Air pollution benefit-cost analyses depend on dispersion models to predict population exposures to pollutants, but it is difficult to determine the reasonableness of the model estimates. This is in part because validation with field measurements is not feasible for marginal concentration changes and because few models can capture the necessary spatial and temporal domains with adequate sophistication. In this study, we use the concept of an intake fraction (the fraction of a pollutant or its precursor emitted that is eventually inhaled) to provide insight about population exposures and model performance. We apply CALPUFF, a regional-scale dispersion model common in health benefits assessments, to seven power plants in northern Georgia, considering both direct emissions of fine particulate matter (PM$_{2.5}$) and secondarily formed ammonium sulfate and ammonium nitrate particles over a domain within 500 km of Atlanta. We estimate emission-weighted average intake fractions of 6 x $10^{-7}$ for primary PM$_{2.5}$, 2 x $10^{-7}$ for ammonium sulfate from SO$_2$, and 6 x $10^{-8}$ for ammonium nitrate from NO$_x$, with no effect of SO$_2$ on ammonium nitrate. To provide insight about model strengths and limitations, we compare our findings with those from a variety of atmospheric dispersion models to estimate changes in population exposure due to changes in source emissions. Currently available models vary considerably in their assumptions and sophistication regarding numerous key dimensions, including spatial and temporal resolution, spatial and temporal domain, characterization of emissions and background pollutant concentrations, atmospheric chemistry, meteorology, deposition, and other aspects of pollutant fate and transport. Because of computational and other resource and information constraints, no model is currently able to address all necessary dimensions simultaneously with ideal resolution, implying that models must be selected that best suit the analysis at hand.

Many recent air pollution benefits assessments (1, 2) have found that premature mortality from primary and secondary fine particulate matter (PM$_{2.5}$), as derived from long-term cohort epidemiological studies, dominates aggregate benefits. The associations reported by these cohort studies (3, 4) suggest generally linear concentration–response functions with no population thresholds within the range of documented ambient concentrations. This implies that exposure models for air pollution benefits assessment must be able to adequately characterize the influence of policies on long-term average concentrations of particulate matter, covering a broad geographic region given long-range transport of PM$_{2.5}$.

One frequently used approach has been to apply detailed fate and transport models for shorter pollution episodes and then to extrapolate the findings to an annual average basis. For example, in the U.S. EPA benefit-cost analysis of the Clean Air Act (1), particulate matter impacts were characterized using the Regional Acid Deposition Model (RADM)/Regional Particulate Model (RPM) in the Eastern United States and the Regulatory Modeling System for Aerosols and Deposition (REMSAD) in the Western United States. Both models covered only a small number of days (30 randomly selected 5-day periods over 4 years for RADM/RPM, four 10-day periods for REMSAD) and had relatively coarse grid resolution (80 km x 80 km for RADM/RPM, 56 km x 56 km for REMSAD). In addition, U.S. EPA has been developing the Community Multiscale Air Quality (CMAQ) model for future benefits assessments, which can capture many chemical and meteorological complexities but has largely been run on an episodic basis. While the sophistication of these models provides some clear advantages, use of this general approach for cohort mortality benefits estimation has been criticized, since these models were generally intended for evaluation of pollution episodes and exceedances of standards and are therefore relatively less useful to assess impacts due to changes in long-term exposure (5).

On the other hand, other health impact assessments have adopted somewhat different strategies. The regulatory impact analysis of the Tier 2 motor vehicle emission standards (2) used a source-receptor (S–R) matrix based on the Climatological Regional Dispersion Model (CRDM), which estimated impacts on an annual average basis at county-level resolution but was relatively less sophisticated in its consideration of atmospheric chemistry or pollutant fate and transport. Multiple applications have used CALPUFF (6–10), a U.S. EPA Guideline model that can account for secondary chemistry and long-range transport given detailed meteorological inputs, but with limited characterization of background concentrations. One study applied both REMSAD (as run in the benefit-cost analysis of the Clean Air Act) and S–R matrix to evaluate the health benefits of power plant pollution control (11). What these models and applications have in common is an ability to estimate concentration...
changes on an annual average basis, although with generally
less sophistication than the episodic models.

Despite the multitude of available models and past
applications, there has been very little deliberation about
the strengths and weaknesses of each model within the
context of health benefits assessment. This is in part because
most models have been validated from a pollution episode
perspective with a focus on performance at specific receptors
rather than a consideration of total population exposure. In
addition, model evaluation is hindered by the fact that it is
not possible to use field measurements for validation of
marginal source contributions when there are numerous
contributors to ambient concentrations and because alter-
native scenarios cannot be observed under identical condi-
tions. One approach for determining the robustness of model
results to assumptions about uncertain parameters is to
conduct sensitivity analyses for variables such as chemical
conversion and wet and dry deposition rates and evaluate
the impact on population exposure estimates. This approach
was adopted in two CALPUFF-based studies of power plant
health risks (9, 10). While this is an important step, many of
the parametric perturbations were somewhat artificial (e.g.,
omission of dry deposition), and the resulting changes in
population exposures were minor. More broadly, because
many model assumptions are fixed or constrained, this
approach may provide excessive confidence in the validity of
model estimates.

