Environ. Sci. Technol. **2011,** *45,* 189–196

Likelihood of Achieving Air Quality Targets under Model Uncertainties

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Received July 29, 2010. Revised manuscript received November 9, 2010. Accepted November 12, 2010.

Regulatory attainment demonstrations in the United States typically apply a bright-line test to predict whether a control strategy is sufficient to attain an air quality standard. Photochemical models are the best tools available to project future pollutant levels and are a critical part of regulatory attainment demonstrations. However, because photochemical models are uncertain and future meteorology is unknowable, future pollutant levels cannot be predicted perfectly and attainment cannot be guaranteed. This paper introduces a computationally efficient methodology for estimating the likelihood that an emission control strategy will achieve an air quality objective in light of uncertainties in photochemical model input parameters (e.g., uncertain emission and reaction rates, deposition velocities, and boundary conditions). The method incorporates Monte Carlo simulations of a reduced form model representing pollutant-precursor response under parametric uncertainty to probabilistically predict the improvement in air quality due to emission control. The method is applied to recent 8-h ozone attainment modeling for Atlanta, Georgia, to assess the likelihood that additional controls would achieve *fixed* (well-defined) or *flexible* (due to meteorological variability and uncertain emission trends) targets of air pollution reduction. The results show that in certain instances ranking of the predicted effectiveness of control strategies may differ between probabilistic and deterministic analyses.

Introduction

The United States Environmental Protection Agency (U.S. EPA) sets national ambient air quality standards (NAAQS) for ozone (O_3) and other criteria pollutants. States with ambient monitors violating those standards must develop State Implementation Plans (SIPs) for attaining the NAAQS by a future date. Recent proposed rules to tighten the NAAQS for O_3 and fine particulate matter ($PM_{2.5}$) will likely prompt a wave of new SIP development (*1, 2*).

In order to demonstrate future attainment, states use photochemical models to simulate the response of ambient pollution to projected reductions at emission sources. The current framework for SIP attainment demonstrations applies a *bright-line test* to deterministically evaluate whether an emission control program is sufficient (*3*). In this process, photochemical models simulate pollutant concentrations under ‘controlled’ (future-year) and ‘base’ (base-year) emission rates, applying identical base-year meteorological episodes in each case. The ratio of future to base pollutant concentrations is termed the relative reduction factor (RRF). This process enables the use of model results in a *relative* rather than an *absolute* sense. The RRF is then multiplied by the measured base-year design value (DVB) for each monitor to estimate the future design value (DVF), which determines whether the monitor is projected to attain the NAAQS with the considered set of control measures (*3*). Although U.S. EPA also advocates consideration of other “weight of evidence” factors in close cases, the deterministic *bright-line test* forms the core of most SIP attainment demonstrations.

However, photochemical model results are known to be uncertain due to uncertain model formulation (structural uncertainty) and uncertain input parameters (parametric uncertainty) (*4*−*6*). Thus, RRFs computed by photochemical models will be uncertain (*7*). Moreover, future meteorology will differ from the past, and those changes will impact pollutant concentrations (*8*). Whether a given control strategy will be sufficient is thus a probabilistic rather than a deterministic question, but the current *bright-line test* fails to quantify the likelihood that attainment will actually be achieved. In fact, many regions have failed to attain NAAQS by the targeted year despite SIP modeling that predicted attainment (*9*).

Hogrefe and Rao (2001) suggested that probabilistic analyses should supplement the pass/fail test of current regulatory practice (*10*). However, most previous efforts to characterize the probabilistic response of air pollutants to emission controls have relied upon numerous Monte Carlo photochemical model simulations (*11*−*13*), which is impractical for extensive SIP modeling. New methods would be needed to enable states to objectively characterize the attainment likelihood of various potential control packages in a computationally efficient manner.

