

Considerations in the use of ozone and PM_{2.5} data for exposure assessment

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Abstract The US national ambient-air monitoring network, created to verify compliance with health-based standards, now doubles as an important source of exposure data for the epidemiological analyses on which these standards increasingly rest, particularly in the case of ozone and PM_{2.5}. This paper was written for a workshop called to facilitate and inform the use of routine ozone and PM_{2.5} data by the Environmental Public Health Tracking Network. It examines the fit between priorities that shape regulatory monitoring and modeling and the data needs of public health tracking.

Keywords Ozone · PM_{2.5} · Ambient air quality standard · Monitor · Measurement · Model

Introduction

The 1963 Clean Air Act and its subsequent amendments identify “criteria pollutants” for which the US Environmental Protection Agency (EPA) is required to establish National Ambient Air Quality Standards (NAAQS) to protect human health with adequate margins of safety. Setting standards in terms of community air concentrations rather than source emission rates is a key feature of the statutory framework in which US air quality is managed (Seinfeld 1975, pp 27–32). This focus has led EPA and its local regulatory partners to develop a substantial national infrastructure of ambient air monitoring to assess communities’ compliance and atmospheric modeling to guide their emissions-reduction strategies. The national ambient air monitoring network, created to

verify compliance with health-based regulatory standards, yields also a consistent and extensive record of air quality that is readily available for atmospheric research and other uses. It is an important source of exposure data for the epidemiological analyses that inform the NAAQS, bringing analytical closure to an ongoing cycle of air quality management (USEPA 2006b; USEPA 2008).

In 2002, the Centers for Disease Control and Prevention (CDC) initiated a program of Environmental Public Health Tracking (EPHT) seeking to promote more systematic linking of information on environmental hazards, exposures, and health effects (McGeehin et al. 2004). Ozone and PM_{2.5} are the air pollutants selected for initial study because air monitoring data are available, high levels of these contaminants are the main cause of poor air quality in much of the country, and they have been strongly linked with respiratory and cardiovascular health effects (CDC 2004). CDC joined EPA and the Health Effects Institute in sponsoring a January 2008 workshop in Baltimore titled Methodologies for Environmental Public Health Tracking of Air Pollution Effects. The workshop brought participants in the EPHT program together with US and international experts in biostatistics and air pollution epidemiology, as described elsewhere in this issue.

This paper was originally prepared as a background document for the Baltimore workshop. It focuses on the spatial and temporal compatibility issues that motivate much of the early work on the air component of EPHT (Boothe et al. 2005, 2006), examining three areas where the priorities that shape regulatory monitoring and modeling for NAAQS compliance may not align exactly with those for public health tracking:

1. High concentrations of ozone and fine particle mass are indicators of unhealthful air, but these two parameters

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do not fully characterize the air's composition and toxicity. An exclusive focus on improving the spatio-temporal resolution of ozone and $PM_{2.5}$ data may not yield proportionate benefits for tracking public health effects.

- Compliance determinations typically place more weight than do epidemiologic analyses on a measurement's absolute accuracy. EPHT tools could usefully exploit some of the abundant additional air data that EPA collects or produces for other purposes.
- Standard regulatory models of the atmosphere are optimized for predicting the impacts of known emissions from controllable major sources. They offer less reliable representations of the myriad emissions from smaller, less well-characterized sources that contribute to a population's total exposure.