One way to explore the strengths and weaknesses of
models for health benefits assessment is to apply two
atmospheric models with different structures and assump-
tions to the same case study. Past studies involving head-
to-head dispersion model comparisons (12, 13) have focused
largely on plume characteristics or relative performance as
a function of distance rather than considering ability to
adequately capture population exposures. One application
that applied two models in a benefits assessment context
(11) provided estimates of total health benefits with each
model but did not give pollutant-specific estimates or insight
about model strengths and weaknesses. Another comparison
of S–R matrix and REMSAD predicted population-weighted
average concentrations of PM$_{10}$ and PM$_{2.5}$ with both models,
including regional and urban/rural stratification, but similarly
lacked information about the relative performance for
different particle constituents (14).

In this study, we apply CALPUFF to a set of seven power
plants in northern Georgia, using the same meteorological
year and source characteristics as in S–R matrix (Table 1,
Figure 1). We chose S–R matrix because of its role in past
regulatory impact analyses (2, 15) and studies of the health
impacts of power plant emissions (11, 16), its comprehensive
emissions inventory, its coverage of the continental United
States, and its calibration to ambient monitor data. However,
relative to CALPUFF, it has simplified meteorology, atmo-
spheric chemistry, and source characterization. CALPUFF
has also been used in past power plant applications (8, 9, 17)
but has substantially different assumptions from S–R matrix,
including more detailed meteorological data and complex
treatment of atmospheric chemistry but less comprehensive

<table>
<thead>
<tr>
<th>TABLE 1. Characteristics of Seven Power Plants in Georgia Used in Atmospheric Dispersion Models$^a$</th>
</tr>
</thead>
<tbody>
<tr>
<td>initial year on-line</td>
</tr>
<tr>
<td>emissions (tons, 1996)</td>
</tr>
<tr>
<td>PM$_{2.5}$</td>
</tr>
<tr>
<td>SO$_2$</td>
</tr>
<tr>
<td>NO$_x$</td>
</tr>
</tbody>
</table>

$^a$ All emissions and stack data taken from 1996 National Emission Trends data; emissions rounded to two significant figures.

FIGURE 1. CALPUFF receptor grid (points within 500 km of Atlanta) and sites of seven power plants in northern Georgia.
emissions and ambient concentration data.

To summarize the results of the two models in a way that is directly applicable to the estimation of health effects, we apply the concept of an intake fraction, used in selected settings for decades (6, 18–20) and recently articulated as a broadly applicable definition (21). Intake fraction (also referred to as exposure efficiency or dose fraction) is a unitless value defined as the fraction of a pollutant or its precursor emitted from a source that is eventually inhaled by someone. It can be calculated as

\[
IF = \frac{BR \times \sum (AC_i \times N_i)}{Q}
\]

(1)

where \(IF\) = intake fraction; \(BR\) = population-average breathing rate (assumed to be 20 m\(^3\)/day); \(AC_i\) = change in concentration of pollutant at receptor \(i\) given emissions from selected source (\(\mu g/m^3\)); \(N_i\) = number of people at receptor \(i\); and \(Q\) = emission rate of pollutant or pollutant precursor from selected source (\(\mu g/day\)). If the health effects from the pollutant have a linear dose–response function with no threshold above ambient concentrations or dose rate dependence, a straightforward calculation of an intake fraction based on annual average concentrations corresponds directly to health benefits estimates.

Using each dispersion model, we calculate pollutant-specific intake fractions for each plant to gain insight about the magnitude of population exposures per unit emissions and the implications of model differences. We conduct a parametric sensitivity analysis on the relative ambient concentrations of sulfate, nitrate, and ammonia within S–R matrix, both for the case study power plants and for other plants around the country for which CALPUFF-based intake fractions have been estimated. We conclude by estimating the mortality impacts corresponding with the power plant emissions to evaluate which pollutants are likely to contribute most to overall health benefits and are therefore more important to estimate accurately in this context. Although this analysis does not yield definitive model validation or uncertainty estimates per se, this approach provides significant insight into the importance of underlying variables for assessing population exposure and risks.

Source Characteristics

For our analysis, we selected a subset of power plants in a defined geographic region that were potentially relevant in a public policy context. We initially chose all older fossil-fueled power plants (“grandfathered” under the Clean Air Act) in Georgia within a 100 mile (161 km) radius of Atlanta. This region was chosen given the large number of coal-fired power plants with limited pollution controls and the lack of previous power plant health impact assessments in the Southeast. We focused on the Atlanta area since proximity to a major metropolitan area is likely to correspond with increased risk per unit emissions and because ongoing studies of air pollution health effects in Atlanta (22, 23) could ultimately feed into a more comprehensive risk assessment. For simplicity, we eliminated two plants (Arkwright and Atkinson) that have minimal emissions and operate only a fraction of the year, leaving seven power plants for our analysis. Basic characteristics of these plants are provided in Table 1.

