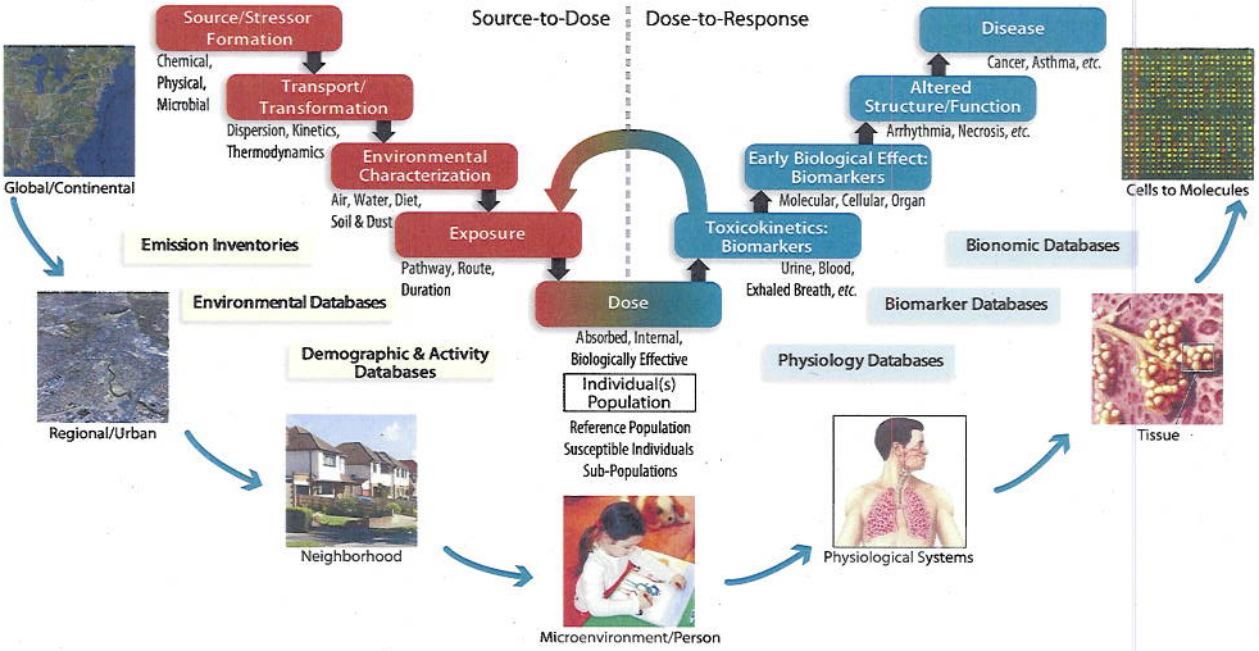

Figure 1: An overview of the sequence of processes and spatial scales involved in the source-to-dose-to-outcome sequence for human health risks due to inhalation exposures.