This manuscript introduces methods for estimating the likelihood that a given level of emission reductions will achieve a targeted improvement in air quality, in light of parametric uncertainties in the photochemical model. Two types of targeted pollutant reduction are considered: a *fixed* amount of air pollution reduction needed at a monitor and a *flexible* function acknowledging that unknown future meteorology and uncertain projections of emission trends generate uncertainty in how much additional improvement is needed. The new methods are applied to recent attainment modeling from the Atlanta, Georgia, 8-h O_3 SIP to assess the likelihood that additional emission controls would achieve targeted amounts of air quality improvement.

Methodology

Reduced Form Models. Recent work has shown that high-order sensitivity analysis of a photochemical model can be applied to construct reduced form models (RFMs) that represent how perturbations in multiple input parameters (e.g., emission rates, reaction rate constants, boundary conditions, and deposition velocities) influence the responsiveness of pollutant concentrations to precursor emissions (*14, 15*). These RFMs provide analytical representations for the amount of ambient pollutant reduction that would be achieved as a function of the fractional changes (ε_j) in targeted

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10.1021/es102581e
Published on Web 12/07/2010

emission rates j = 1,2,...,J and the fractional perturbations ϕ_k needed to adjust uncertain parameters k = 1,2,...,K to their 'actual' values. Digar and Cohan (2010) introduced methods for efficiently computing the impacts of emissions perturbations while input parameters are perturbed (*14*). The Continuum RFM considers adjustable fractional perturbations in emissions, while the Discrete RFM is applicable when the tonnage of emission perturbation is predetermined (e.g., a specific control technology at a point source).

For the Continuum RFM, the change in concentrations (ΔC^*) resulting from fractional emission perturbation (ε_j) while input parameters P_k are perturbed by fractions ϕ_k is given by

$$\Delta C^* = C_{\varphi_k P_k} - C_{\varphi_k P_k, \varepsilon_j E_j} \approx -\left[\varepsilon_j S_j^{(1)} + \frac{1}{2}\varepsilon_j^2 S_j^{(2)} + \varepsilon_j \sum_k \varphi_k S_{j,k}^{(2)}\right] \tag{1a}$$

where $C_{\phi_k P_k}$ denotes concentrations under the input perturbations, and $C_{\phi_k P_k, \varepsilon_j E_j}$ are the corresponding concentrations when emission rate E_j is perturbed by fraction ε_j. $S_j^{(1)}(= \partial C/\partial \varepsilon_j)$ and $S_j^{(2)}(= \partial^2 C/\partial \varepsilon_j^2)$ are the local first- and second-order sensitivity coefficients of 'C' to the targeted emission rate, and $S_{j,k}^{(2)}(= \partial^2 C/\partial \varepsilon_j \partial \varphi_k)$ is the cross-sensitivity between parameter j and k. These coefficients are computed using the high-order decoupled direct method (HDDM) (*16, 17*), except for $S_{j,k}^{(2)}$ involving deposition velocities, which is computed by finite differencing of model runs. If the targeted emission rate E_j is also uncertain, then eq 1a can be rewritten as

$$\Delta C^* = -\left[(1 + \varphi_j)\varepsilon_j S_j^{(1)} + \frac{1}{2}(1 + \varphi_j)^2 \varepsilon_j^2 S_j^{(2)} + (1 + \varphi_j)\varepsilon_j \sum_k \varphi_k S_{j,k}^{(2)}\right] \tag{1b}$$

The $(1 + \phi_j)$ terms accounts for the influence of the uncertain emission inventory on the amount of tons controlled by fractional perturbation ε_j. For our analysis, ε_j represents emission control (i.e., $\varepsilon_j < 0$), so $C_{\phi_k P_k, \varepsilon_j E_j}$ is typically less than $C_{\phi_k P_k}$, and positive values of ΔC^* indicate pollutant reduction. Extensive testing of eq 1 (a and b) showed that ΔC^* is accurately predicted (normalized mean bias ≈ 6%, normalized mean error ≈ 10%) even for 50% emission controls under 50% simultaneous perturbations in 3 parameters (*14*).