To ensure comparability between the two dispersion models, we matched stack characteristics as closely as the model structures would allow. Since S–R matrix results were taken from an existing database, this implied that CALPUFF inputs were constructed to be comparable to inputs for S–R matrix. We modeled impacts of primary filterable PM\(_{2.5}\) emissions as well as emissions of SO\(_2\) and NO\(_x\) (as precursors of ammonium sulfate and ammonium nitrate). Although these emissions do not necessarily correspond with projected future emissions, the total tonnage emitted in 2000 was similar to 1996 emissions (approximately 10% greater in 2000 for all three pollutants). For comparability with S–R matrix, all emission rates were assumed to be uniform across the year. In addition, for comparability with past and future regulatory applications, each model employed EPA-recommended chemical conversion modules and assumptions. The only difference in the stack characteristics in the two models was the use of actual stack heights in CALPUFF, which were not used directly in S–R matrix as described below.

S–R Methodology

The details of the derivation of the S–R matrix used in our analysis are available elsewhere (11). Briefly, this matrix contains county-to-county transfer factors across the United States for primary particles (both PM\(_{2.5}\) and PM\(_{10}\) and secondary particles (including sulfates and nitrates), based on an adjusted version of the Climatological Regional Dispersion Model (CRDM). CRDM uses simple climatological summaries based on 1990 meteorological data (annual average mixing heights and joint frequency distributions of wind speed and direction) along with a sector-averaged dispersion model across 16 wind directions. Deposition and chemical conversion are incorporated at each receptor location with relatively simple assumptions, although it should be noted that these assumptions are used to develop “first guess” ambient concentration estimates, which are then calibrated based on ambient concentration monitoring data. The model assumes dry deposition rates of 0.1 cm/s for particles, 0.5 cm/s for SO\(_2\), and 1 cm/s for NO\(_x\), gaseous nitrate, and ammonia. Wet deposition rates (in cm/s) are calculated as a function of the annual precipitation rate in each county in inches (P): 0.08P for particles, 0.008P for SO\(_2\), 0.014P for ammonia, and 0.025P for NO\(_x\). Chemical conversion is incorporated by assuming sulfate oxidation to be a function of relative humidity (0.5% hour for relative humidity less than 40% up to 1.5% hour for relative humidity above 70%) and nitrate oxidation to be a constant 2%/hour.

In CRDM, sources were grouped into four categories to simplify the modeling—point sources with effective stack heights less than 250 m, 250–500 m, and more than 500 m, along with area sources. All seven power plants in our analysis fall within the 250–500 m category. Unit emission rates of 1 \(\mu g/s\) were used for all sources to develop the S–R matrix, which was applied to a 1996 emissions inventory and then adjusted to match with ambient concentration monitor data. The ambient concentration adjustment included separate consideration of sulfate, nitrate, and ammonium concentrations, with a simplifying assumption that particle sulfate is neutralized by ammonium and particle nitrate forms when ammonium levels are sufficient and the temperature is sufficiently low (which is assumed to occur only during one-quarter of the year) (24). The S–R matrix methodology to deal with rate-limiting constituents implied that reductions in SO\(_2\) could lead to increases in ammonium nitrate formation by increasing the available ammonium.

To appropriately incorporate the nonlinearities of the sulfate–nitrate–ammonium system, we estimated ambient concentrations with all sources and then with the removal of each power plant individually. We separated the effects of the SO\(_2\) and NO\(_x\) emissions by first considering the influence of SO\(_2\) emission reductions on the formation of ammonium sulfate and ammonium nitrate (with NO\(_x\) emissions held constant) and then modeling the influence of NO\(_x\) emission reductions on the formation of ammonium nitrate (with SO\(_2\) emissions held constant).
Our domain contained approximately 33 million individuals, weighted averages from all census tracts within a county. R matrix results, we calculated population-county-level S domain (total of 1806 receptors). For direct comparisons and used county-level resolution in the remainder of the placed receptors at each census tract within 200 km of Atlanta to this restricted domain is evaluated in our analysis. To limitations at longer range. Potential underestimation due evaluated impacts across the continental United States, we plants in Georgia

for Environmental Prediction global reanalysis, which pro-
av 6.2 × 10–7
SO2/sulfate
Bowen 1.6 × 10–7
Hammond 1.6 × 10–7
Harlee Branch 1.5 × 10–7
J ack McDonough 1.7 × 10–7
Scherer 1.5 × 10–7
Wansley 1.5 × 10–7
Yates 1.6 × 10–7
emission-weighted av 1.6 × 10–7