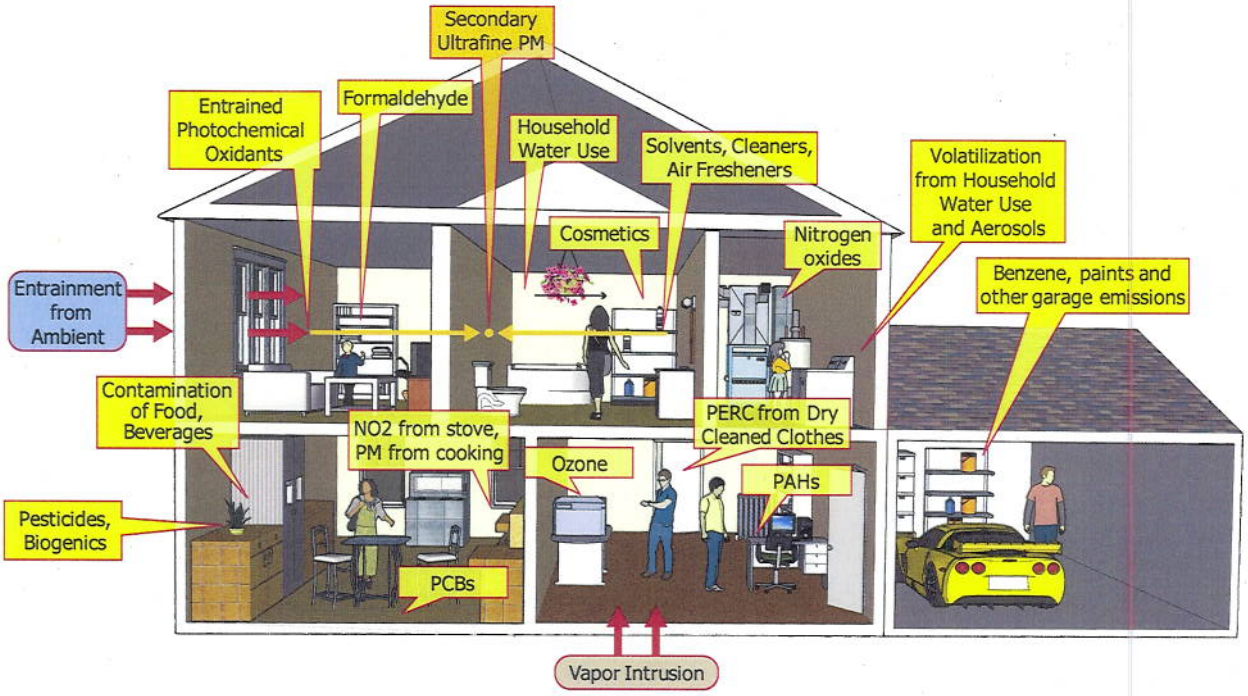

Figure 2: For most people the majority of exposures to airborne contaminants takes place through contact and inhalation of chemicals in indoor (residential or occupational) microenvironments. The air in these microenvironments contains a complex mixture of contaminants including those entrained from outdoor (ambient) air, those emitted indoors, and those formed via chemical transformations in indoor air (e.g. ultrafine particles formed from the interaction of entrained ozone with emissions from household air fresheners and solvents).