The Discrete RFM allows accurate (normalized mean bias ≈ 3% and error ≈ 13% for 50% perturbations in 3 input parameters (*14*)) and efficient estimation of concentration response under input uncertainty when the magnitude of the emission reduction is predetermined. It computes the error-adjusted concentration response ΔC^* to an emission control by computing a function F_k that represents how concentration response to targeted emission change $\varepsilon_j E_j$ varies with change ϕ_k in parameter k (*14*)

$$F_k = (\Delta C_{perturbed} - \Delta C_{base})/\varphi_k \tag{2}$$

where $\Delta C_{perturbed}(= C_{\phi_k P_k} - C_{\phi_k P_k, \varepsilon_j E_j})$ and $\Delta C_{base}(= C_{base} - C_{\varepsilon_j E_j})$ represent concentration response under perturbed and base input conditions, respectively. Finite differencing of model runs with 10% input perturbations ($\phi_k = 0.1$) was used to compute F_k. ΔC^* is then calculated by the following Discrete RFM

$$\Delta C^* \approx \Delta C_{base} + \sum_k \varphi_k F_k \tag{3}$$

in which input perturbations can be set by Monte Carlo sampling of ϕ_k.

Probabilistic Framework and Reduction Targets. The Continuum (eq 1) and Discrete (eq 3) RFMs are analytical equations that can be evaluated readily for any perturbations ϕ_k in uncertain parameters k, in contrast to direct Monte Carlo simulation of a photochemical model (*11−13*). Here, we conduct Monte Carlo simulations of these RFMs, treating each input parameter as an independent log-normally distributed random variable with 1σ uncertainty listed in Table 1 based on earlier studies (*13, 14, 18−20*). The basis for selecting the input parameters is explained later. One million Monte Carlo sampling of ϕ_k are made to generate a probability distribution of the concentration reduction resulting from each targeted emission perturbation ε_j (Figure 1).

Our goal is to estimate the probability that a control strategy would actually achieve an air quality target in light of parametric uncertainty in the photochemical model. In this study, two types of pollutant reduction targets are considered:

(A) A *fixed* reduction target (T_{fixed}) which assumes that the amount of additional pollutant reduction needed for achieving the air quality improvement target is perfectly known, and only the impact (ΔC^*) of the control measures is uncertain due to input uncertainty. Thus, likelihood of attainment (L_{fixed}) is simply the probability that ΔC^* is greater than or equal to T_{fixed}, i.e.

$$L_{fixed} = p(\Delta C^* \geq T_{fixed}) \tag{4}$$

(B) A *flexible* reduction target ($T_{flexible}$) which recognizes that the needed amount of ambient pollutant reduction (ΔC^*) cannot be predicted perfectly because factors such as future weather and emission trends are unpredictable. In this case, likelihood of attainment ($L_{flexible}$) is assumed to be a function that increases with the amount of pollutant reduction (ΔC^*) that is achieved. Though various target functions could be posited, for analysis purposes we define a target function, $T(\Delta C^*)$, based on a cumulative distribution (cdf) of a Gaussian function as follows

$$T(\Delta C^*) = \int_{-\infty}^{\Delta C^*} N(x)dx \tag{5}$$

where $N(x) = e^{(-(x-\mu)^2/2\sigma^2)}/(\sigma(2\pi)^{1/2})$. The mean reduction target μ (at which a strategy would have 50% likelihood to be sufficient) and standard deviation $\pm\sigma$ can be assigned values depending on the case under consideration. In this study, an uncertainty of ±3 ppb (95% confidence interval) has been used, because current EPA methodology requires weight of evidence analysis if the deterministic attainment modeling results are within 3 ppb of the standard (*3*). Moreover, uncertainties in O_3 DVFs have been estimated to be 3−5 ppb due to variation in emission inventories and photochemical models (*21*) and 2−4 ppb due to variability in meteorology and chemical mechanisms (*7*). The final likelihood of attainment ($L_{flexible}$) for given emission controls under parametric uncertainty with the flexible reduction target (Figure S-1) can then be calculated using the probability density as

$$L_{flexible} = \int_{-\infty}^{+\infty} P(\Delta C^*)T(\Delta C^*)d\Delta C^* \tag{6}$$