VOL. 37, NO. 24, 2003 / ENVIRONMENTAL SCIENCE & TECHNOLOGY

In contrast with S–R matrix, CALPUFF is a Lagrangian puff model that utilizes time-resolved three-dimensional wind-fields and detailed meteorology, source characteristics, and limited background pollution data. For our CALPUFF modeling, our receptor region consisted of points within 500 km of Atlanta (33.65°N, 84.42°W) (Figure 1). Although S–R matrix evaluated impacts across the continental United States, we selected a smaller domain for CALPUFF due to the complexity of developing meteorological fields and because of model limitations at longer range. Potential underestimation due to this restricted domain is evaluated in our analysis. To provide greater spatial resolution closer to the sources, we placed receptors at each census tract within 200 km of Atlanta and used county-level resolution in the remainder of the domain (total of 1806 receptors). For direct comparisons with county-level S–R matrix results, we calculated population-weighted averages from all census tracts within a county. Our domain contained approximately 33 million individuals, using 2007 population projections to provide comparability with past S–R matrix analyses (11). Intake fractions and health risks would be slightly lower using current population estimates; the ratio of 2007 estimated population to 1990 population (the basis of many past assessments) is approximately 1.2.

Our CALPUFF modeling methodology is described in depth elsewhere (9). Briefly, we used CALMET version 5.2 (000602a), CALPUFF version 5.5 (010730 1), and CALPOST version 5.2 (991104d) (Earth Tech, Concord, MA). Given available data for 1990, upper-air data were taken from the National Center for Atmospheric Research/National Centers for Environmental Prediction global reanalysis, which provided spatial resolution of 2.5° × 2.5° (roughly 250 km) at 19 vertical levels with 6-h time resolution. Surface observations were taken from the SAMSON dataset from the National Climatic Data Center, providing hourly observations from 262 stations across the United States. Despite the lower resolution, the meteorological data were generally complete and the resulting CALMET fields displayed reasonable characteristics and behaviors.

In our CALPUFF runs, we used the default wet and dry deposition model routines and the MESOPUFF II chemical transformation mechanism for SO2 and NOx (25). Dry deposition rates vary as a function of terrain as well as meteorological conditions and pollutant, while wet depo-

<table>
<thead>
<tr>
<th>TABLE 2. Intake Fractions for Primary PM2.5 and Secondary Ammonium Sulfate Using CALPUFF and S–R Matrix for Seven Power Plants in Georgia</th>
</tr>
</thead>
<tbody>
<tr>
<td>(&lt;500 km of Atlanta)</td>
</tr>
<tr>
<td>primary PM2.5</td>
</tr>
<tr>
<td>Bowen</td>
</tr>
<tr>
<td>Hammond</td>
</tr>
<tr>
<td>Harlee Branch</td>
</tr>
<tr>
<td>J ack McDonough</td>
</tr>
<tr>
<td>Scherer</td>
</tr>
<tr>
<td>Wansley</td>
</tr>
<tr>
<td>Yates</td>
</tr>
<tr>
<td>emission-weighted av</td>
</tr>
<tr>
<td>SO2/sulfate</td>
</tr>
<tr>
<td>Bowen</td>
</tr>
<tr>
<td>Hammond</td>
</tr>
<tr>
<td>Harlee Branch</td>
</tr>
<tr>
<td>J ack McDonough</td>
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<tr>
<td>Scherer</td>
</tr>
<tr>
<td>Wansley</td>
</tr>
<tr>
<td>Yates</td>
</tr>
<tr>
<td>emission-weighted av</td>
</tr>
</tbody>
</table>

Results

For primary PM2.5, intake fractions as calculated by CALPUFF and S–R matrix are quite similar across the seven power plants within the 500 km receptor region defined by CALPUFF (Table 2). Intake fractions range between 5 × 10–7 and 1 × 10–6 using CALPUFF, compared with a range of 5 × 10–7 to 6 × 10–7 when S–R matrix is applied to the identical receptor region. When we compare emission-weighted averages, which would be proportional to health impact estimates, the values are within 10% of one another. The one plant where values differ more substantially (Jack McDonough) has high population density close to the plant, potentially demonstrating the influence of a more refined receptor domain in CALPUFF. The 500 km radius receptor region captures approximately 70% of the total intake fraction in S–R matrix, indicating that most of the impact is found within the restricted CALPUFF domain with a small but consistent degree of underestimation.

As anticipated, due to the relative importance of long-range transport, there is less variation in secondary particulate matter intake fractions across power plants in a similar geographic region. For secondary ammonium sulfate per unit SO2 emissions, intake fractions as calculated by CALPUFF are essentially identical across power plants, with values of...
TABLE 3. Intake Fractions for Secondary Ammonium Nitrate Associated with NO\textsubscript{x} Emissions, Using CALPUFF, S–R Matrix with Ambient Ammonia, and S–R Matrix with No Ammonia Limitations for Seven Power Plants in Georgia