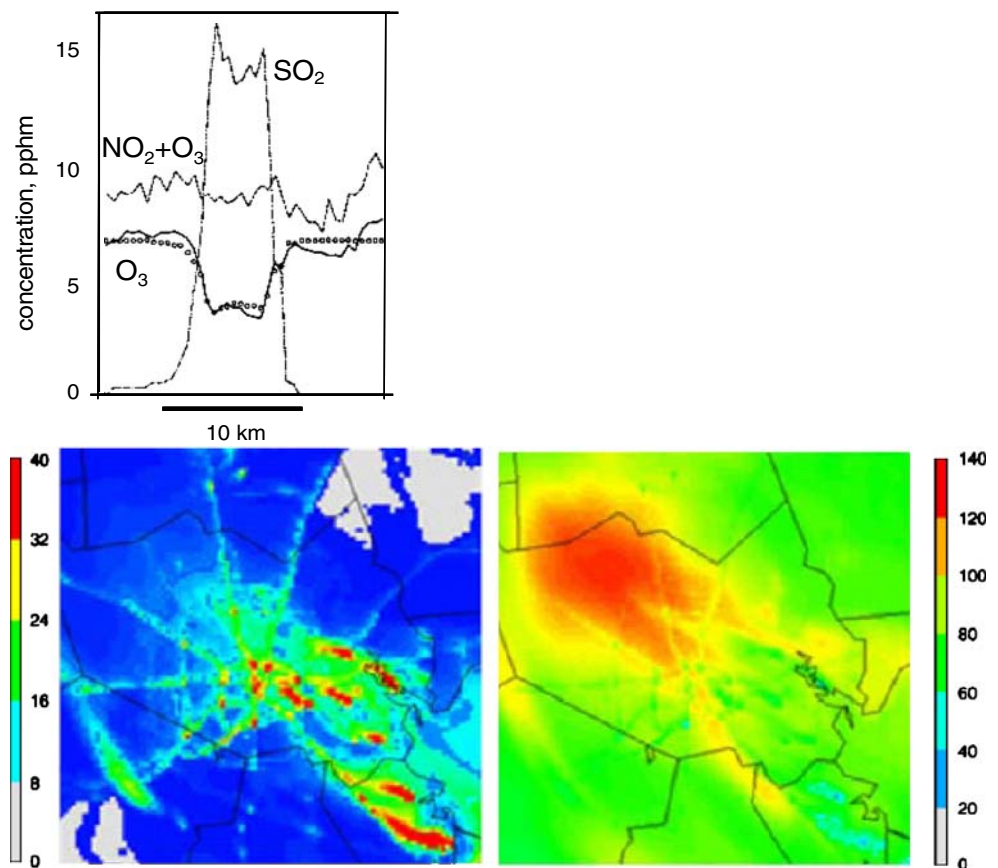
Parameterization of "air quality"

Community air is a complex and dynamic mix of gases and suspended particles in constant interaction; it would not be feasible for a routine national network to monitor all of its constituents. While carbon monoxide and particulate lead have NAAQS specific to their health effects, ozone and $PM_{2.5}$ are monitored for NAAQS that address effects from

mixtures of pollutants associated chemically or physically with their eponymous indicators (Arnold et al. 2007). Ozone is a unique species, but the ozone concentration serves as the indicator for a reactive mix of ozone and other photochemical oxidants. Fine particulate matter is a heterogeneous mixture, and its total mass concentration $PM_{2.5}$ serves as an indicator that a recent review (NRC 2004, p 9) comments "greatly oversimplifies complex biological phenomena" addressed by its NAAQS.

Ozone is one of many oxidants participating in a system of photochemical reactions that involves nitrogen oxides, hydrocarbons, and free radical intermediates. During daylight hours, these reactions commonly yield a gradual accumulation of total oxidants to which ozone is a major contributor. The proportion of ozone to other oxidants can vary greatly from one air parcel to another, as the fast reaction $O_3 + NO \rightarrow O_2 + NO_2$ of ozone with nitric oxide effectively replaces ozone with another oxidant, nitrogen dioxide. NO is the predominant nitrogen oxide produced by high-temperature combustion, so the near-source effect of combustion emissions is to depress ozone concentrations (Fig. 1). Monitoring strategies and exposure metrics for ozone are designed to minimize this masking by fresh emissions: the ozone NAAQS thus address the maximum concentrations recorded anywhere in an area by monitors set well back from roadways.