Application

Photochemical Modeling Episode. We demonstrate this method by applying it to reconsider attainment modeling from a recent 8-h O_3 SIP for Atlanta, Georgia (*22*). Modeling is conducted for an 18-day summer episode (May 30 to June 16, 2002; first three days discarded for model initialization) for a southeastern U.S. modeling domain with 12 km grid resolution and 19 vertical layers of increasing thickness,

TABLE 1. Selection of Uncertain Input Parameters for Monte Carlo Analysis Based on the Impact Analysis by Digar and Cohan (2010) (*14*)

parameter	uncertainty in parameter[a] (1σ)	cross-sensitivity[b] (ppb)	impact on O_3 sensitivity[c]
Emission Rates			
domain-wide NO_X	0.336	−32.92	−**0.762**
domain-wide biogenic VOC	0.405	17.58	+**0.491**
domain-wide anthropogenic VOC	0.336	4.70	+**0.109**
Reaction Rate Constants			
all photolysis frequencies	0.347	16.45	+**0.393**
R($OH+NO_2$)	0.131	−9.30	−**0.084**
R($NO+O_3$)	0.095	−9.39	−**0.061**
R(all VOCs+OH)	0.095	8.24	+**0.054**
R($NO+HO_2$)	0.095	5.48	+0.036
R(C_2O_3+NO)	0.182	1.98	+0.025
R(PAN decomposition)	0.262	1.33	+0.024
R($C_2O_3+HO_2$)	0.294	−0.67	−0.014
R(RO_2+HO_2)	0.262	−0.54	−0.010
R(RO_2+NO)	0.262	0.40	+0.007
R(HO_2+HO_2)	0.095	−0.86	−0.006
R(NO_3+NO)	0.294	−0.10	−0.002
R($HCHO+NO_3$)	0.294	0.00	+0.000
Boundary Conditions			
boundary cond. O_3	0.203	0.41	+**0.006**
boundary cond. NO_Y	0.549	−0.10	−**0.004**
Others			
dry deposition velocity (all gaseous species)	0.223	−2.42	−**0.037**

[a] All distributions are log-normal (*13, 14, 18–20*). [b] Cross-sensitivity of O_3 to Atlanta anthropogenic non-EGU NO_X emissions and each uncertain parameter, evaluated at the grid-cell with maximum daily 8-h average O_3 in a 3 × 3 array centered on the Confederate Avenue monitor, averaged over the episode. [c] Impact factor: The fractional change in first-order sensitivity of ozone to emissions, due to a 1σ change in an input parameter. Computed as Impact Factor = $\sigma S^{(2)}_{j,k}/S^{(1)}_j$ where $S^{(1)}_j$ is the first-order sensitivity of O_3 to Atlanta NO_X and $S^{(2)}_{j,k}$ is the cross sensitivity of $S^{(1)}_j$ with an uncertain parameter. Bolding indicates parameters selected for analysis in this study.

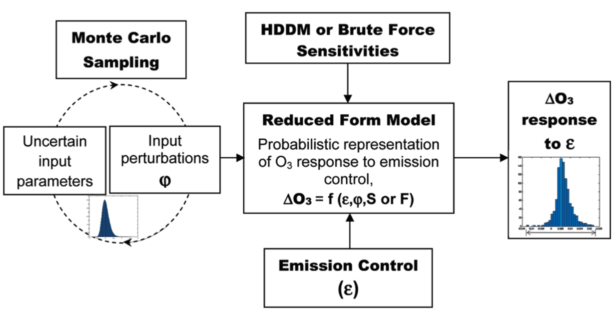

FIGURE 1. Probabilistic framework for characterizing ozone response to a control strategy under model parametric uncertainty.