<table>
<thead>
<tr>
<th>Plant</th>
<th>S–R matrix (≤ 50 km, ambient ammonia)</th>
<th>S–R/CALPUFF (≤ 50 km, ambient ammonia)</th>
<th>S–R/CALPUFF (≤ 50 km, unlimitted ammonia)</th>
<th>S–R matrix (total, ambient ammonia)</th>
<th>S–R matrix (total)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bowen</td>
<td>6.7 x 10\textsuperscript{-8}</td>
<td>2.7 x 10\textsuperscript{-8}</td>
<td>0.4</td>
<td>4.2 x 10\textsuperscript{-8}</td>
<td>0.6</td>
</tr>
<tr>
<td>Hammond</td>
<td>7.1 x 10\textsuperscript{-8}</td>
<td>2.7 x 10\textsuperscript{-8}</td>
<td>0.4</td>
<td>4.2 x 10\textsuperscript{-8}</td>
<td>0.6</td>
</tr>
<tr>
<td>Harlee Branch</td>
<td>5.9 x 10\textsuperscript{-8}</td>
<td>2.3 x 10\textsuperscript{-8}</td>
<td>0.4</td>
<td>3.9 x 10\textsuperscript{-8}</td>
<td>0.7</td>
</tr>
<tr>
<td>J ack McDonough</td>
<td>7.0 x 10\textsuperscript{-8}</td>
<td>2.6 x 10\textsuperscript{-8}</td>
<td>0.4</td>
<td>4.1 x 10\textsuperscript{-8}</td>
<td>0.6</td>
</tr>
<tr>
<td>Scherer</td>
<td>5.9 x 10\textsuperscript{-8}</td>
<td>2.2 x 10\textsuperscript{-8}</td>
<td>0.4</td>
<td>3.9 x 10\textsuperscript{-8}</td>
<td>0.7</td>
</tr>
<tr>
<td>Wansley</td>
<td>6.0 x 10\textsuperscript{-8}</td>
<td>2.7 x 10\textsuperscript{-8}</td>
<td>0.4</td>
<td>4.3 x 10\textsuperscript{-8}</td>
<td>0.7</td>
</tr>
<tr>
<td>Yates</td>
<td>6.9 x 10\textsuperscript{-8}</td>
<td>2.7 x 10\textsuperscript{-8}</td>
<td>0.4</td>
<td>4.4 x 10\textsuperscript{-8}</td>
<td>0.6</td>
</tr>
<tr>
<td><strong>emission-weighted av</strong></td>
<td><strong>6.4 x 10\textsuperscript{-8}</strong></td>
<td><strong>2.5 x 10\textsuperscript{-8}</strong></td>
<td><strong>0.4</strong></td>
<td><strong>4.1 x 10\textsuperscript{-8}</strong></td>
<td><strong>0.6</strong></td>
</tr>
</tbody>
</table>

TABLE 4. Intake Fractions for Secondary Ammonium Sulfate and Ammonium Nitrate Per Unit of SO\textsubscript{2} Emissions, Using S–R Matrix for Seven Power Plants in Georgia

<table>
<thead>
<tr>
<th>Plant</th>
<th>SO\textsubscript{2}/sulfate IF</th>
<th>SO\textsubscript{2}/nitrate IF</th>
<th>% reduction in particle formation by including SO\textsubscript{2}/nitrate effect (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bowen</td>
<td>3.4 x 10\textsuperscript{-7}</td>
<td>-3.9 x 10\textsuperscript{-8}</td>
<td>11</td>
</tr>
<tr>
<td>Hammond</td>
<td>3.4 x 10\textsuperscript{-7}</td>
<td>-3.8 x 10\textsuperscript{-8}</td>
<td>11</td>
</tr>
<tr>
<td>Harlee Branch</td>
<td>3.0 x 10\textsuperscript{-7}</td>
<td>-3.8 x 10\textsuperscript{-8}</td>
<td>13</td>
</tr>
<tr>
<td>J ack McDonough</td>
<td>3.2 x 10\textsuperscript{-7}</td>
<td>-3.9 x 10\textsuperscript{-8}</td>
<td>12</td>
</tr>
<tr>
<td>Scherer</td>
<td>3.0 x 10\textsuperscript{-7}</td>
<td>-3.8 x 10\textsuperscript{-8}</td>
<td>13</td>
</tr>
<tr>
<td>Wansley</td>
<td>3.3 x 10\textsuperscript{-7}</td>
<td>-3.9 x 10\textsuperscript{-8}</td>
<td>12</td>
</tr>
<tr>
<td>Yates</td>
<td>3.3 x 10\textsuperscript{-7}</td>
<td>-4.0 x 10\textsuperscript{-8}</td>
<td>12</td>
</tr>
<tr>
<td><strong>emission-weighted av</strong></td>
<td><strong>3.2 x 10\textsuperscript{-7}</strong></td>
<td><strong>-3.8 x 10\textsuperscript{-8}</strong></td>
<td><strong>12</strong></td>
</tr>
</tbody>
</table>

*Domain includes continental United States.*

2 x 10\textsuperscript{-8} for all plants to one significant figure (Table 2). Over the same geographic domain, the S–R matrix intake fractions are nearly identical to those calculated by CALPUFF. For ammonium sulfate particulate matter, only half of the total population impacts are realized in the restricted 500 km radius domain.