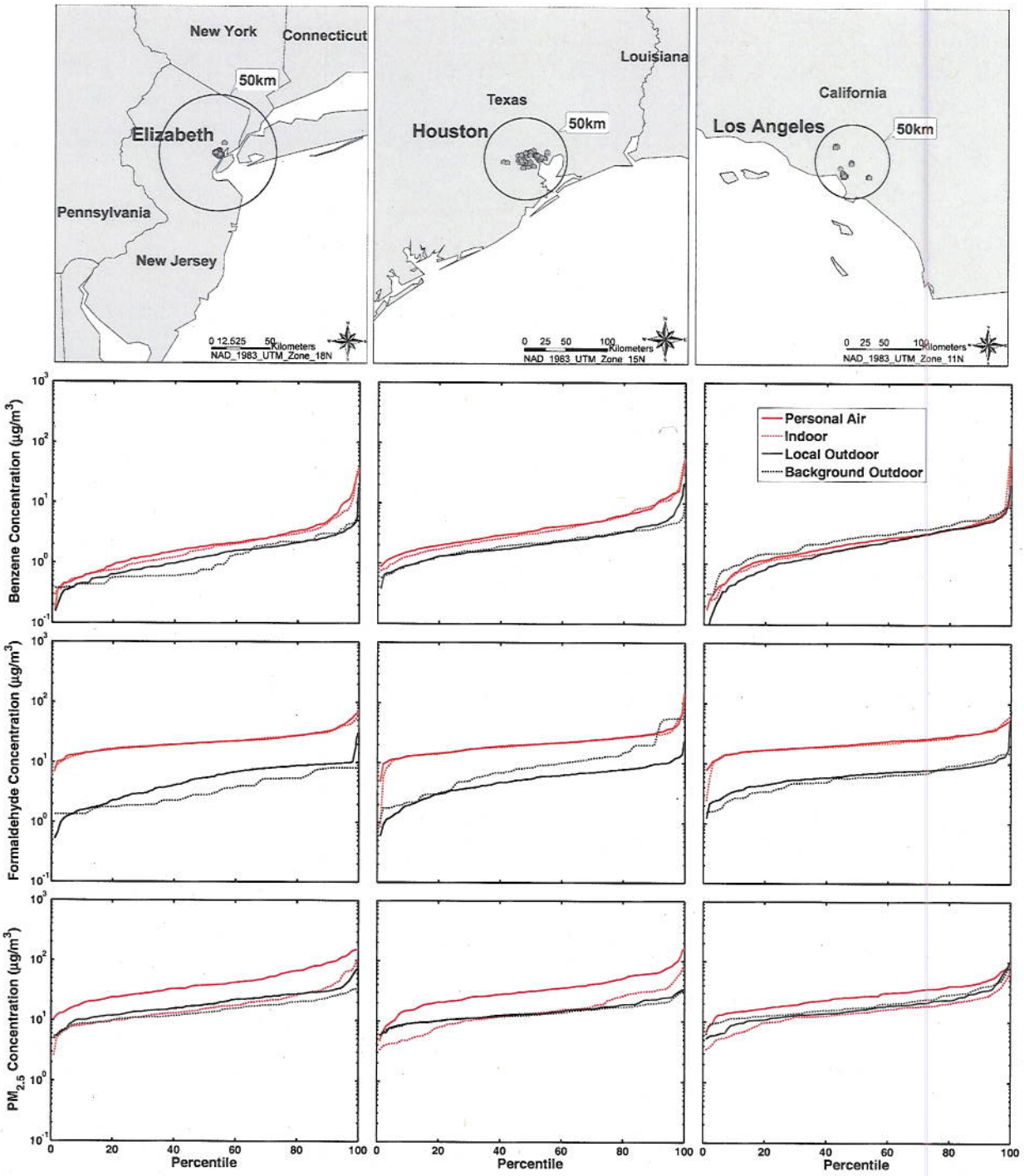

Figure 3: Personal, local outdoor, and indoor levels of three common air pollutants across diverse geographical areas. Shown are distributions of 48-hour integrated indoor, local outdoor, background ourdoor (concentrations from the nearest ambient air quality monitor) and personal air samples of target compounds collected simultaneously from approximately 100 homes of non-smokers (and with no attached garages), each in Elizabeth, NJ, Houston, TX, and Los Angeles, CA between 1999 and 2001 (data from [46]). The three contaminants shown are benzene (representing a non-reactive gas), formaldehyde (representing a highly reactive gas that is both emitted and formed through atmospheric photochemistry) and $PM_{2.5}$.