covering Alabama, Georgia, Mississippi, South Carolina, Tennessee, and parts of Kentucky, North Carolina, and Florida. The episode is a subset of the full ozone season simulated for the Georgia SIP. Otherwise, modeling methods mimicked those of the Georgia SIP, including use of the Community Multiscale Air Quality (CMAQ) Model v4.5 (*23*) with Carbon Bond 4 (CB-IV) mechanism (*24*) with aerosol and aqueous updates; input meteorological conditions from the fifth generation Mesoscale Model (MM5) (*22, 25, 26*) simulations; and input emissions from Visibility and Improvements State and Tribal Association of the Southeast (VISTAS) year 2009 projections (projected from a 2002 base inventory) (*27, 28*), with updates to Georgia emissions projections based on GA EPD SIP modeling (*22*). Accuracy of O_3 predictions for the 2002 base case was thoroughly tested in Georgia SIP modeling and found to be well within U.S. EPA benchmarks (*22*).

Control Strategies. Ozone in Georgia is predominantly sensitive to NO_X emissions because of the dense forest cover leading to high biogenic VOC emissions (*29*); our modeling showed O_3 in the region to be at least an order of magnitude more sensitive to NO_X than to VOCs, consistent with earlier studies (*30*). Hence, for the selection of control options, NO_X emission reductions were emphasized. For simplicity, Georgia is divided into three broad regions (see Figure 2): Atlanta (the 20 county O_3 nonattainment region), Macon (7 counties), and the Rest of GA (= Georgia − Atlanta − Macon).

Our analysis sought to identify scenarios of control measures that could be implemented at the state level within a SIP time frame. These scenarios were constructed by applying AirControlNET v. 3.2 (*31*) to identify potential control options for the emission inventory. [A limited list of control technologies and associated control efficiencies obtained from AirControlNET is furnished as Supporting Information (Table S-1). Additional potential measures were also incorporated as described in Table S-2.] The maximum percent emission reduction from applying all identified control options in each region is tabulated in Table 2.

Power plant emissions are excluded from the regional categories and considered separately. Specifically we consider five large coal-fired power plants, which are among the largest NO_X point-sources near Atlanta and lacked selective catalytic reduction (SCR) control for NO_X when the Georgia SIP was being developed. Potential emission reductions at the power plants were computed by applying control efficiencies from U.S. EPA Integrated Planning Model methodology (*32*) to the inventoried emission rates, accounting for pre-existing control technologies where applicable (Table 2). Note that power plant controls are based on fixed tonnage reductions, whereas regional emission controls are based on percentage reductions.

Parameters for Uncertainty Analysis. Table 1 shows the input parameters that were targeted for uncertainty analysis due to the following reasons. Uncertainties in domain-wide NO_X and VOC emissions rates and in boundary conditions