For secondary ammonium nitrate per unit NO\textsubscript{x} emissions, CALPUFF yields intake fractions approximately a factor of 2.5 higher than S–R matrix within the 500 km domain (Table 3). This is likely related to differences in how the models address atmospheric transport, conversion, and partitioning of important species. We were able to quantify the influence of one potential factor, differences in ammonia background concentration and availability. Because the version of CALPUFF available at the time of the analysis calculated only plume sulfate and nitrate but characterized ambient ammonia as a constant and spatially uniform concentration, there has less ability to capture ammonia-limited conditions. If we increase ammonia concentrations in S–R matrix to avoid ammonia-limited conditions, the ammonium nitrate intake fraction estimates are increased significantly, although slightly lower than estimates using CALPUFF (Table 3).

A major advantage of S–R matrix over CALPUFF is its ability to estimate the effect of SO\textsubscript{2} emissions on ammonium nitrate concentrations for a marginal analysis. While CALPUFF does not calculate this marginal dependence, the output of S–R matrix allows it to be estimated using annual sulfate and nitrate changes along with available ammonia concentrations. In cases where there is sufficient nitrate, marginal increases in ammonia from decreased sulfate production allow for increases in ammonium nitrate and result in negative IF estimates for SO\textsubscript{2}/nitrate. The net effect is approximately 10% less particulate matter formation per unit SO\textsubscript{2} emissions than if effects on ammonium sulfate were considered alone. Table 4 shows IF estimates and ratios for these plants compared to a continental U.S. domain. The emission-weighted average IF for ammonium nitrate from SO\textsubscript{2} in the 500 km domain is approximately 10% less particulate matter formation per unit SO\textsubscript{2} emissions.

The majority of the results presented here concern the relative effects of CALPUFF and S–R matrix on aggregate population impacts. However, the analysis can also be informed by comparing the geographic patterns of incremental concentrations predicted by each model. For simplicity, we present the impacts of all seven power plants combined in Figure 2 and in Supporting Information Figures S1 through S3, since the general patterns for individual plants are similar.

For primary PM\textsubscript{2.5}, the concentrations predicted by S–R matrix and CALPUFF are within a factor of 2 over most of the domain (Figure 2 and S1); the difference is less than 20% over a significant portion of the domain. For secondary sulfates (Figures 2 and S2), the findings are similar, with minimal differences over much of the domain and slight elevations in S–R matrix relative to CALPUFF in the westernmost areas of the domain. For secondary nitrates, the story is a bit more complex, given the differences in how the models characterize the impacts of SO\textsubscript{2} emissions on ammonium nitrate reductions. For comparability with Table 2, we depict the ratio between S–R matrix and CALPUFF using the nitrate increments only associated with NO\textsubscript{x} emissions in S–R matrix (Figures 2 and S3). Because of the zero values recorded for some receptors in S–R matrix but not in CALPUFF, the ratio is zero at a number of locations but otherwise demonstrates similar spatial patterns as the other pollutants. In aggregate, the maximum annual average PM\textsubscript{2.5} increment associated with the seven power plants is 0.6 \mu g/m\textsuperscript{3} using CALPUFF and 0.9 \mu g/m\textsuperscript{3} using S–R matrix, with the difference principally related to the greater sulfate maximum for S–R matrix (Figure S2). This provides some indication that similar intake fractions could arise from differing spatial concentration patterns.

Finally, to demonstrate the approximate magnitude of health impacts associated with these sources and to illustrate the relative importance of various particle constituents, we calculate a point estimate for mortality impacts using standard epidemiological assumptions. We select a concentration–response function from a recent follow-up to the American Cancer Society cohort study (3), which implies a 0.6% increase in all-cause mortality per \mu g/m\textsuperscript{3} increase in annual mean PM\textsubscript{2.5} concentrations. Although the authors report multiple alternative values, this estimate is based on the average concentration data across the entire study period and is bounded by the concentration–response function implied by the other major population-based cohort mortality study (4) and the value used if cohort mortality is omitted entirely and only time-series epidemiology is used. For this illustrative calculation, we assume that all particle types have equal toxicity and that any population threshold would be

5532 • ENVIRONMENTAL SCIENCE & TECHNOLOGY • VOL. 37, NO. 24, 2003
FIGURE 2. Ratio of S–R matrix concentration impacts to CALPUFF concentration impacts for seven power plants in Georgia combined (primary PM$_{2.5}$, sulfate, and nitrate from NO$_x$, alone).
below the lowest ambient concentration in our receptor region.