Fig. 1 Right observed ozone deficit in the sulfur dioxide plume downwind of a power plant (adapted from White 1977). Regional background ozone is depleted within the plume by coemitted nitric oxide. Circles show ozone profile from a simple photostationary model. Below simulated NO_x (left) and O_3 (right) concentration fields in Houston from a CMAQ model run at 1 km grid resolution (adapted from Ching et al. 2006). Arterial roads show NO_x excesses and O_3 deficits, as do source regions to the southeast



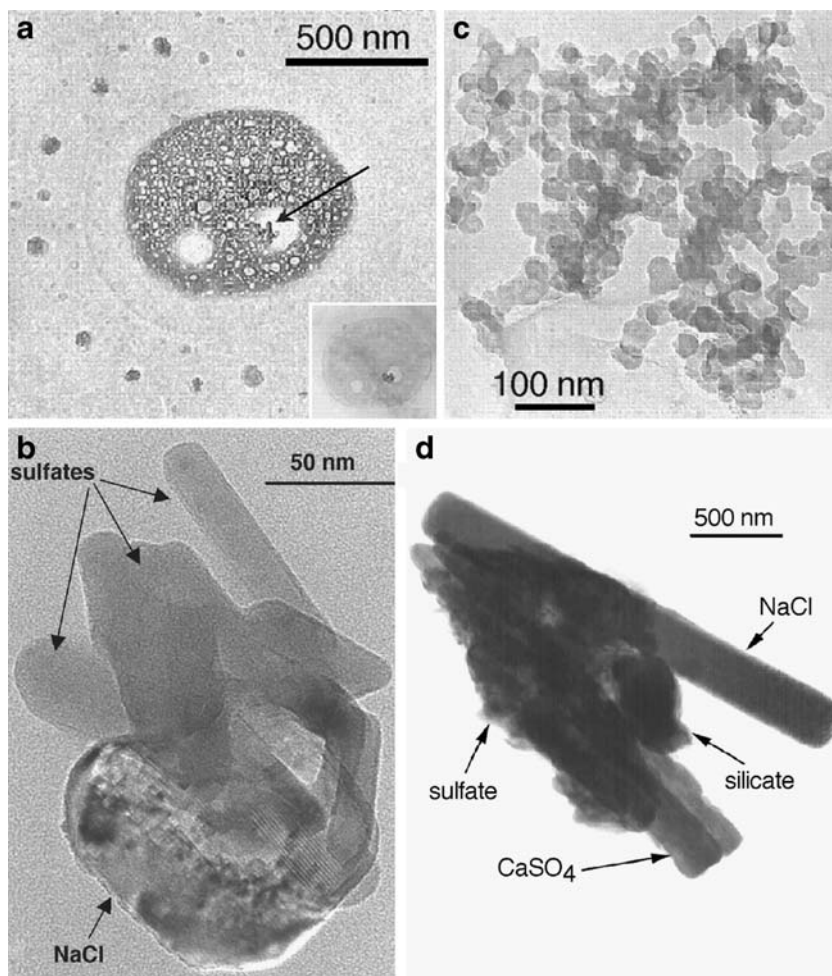
PM_{2.5} is defined operationally as the mass of particulate matter collected on specified filter media under specified conditions behind a specified inlet designed to exclude particles greater than about 2.5 μm in aerodynamic diameter. The collected material is a heterogeneous agglomeration of solid and liquid particles, some of them semivolatile, that when airborne were of diverse size, shape, and chemical composition (Fig. 2). It is useful to distinguish between primary particles, which are emitted directly to the atmosphere, and the secondary condensates that atmospheric reactions produce from gaseous emissions. Whereas primary PM_{2.5} concentrations rapidly fall as emissions mix with cleaner background air, secondary PM_{2.5} concentrations may increase for some distance downwind. The primary and secondary components of PM_{2.5} are thus differently distributed even when both arise from the same source of emissions.