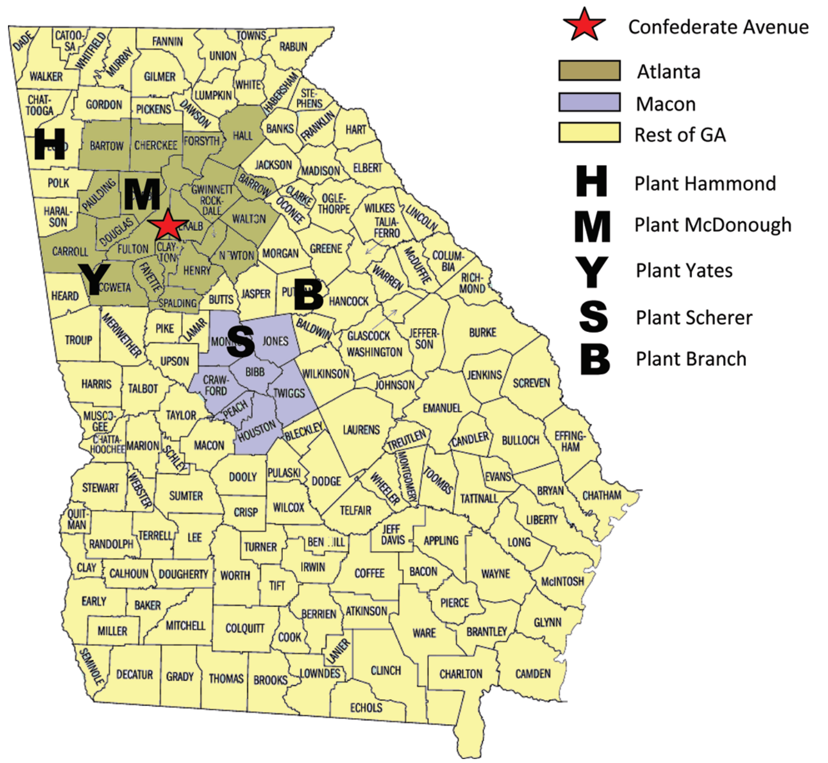

FIGURE 2. Point sources and emission regions in Georgia considered for control strategy analyses.

TABLE 2. Hypothetical NO_X Emission Control Options in Georgia

control scenario	description	emission controlled[a] fixed % reduction of total emission
Regional Sources		
ATL(12)	maximum available anthropogenic NO_X control in Atlanta	12% (42.7 tpd[b])
ATL(6)	half of available anthropogenic NO_X control in Atlanta	6% (21.3 tpd)
MAC	maximum available anthropogenic NO_X control in Macon	20% (10.7 tpd)
REST	maximum available anthropogenic NO_X control in Rest of Georgia (i.e., Georgia - Atlanta - Macon)	15% (81.5 tpd)
control scenario	**description**	**emission controlled[a] fixed tonnage reduction**
Point Sources (EGU)		
EGU(M)	convert Plant McDonough from coal to gas plus SCR[c]	10.0 tpd (85%)
EGU(S)	add SCR at Plant Scherer	26.5 tpd (50%)
EGU(Y)	add SCR at Plant Yates	29.8 tpd (80%)
EGU(H)	add SCR to units 1−3 at Plant Hammond	11.6 tpd (63%)
EGU(B)	add SCR at Plant Branch	51.7 tpd (80%)

[a] The basis for emission control estimates is explained in Tables S-1 and S-2. [b] tpd - tons per day. [c] SCR - Selective Catalytic Reduction.

of O_3 and total reactive nitrogen ($NO_Y = NO_X$ and its oxidation products) have been shown to substantially influence the sensitivities of O_3 to NO_X emissions (*4, 5, 11−13, 33*). Past studies have also shown that reaction rates for NO_2+OH (*34−36*) and the photolysis reactions (*37, 38*) and several other uncertain reactions (*13, 18*) can also significantly influence ozone sensitivity (Table 2). We also consider dry deposition velocities of all gaseous species jointly as an uncertain input parameter (*39*).

Our previous study evaluated the relative impacts of the 19 input parameters in Table 1 on estimates of O_3-precursor sensitivity in this region (*14*). For this study, we consider 10 of the 19 uncertain parameters marked in bold in Table 1, limiting the uncertain reaction rate constants to the four that most influenced O_3 sensitivity.

Results and Discussion

Based on the standard U.S. EPA attainment demonstration methodology (*3*), Georgia's 2009 SIP modeling predicted that one monitor (Confederate Avenue, AIRS ID: 13-121-0055, for location see Figure 2) would exceed the 1997 8-h O_3 NAAQS of 84 ppb (Ref Table 6-1 on page 133 of ref *22*). The SIP reports additional modeling and weight of evidence analyses to argue that attainment would actually be achieved. However, it can be computed that an additional 1.5 ppb reduction in modeled 2009 8-h O_3 would have been needed to reduce the relative reduction factor (RRF) in the Georgia SIP (Ref Table 6-1 on p 133 of ref *22*) sufficiently to demonstrate NAAQS attainment using the standard methodology (Supporting Information). Hence for this study, we consider the hypothetical scenario that an additional 1.5 ppb of improvement is necessary at this monitor and explore various control scenarios available in Georgia for reaching that target.