To one significant figure, the resulting impacts from current emissions of seven selected power plants in northern Georgia are 500 deaths per year within 500 km of Atlanta, using either CALPUFF or S–R matrix. As expected from the relative magnitudes of the emissions (Table 1) and resulting concentrations, most of the health impacts are associated with sulfates. Approximately 86% of the total impacts for CALPUFF are due to sulfates, while the impact of sulfates is actually greater than the total for S–R matrix, because the decreased ammonium nitrate due to SO₂ emissions offsets the increased ammonium nitrate due to NOₓ emissions. If we extend S–R matrix across the continental United States, the health impact estimate for the seven power plants in Georgia doubles to approximately 1000 deaths per year. This reinforces the conclusion that the difference between the nitrate partitioning matter due to NOₓ emissions—are not unexpected, given the different ways each model addresses atmospheric transport, conversion, and partitioning of important species, particularly ammonia scavenging, deposition rates, and the oxidation of SO₂ to sulfate and NOₓ to nitrate. While many of these factors exert similar influences on secondary chemistry for both sulfate and nitrate particulate matter, the larger discrepancy between nitrate iF predictions may be attributed to additional differences in how each model accounts for the low temperature, high relative humidity, and high ammonia and nitric acid concentrations required for ammonium nitrate particulate matter formation. While the MESOPUFF II chemical conversion scheme in CALPUFF includes explicit state variables to estimate how nitric acid partitions between the gas and aerosol phases, S–R matrix assumes that necessary conditions only occur for one-quarter of the year and reduces raw predicted ammonium nitrate accordingly, by a simple factor of 4. Further, generally lower oxidation rates for NOₓ and higher deposition rates for NOₓ relative to SO₂ and sulfate in

![TABLE 5. Region-Stratified Mean Intake Fraction Ratios (S–R Matrix Divided by CALPUFF) for 40 Power Plants Across the United States](image)

<table>
<thead>
<tr>
<th>Region</th>
<th>N</th>
<th>PM₂.₅</th>
<th>SO₂/nitrate</th>
<th>NOₓ/nitrate, ambient ammonia</th>
<th>NOₓ/nitrate, unlimited ammonia</th>
</tr>
</thead>
<tbody>
<tr>
<td>South</td>
<td>12</td>
<td>0.5</td>
<td>2.2</td>
<td>0.6</td>
<td>1.1</td>
</tr>
<tr>
<td>West</td>
<td>5</td>
<td>1.8</td>
<td>3.4</td>
<td>0.3</td>
<td>0.6</td>
</tr>
<tr>
<td>Midwest</td>
<td>9</td>
<td>0.6</td>
<td>1.9</td>
<td>0.5</td>
<td>0.7</td>
</tr>
<tr>
<td>East</td>
<td>14</td>
<td>0.4</td>
<td>2.2</td>
<td>0.4</td>
<td>1.2</td>
</tr>
<tr>
<td>Nation</td>
<td>40</td>
<td>0.5</td>
<td>1.9</td>
<td>0.4</td>
<td>1.0</td>
</tr>
</tbody>
</table>

S–R matrix lead to decreased nitric acid production and thus may be a significant cause of relatively lower ammonium nitrate iF estimates in S–R matrix.

There are some limitations in the conclusions that can be drawn from our analysis. First, although we have shown the relationship between CALPUFF and S–R matrix in Georgia, this does not necessarily imply that the patterns would be identical in other locations. Given the dependence of secondary sulfate and nitrate formation on factors such as temperature and relative humidity, the relative model differences could vary regionally. To provide some preliminary insight about this question, we compare S–R matrix findings with intake fractions calculated using CALPUFF for 40 power plants across the United States (6, 17). Note that these 40 CALPUFF estimates were made in a prior analysis and input assumptions between CALPUFF and S–R matrix here do not parallel as neatly as in the main analysis.

Discussion

This analysis demonstrates that two dispersion models with somewhat different model structures and assumptions yield generally similar intake fraction estimates when compared on the same geographic domain, with greater differences for nitrates than for sulfates or primary PM₂.₅. We also show that use of a 500 km radius domain is likely to significantly underestimate population exposure and health risks, particularly for secondary pollutants. The magnitude of underestimation for power plants in Georgia is similar to that estimated previously for nine power plants in Illinois (9), which reported ratios (impacts within 500 km divided by total impacts across the continental United States) of approximately 0.8 for primary PM₂.₅ and 0.3 for secondary particles. The degree of underestimation is likely to be less for sites where many long-range downwind receptors have minimal populations (8), but it is clear that a larger domain is necessary for risk assessment of particulate matter from elevated point sources and will increase risk estimates over those calculated in regional analyses.

Though the reduced form of S–R matrix predictions and its subsequent calibration with ambient concentration data make a systematic sensitivity analysis and comparison to CALPUFF impractical, plausible explanations for many of the similarities and differences in model predictions may be made by contrasting the differing assumptions and subroutines between these models. The most significant differences in predicted intake fractions—which for ammonium nitrate particulate matter due to NOₓ emissions—are not unexpected, given the different ways each model addresses atmospheric transport, conversion, and partitioning of important species, particularly ammonia scavenging, deposition rates, and the oxidation of SO₂ to sulfate and NOₓ to nitrate. While many of these factors exert similar influences on secondary chemistry for both sulfate and nitrate particulate matter, the larger discrepancy between nitrate iF predictions may be attributed to additional differences in how each model accounts for the low temperature, high relative humidity, and high ammonia and nitric acid concentrations required for ammonium nitrate particulate matter formation. While the MESOPUFF II chemical conversion scheme in CALPUFF includes explicit state variables to estimate how nitric acid partitions between the gas and aerosol phases, S–R matrix assumes that necessary conditions only occur for one-quarter of the year and reduces raw predicted ammonium nitrate accordingly, by a simple factor of 4. Further, generally lower oxidation rates for NOₓ and higher deposition rates for NOₓ relative to SO₂ and sulfate in

...
presence of emissions from the source being assessed. Since source apportionment techniques of the precision required in the first condition are not available, and because the second condition is by definition impossible, we are left to rely upon modeling estimates of the marginal contribution to ambient PM from individual sources.