A limitation of ozone and PM_{2.5} as a parameterization of air quality is that they do not represent the same oxidant and particulate mixes near a source as they do farther away. They do not, for example, capture the localized gradients in primary emissions from cars and trucks thought to account

for chronic health risks found near roadways (e.g., Hoek et al. 2002). As noted above, highways are marked by ozone deficits even in oxidant-rich air. The combustion nuclei in vehicle exhaust, whatever their possibly disproportionate impact on health, generally represent a minor fraction of total PM_{2.5} mass (e.g., Roosbroeck et al. 2008). Air quality maps limited to ozone and PM_{2.5} will thus fail to fully characterize traffic-related air pollution regardless of their spatial resolution. Similar considerations apply to industrial or municipal point sources that may create localized “hot spots” for especially toxic trace species.

Increasing the number of species measured at a central monitoring station—adding species that are peculiar to particular source types, for example—can provide useful insights (Laden et al. 2000). The added value of such information for time series analyses can be limited, however, by the substantial temporal covariance typically observed among all species (e.g., Burnett et al. 2000, Table 17). Figure 3 sketches a general mechanism by which the localized health effects of specific primary emissions can be misattributed to more broadly distributed indicators such as PM_{2.5} or ozone, even when the toxic species is

Fig. 2 Electron micrographs of example atmospheric particles (NRC 2004; adapted from Buseck and Posfai 1999). **a** Internal mixture of sulfate and soot; *arrow* points to a soot aggregate. The surrounding halo is ammonium sulfate crystals formed as the sulfate dehydrated in the microscope’s vacuum. **b** Sea salt. **c** Branching soot aggregate typical of combustion processes. **d** Internal mixture of terrestrial silicate with sea salt and anhydrite (calcium sulfate) likely formed by the reaction of sulfur dioxide with carbonate particles



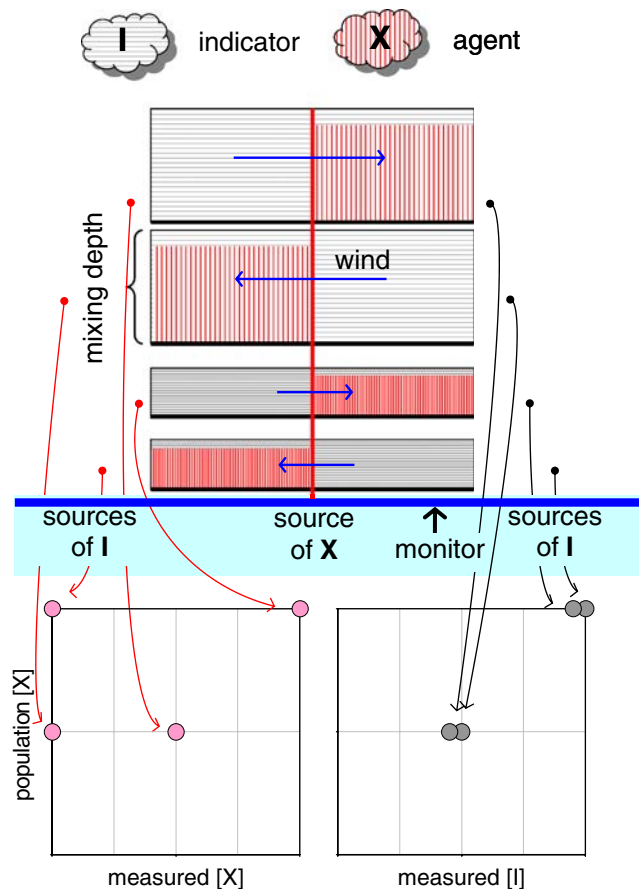


Fig. 3 Pollution climate of Line City. Blue bar maps the linear arrangement of emissions sources and monitor. Rectangles above the bar show x - z distributions of atmospheric concentrations under four different meteorological regimes. Curved arrows link individual meteorological regimes to individual points in graphs below the bar. These graphs plot city-average concentrations of the harmful agent X against monitored concentrations of X and the generic air quality indicator I