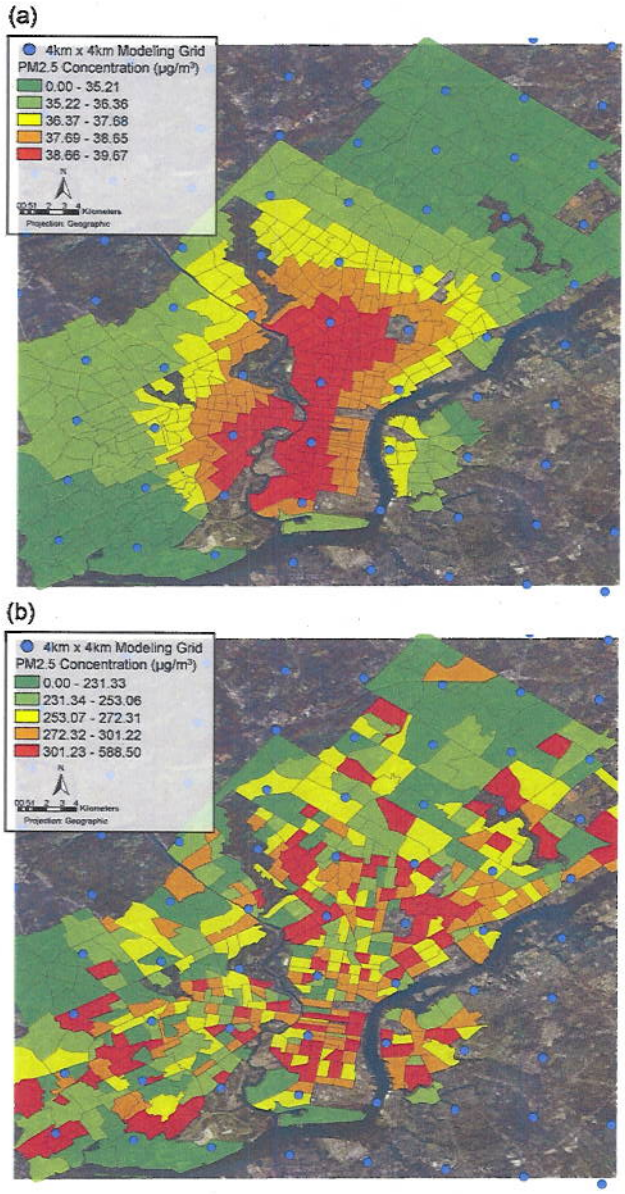

Figure 4: (a) 24-h averaged local outdoor concentrations on July 19, 1999 for 482 urban Philadelphia census tracts, derived from hourly $PM_{2.5}$ predictions of the Community Multiscale Air Quality (CMAQ) model that were "downscaled" at census tract level, using the Bayesian Maximum Entropy (BME) method; (b) Corresponding 95th percentiles, calculated for each census tract, of 24-h aggregated total inhalation doses from both outdoor and indoor sources. The color scheme shows quantiles of the concentrations and dose distributions (source: [2]).

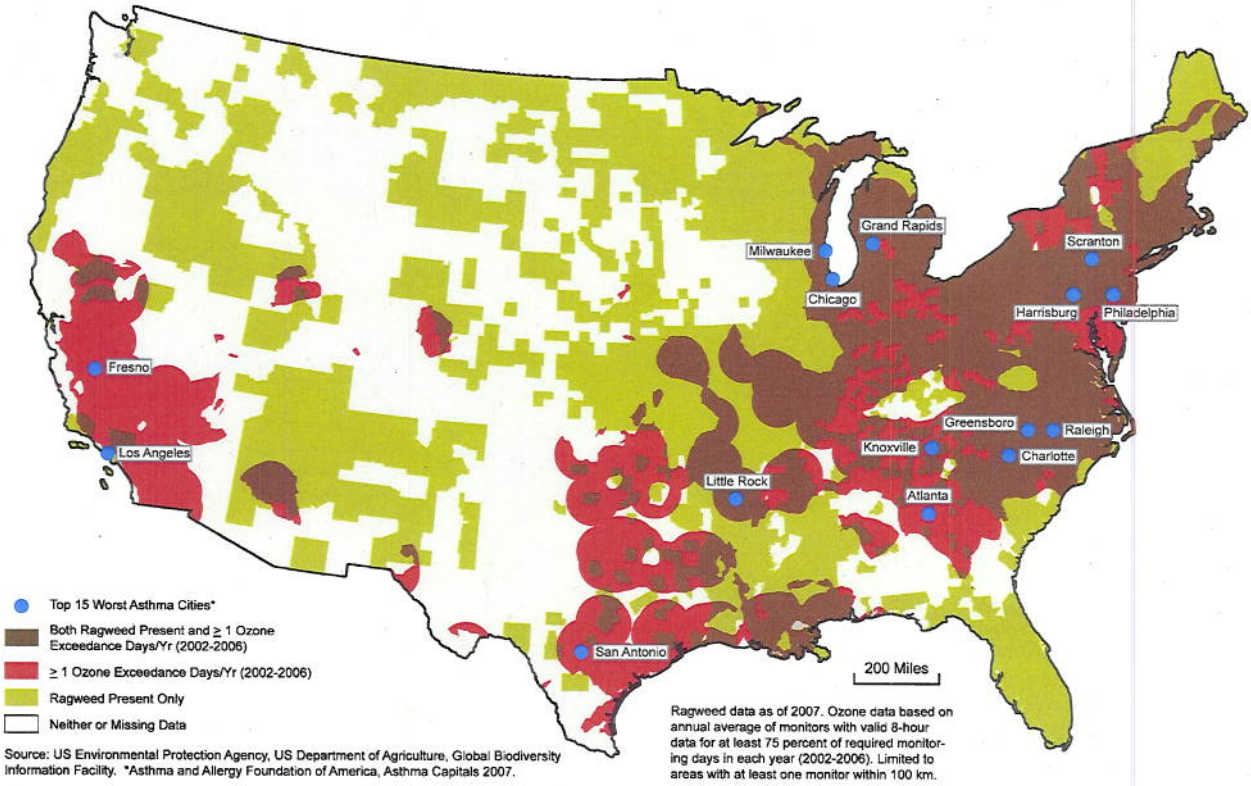

Figure 5: Co-occurrence of high ragweed pollen levels and high ozone levels across the continental US; co-exposures to pollen and ozone can have synergistic adverse health effects. Unified modeling frameworks are needed to study co-exposures to gaseous, particulate phase pollutants, and bioaerosols, in a consistent manner. Source: NRDC[47] based on ozone monitoring data from USEPA, USDA, and AAFA.