Secondly, lines of evidence do exist, however, to increase our confidence in model predictions. CALPUFF has performed favorably when compared with tracer field experiment results in Georgia, with similar transport times and directions and modeled estimates within a factor of 2 of observed concentrations (26). In other settings, CALPUFF has demonstrated little mean bias with respect to tracer releases (27). In studies with comprehensive emissions inventories, CALPUFF has yielded similar estimates to monitored data both for short-term pollution episodes (28) and for long-term average concentrations (29, 30). Clearly, good performance under these circumstances is not equivalent to the ability to predict population-weighted annual average concentration increments over a large domain, especially since these studies focused on primary pollutants, but it provides some support for the validity of CALPUFF in general.

Furthermore, according to CASTNET data from Pike County in Georgia (31), ambient concentrations in 1990 were approximately 6.5 g/m$^3$ of sulfate and 0.4 g/m$^3$ of nitrate. We estimated the contributions of seven power plants in Georgia to be about 10% of these ambient levels. This magnitude is plausible given the substantial contribution of long-range transport for secondary PM; while the modeled sources emit about 80% of SO$_2$ from the state of Georgia, they provide about 10% of SO$_2$ emissions from the states in our CALPUFF region (32). In addition, a recent study modeled the effects of SO$_2$ emission controls and estimated that a 1000 ton/day reduction from point sources in Georgia would yield average PM$_{2.5}$ reductions in Georgia of approximately 0.2–0.7 g/m$^3$ during two warm weather episodes (33). Our corresponding estimate was approximately 0.2 g/m$^3$ per 1000 tons/day of SO$_2$ emitted on an annual average basis. This evidence, along with the fact that CALPUFF and S–R matrix have very different model frameworks but yield similar findings, lends some support to the plausibility of our estimates, but the potential for bias must be explored in greater detail.

Finally, our findings are most relevant to health risk assessment given the assumption that the annual average concentration increment is the appropriate measure for estimating health risks. If future investigations demonstrate otherwise, between-model differences should be reexamined with different averaging periods. Similarly, our comparison implicitly assumes that decision makers are interested only in aggregate health impacts. If distributional concerns are important, differences in geographic patterns of concentrations may influence the evaluation of health risks and the development of optimal control strategies (although Supporting Information Figures S1 through S3 indicate that geographic distributions are generally similar).

Despite these limitations, our analysis allows us to draw some conclusions relevant for policy decisions and future analyses. First, it appears that both CALPUFF and S–R matrix are able to provide similar estimates of primary and secondary particulate matter exposure and thus risk assessments are likely to be relatively insensitive to choice between these two models. Between-model differences are greater for nitrates than for either sulfates or primary PM$_{2.5}$, implying that analyses of NO$_x$ control strategies may have greater dispersion modeling uncertainties than analyses of SO$_2$ or primary particulate matter control strategies. However, because mass emissions of NO$_x$ are substantially lower than SO$_2$ emissions for many fossil-fueled power plants, the impact of greater exposure uncertainty per unit emission of NO$_x$ is somewhat offset by the proportionally greater SO$_2$ emissions in the context of multipollutant control strategies. For all pollutants, it appears that a 500 km radius is inadequate to capture a substantial percentage of the intake fraction and population health risks, emphasizing the importance of accurate long-range dispersion modeling. A formal structured uncertainty analysis or value of information analysis would be needed to provide better quantitative evidence of the relative importance of different components of a power plant risk assessment. Future research efforts should address such needs, by supplementing these comparisons with calculations from other atmospheric models and other regions and evaluating within-model and between-model uncertainties for other source categories (including mobile sources).

Acknowledgments
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Supporting Information Available
Incremental primary PM$_{2.5}$ concentration impacts using CALPUFF and S–R matrix, seven power plants in Georgia combined (Figure S1); incremental secondary ammonium sulfate concentration impacts using CALPUFF and S–R matrix, seven power plants in Georgia combined (Figure S2); and incremental secondary ammonium nitrate concentration impacts of seven power plants in Georgia combined, using CALPUFF, S–R matrix with only NO$_x$ emissions, and S–R matrix with both SO$_2$ and NO$_x$ emissions (Figure S3). This material is available free of charge via the Internet at http://pubs.acs.org.

Literature Cited


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