itself monitored. The idealized Line City is a collection of pollutant sources and two neighborhoods arrayed along an east–west axis. The two neighborhoods are bracketed by sources of the broadly distributed indicator species I . The

neighborhoods themselves bracket the sole source of X , the primary pollutant actually affecting health. Each source generates a plume of effluent to the east or west depending on wind direction. Line City winds blow from the east on half of the days and from the west on the others. Both neighborhoods receive I emissions every day, but this I is mixed with unhealthy X in only one neighborhood at a time. Wind speed and mixing depth combine each day, independently of wind direction, to yield either good or poor synoptic ventilation. Only one of the two neighborhoods has air quality monitors. Because the I monitor always sees the effects of ventilation, even when the X monitor has nothing to measure, the I measurements give a better indication of overall community exposure to X than do the available measurements of X itself.

Availability of measurements

Ozone and $PM_{2.5}$ are routinely monitored for compliance with their NAAQS by the Federal Reference Method (FRM) or a Federal Equivalent Method (FEM). (EPA specifies a reference method for measurement when it promulgates a standard; it designates other methods as equivalent if they demonstrate agreement with the reference method.) States, tribes, and local agencies establish and operate compliance networks following specific EPA guidelines for siting, instrumentation, and quality assurance. The resulting data are submitted to EPA's Air Quality System (AQS) database by the end of the quarter following the quarter of their collection. AQS is an attractive source of uniform, timely, and quality-assured air monitoring data.

Figure 4 compares the volumes of FRM/FEM data available for ozone and $PM_{2.5}$. FRM/FEM measurements are made daily at far more locations for ozone than for $PM_{2.5}$. Ozone was measured at this frequency at over 550 sites throughout 2006 and at over 1,100 sites during peak ozone months, while daily $PM_{2.5}$ measurements were made

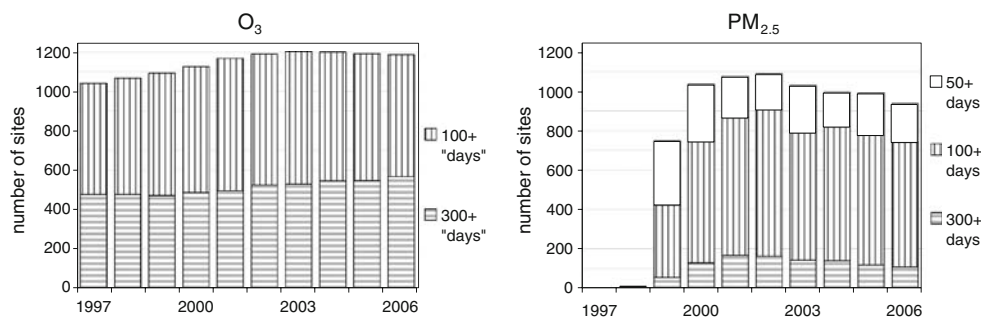


Fig. 4 Trends in monitoring by the FRM or the FEM. The FRM and FEM monitors for ozone report continuously, year-round in some locations and during selected warm months at others. Measured “days” for ozone are plotted as reported hours/24. FRM and FEM monitors for

$PM_{2.5}$ collect 24-h samples year-round, daily or every third or sixth day. At sites with multiple monitors, only the one reporting the most observations was counted. Data were downloaded December 2007 from AirData, http://www.epa.gov/aqspub1/annual_summary.html

at fewer than 110 sites. Most FRM/FEM measurements for $PM_{2.5}$ are made once every 3 or 6 days.

The different frequencies of the FRM/FEM data for ozone and $PM_{2.5}$ reflect the networks' design for compliance monitoring rather than time series analyses of acute health effects. The health-based ozone NAAQS has always targeted extreme values, specifying an 8-h (formerly 1 h) concentration not to be exceeded more than a handful of days each year. Verifying compliance with a standard of this form requires continuous monitoring, at least during seasons with the potential for high concentrations. In contrast, the new $PM_{2.5}$ NAAQS introduced in 1997 included a limit on the annual mean as well as one on extreme 24-h concentrations. The annual standard was generally controlling, in the sense that it was hard to violate the 24-h standard without also exceeding the annual mean. Compliance monitoring for $PM_{2.5}$, therefore, focused on the annual mean, which usually could be estimated from measurements on a representative sample of days. The EPA significantly tightened the 24-h standard in December 2006 and supported this change by moving to daily sampling at about 50 monitoring sites previously sampling 1 day in three (USEPA 2006a).

Non-FRM/FEM data for $PM_{2.5}$ are available on the every-third-day schedule of most FRM/FEM monitors from two networks that monitor particle speciation (VIEWS 2007). The Interagency Monitoring of Protected Visual Environments (IMPROVE) operated about 160 sites in 2006 at predominantly rural or remote locations. The Chemical Speciation Network/Speciation Trends Network (CSN/STN) operated about 60 population-oriented sites every third day and about 125 more every sixth day. These networks weigh 24-h samples on Teflon filters as the FRM does, but use samplers with inlets and flow rates different from FRM specifications.

Daily $PM_{2.5}$ measurements are made at many more locations by continuous monitors, in support of EPA's AIRNow public-reporting program. About 580 sites throughout the US supply hourly data (Chan 2007, personal communication) that are reduced to broad ranges for real-time display on a national map (<http://www.airnow.gov>). Various measurement methods are used (Hanley 2006), including nephelometry, beta attenuation (BAM), and oscillating microbalance (TEOM). These real-time data are qualified as "not fully validated ... [and] only approved for the expressed purpose of reporting and forecasting the Air Quality."

EPA gave its first FEM designation to a continuous $PM_{2.5}$ monitor shortly after the Baltimore workshop, and substantial growth in the availability of continuous FEM data can be expected in the near future. The FRM/FEM designation is important for compliance monitoring because particle measurements are sensitive to methodological details. Some ambient particles are in equilibrium with

surrounding gases, an equilibrium that can shift after they are sampled onto an FRM/FEM filter through which air is drawn for 24 h. Because the $PM_{2.5}$ NAAQS is set in terms of the FRM, a method that avoided such sampling artifacts (or exhibited different ones) would not be suited to monitoring NAAQS compliance. Additionally, nongravimetric methods require calibration factors that can vary with particle composition and ambient humidity. It is clearly undesirable to have site-specific calibrations influence compliance determinations that span diverse climates and regulatory jurisdictions.

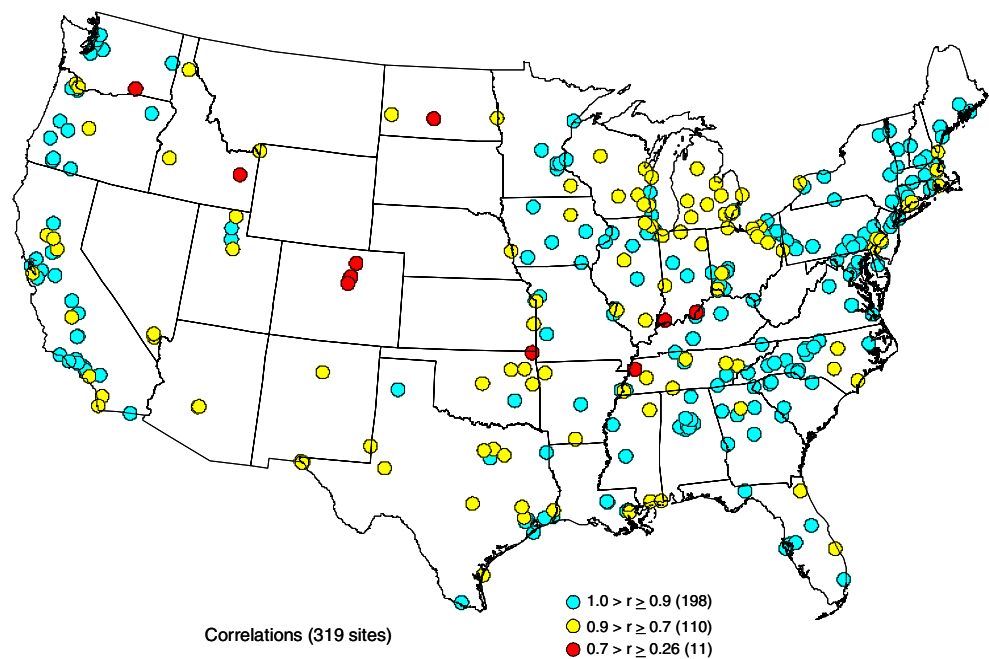
Data quality objectives for epidemiological analyses differ from those for compliance monitoring in ways that are more welcoming of AIRNow data. Compliance determinations address whether or not measured concentrations exceed a specified limit; avoiding errors requires measurements that are especially accurate at concentrations near that limit. Epidemiological analyses examine differences rather than absolute concentrations and require only the correlation of a measurement with the variable of interest. Mintz and Schmidt (2007) report an overall correlation of $r^2=0.77$ between the 24-h AQS and AIRNow $PM_{2.5}$ concentrations in over 110,000 paired observations during 2004–2006 (Fig. 5).

Availability of models

The relationship of inhaled air to monitored air is embedded in any observed statistical association between community health and air quality. As fixed monitors collect only discrete samples from the continuous atmosphere to which people are actually exposed, this relationship must generally be modeled. City-scale studies have commonly modeled general populations' individual exposures as Berksonian departures from a city-wide air quality that is estimated by averaging all local measurements in each monitoring period (e.g., Samet et al. 2000). Such simple models are consistent with the interpretation, sketched earlier, of ozone and $PM_{2.5}$ as generic indicators for broader chemical mixes.

Some metropolitan-scale and regional-scale studies have used contouring, land-use, or atmospheric models to relate measured concentrations to individual exposures. EPA's current operational-level understanding of emissions and transport factors is incorporated in its Community Multi-scale Air Quality (CMAQ) grid model, which has been used since 2004 to produce real-time national forecasts of hourly ozone concentrations (NWS 2007). CMAQ's $PM_{2.5}$ routines are much younger than its ozone routines, which trace their ancestry back through generations of critical scrutiny (e.g., NRC 1991). A recent report (USEPA 2005) found its performance in the western US to be significantly poorer for all $PM_{2.5}$ species than in the eastern US, which

Fig. 5 Correlation of 24-h-averaged PM_{2.5} AIRNow data with FRM/FEM measurements at sites with at least 30 observations during 2004–2006 (adapted from Mintz and Schmidt 2007). AIRNow has since grown to about 580 sites



had been the early focus of evaluations. Users can expect CMAQ's PM_{2.5} performance to evolve and improve as it undergoes more cycles of review and development.

CMAQ's primary function has been to support the evaluation of alternative strategies for managing emissions to attain ambient standards. It is accordingly source-oriented, taking emissions and winds as known and predicting the ambient concentrations that result under various regulatory scenarios. One limitation of any source-oriented PM_{2.5} model is that elevated concentrations in the real world can result from sporadic and hard-to-characterize fugitive emissions. Unlike CO and SO₂, which emerge predictably from tailpipes and stacks through which their fluxes can be measured and documented, episodes of dust and smoke typically reflect agricultural and construction activities, wildfires, and other erratic and diffuse sources. Even concentrations measured near such sources are not reliably convertible to the mass fluxes needed as model inputs. Evaluations of CMAQ repeatedly show the agreement of its predictions with measured 24-h concentrations to be better for sulfate than for any other pollutant (Mebust et al. 2003; USEPA 2005). It is no coincidence that our knowledge of emissions is also better for SO₂, the primary precursor to sulfate, than it is for any other air pollutant. Because well-determined species can "steal" explanatory significance from poorly determined covariates in multivariable regression (White 1998), differential uncertainties in emissions can distort the modeled associations of individual species or source contributions with health effects.

CMAQ and the monitoring networks exhibit somewhat complementary strengths and weaknesses as sources of air data. Point measurements represent "true" concentrations, as

defined by regulations and the epidemiological findings that motivate them. However, they have no necessary relationship to one another; in sparsely monitored areas, they leave uncertain the boundaries between clean and dirty air. In contrast, CMAQ yields a logically coherent grid of concentrations that reflects our understanding of emissions and atmospheric processes. These concentrations can be unrepresentative of reality, however, when they are derived from inaccurate descriptions of emissions and the atmosphere.

Model outputs would ideally be reconciled with available measurements by adjusting uncertain model inputs. Prior probability distributions would be assigned to the intensity and geographical distribution of emissions, to wind fields, and to empirical parameterizations of atmospheric transformations and then revised in light of observed concentrations. The iterations required for a fully Bayesian solution would be impractical with the massive CMAQ code, although initial steps in this direction have been taken with simpler models (Husar et al. 1986; Schichtel et al. 2006). A more tractable but still computationally demanding approach to assimilating observations with CMAQ is the hierarchical Bayesian (HB) fusion model described by McMillan et al. (2009, submitted for publication) and explored in early EPHT efforts (Boothe et al. 2005, 2006). Daily ozone and PM_{2.5} estimates from this statistical hybrid are currently available from 2001 to 2005 for 12- and 36-km grids over the contiguous United States (USEPA 2009).

McMillan et al. (2009, submitted for publication) used PM_{2.5} concentrations in the eastern US during 2001 for their initial development and evaluation of the HB fusion model. In 2001, there were no data quality objectives for continuous PM_{2.5} monitors (USEPA 2002) and no collation

and mapping of continuous data by AIRNow. In this setting, the authors relied on FRM/FEM data for their observational inputs, reserving non-FEM measurements (from the IMPROVE and CSN/STN speciation networks) for cross-validation of the results. In 2004 and later years, the continuous PM_{2.5} data reviewed by Mintz and Schmidt (2007) provide much fuller observational coverage of the space–time grid than the predominantly 1-in-3-day FRM/FEM data offer, and any inequivalence between them can be accounted for within the Bayesian framework. Incorporating the continuous data seems a natural next step for tracking analyses to explore, particularly given EPA's announced intention to facilitate their substitution for filter-based FRM measurements in the future (USEPA 2006a).

Summary

The air component of EPHT seeks to link public health statistics and routine air monitoring data. Ozone and PM_{2.5} were selected for the initial study because they are the foci of current efforts to monitor and manage national air quality. The public health and air monitoring networks deliver their data in different formats with differing native spatial resolutions, and an early thrust of EPHT has accordingly been the investigation of methodologies for integrating the two data streams. This paper has surveyed some of the issues presented by reconciliation efforts, highlighting those which health researchers and air regulators may approach with unrecognized differences in their understandings and expectations. The following are among its observations:

- Spatial gradients in ozone and PM_{2.5} capture variations in the overall hazard from air pollution adequately at some scales and poorly at others, a consideration reflected in the guidelines by which air monitors are sited. Enhanced spatial resolution of these two indicators may, therefore, have limited value for health studies unless accompanied by the enhanced chemical resolution provided by measurement of additional species.
- The common temporal modulation of all species concentrations by synoptic (i.e., regional-scale) weather patterns may, conversely, limit the value of enhanced chemical resolution unaccompanied by enhanced spatial resolution to map the gradients in the mix.
- EPA generates various categories of ozone and PM_{2.5} data to support planning and public communication efforts as well as consequential regulatory actions. Data used for EPHT need not meet some of the more legalistic requirements placed on data used to determine compliance with regulatory standards.

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