Likelihood To Achieve a Fixed Target. We first assess the likelihood that each control scenario will achieve at least 1.5 ppb reduction in 8-h O_3 at the grid-cell corresponding to the Confederate Avenue monitor, averaged over the six days with O_3 in the base year 2002 exceeding 80 ppb (Table 3). The deterministic results are from the base model ($\phi_k = 0$), with the standard deviation of the daily O_3 reductions shown as an indicator of the variability in results due to day-to-day changes in emissions and meteorology. The probabilistic

TABLE 3. Reduction in 8-h Ozone at Atlanta Confederate Avenue Monitor Due to Each Emission Control Package

control strategy[a]	description	deterministic O_3 reduction[b] (ppb)	day-to-day variation (σ) of deterministic O_3 reduction[c] (ppb)	O_3 reduction under parametric uncertainty, mean (5th, 95th percentiles)[d] (ppb)	likelihood to achieve T_{fixed}[e]	likelihood to achieve $T_{flexible}$[f]
C1	ATL(6)	1.1	0.4	1.0 (−0.7, 2.1)	19.6%	37.5%
C2	EGUs (B, S)	1.2	1.1	1.2 (0.7, 1.7)	4.4%	41.1%
C3	ATL(6) + MAC + REST	1.4	0.4	1.2 (−0.4, 2.5)	37.1%	44.9%
C4	ATL(6) + EGU(M)	1.7	0.8	1.5 (−0.6, 3.2)	57.8%	52.5%
C5	EGUs (B, S, H)	1.7	1.7	1.7 (1.2, 2.3)	77.9%	56.1%
C6	ATL(6) + EGUs (B, S)	2.2	1.3	2.1 (0.1, 3.6)	78.4%	63.5%
C7	ATL(12)	2.3	0.8	2.0 (−1.2, 4.5)	71.7%	62.6%
C8	EGUs (M, B, H, S)	2.3	1.1	2.4 (1.5, 3.3)	94.4%	70.6%
C9	ATL(12) + MAC + REST	2.6	0.8	2.3 (−0.9, 5.0)	78.0%	67.8%
C10	ATL(12) + EGU(M)	2.8	1.2	2.7 (−1.1, 5.6)	79.9%	71.9%
C11	EGUs (M, B, H, S, Y)	2.9	2.8	2.9 (1.2, 7.6)	99.9%	81.7%
C12	ATL(12) + EGUs (B, S)	3.4	1.6	3.2 (−0.3, 6.0)	86.6%	79.0%
C13	ATL(12) + EGUs (B, H, S)	4.0	2.1	3.7 (0.3, 6.4)	90.3%	84.5%
C14	ATL(12) + EGUs (M, B, H, S, Y)	5.1	3.2	5.0 (1.1, 8.1)	94.0%	91.5%

[a] In ascending order based on deterministic O_3 reduction. [b] Mean of the impacts among the high ozone days in episode; underlining indicates O_3 reduction $\geq$1.5 ppb. [c] Standard deviation of the daily impacts within the high O_3 days of the episode. [d] 90% confidence intervals. [e] *Fixed* reduction target of 1.5 ppb. [f] *Flexible* reduction target of 1.5 ppb with 3 ppb uncertainty.

results reflect 1 million Monte Carlo samplings of the input ϕ_k's for the RFMs. A Continuum RFM was constructed to predict the impact of each regional control scenario and a Discrete RFM for each power plant option, under parametric uncertainties in the 10 selected parameters from Table 1. Impacts of jointly controlling NO_X from multiple regions or power plants were assumed to be additive. This is a conservative assumption that may slightly underpredict joint impacts, since controlling NO_X in one place makes O_3 more sensitive to NO_X from elsewhere (*30*). The error caused by this assumption is small for controls of these magnitudes (*14*).

Table 3 presents deterministic and probabilistic estimates of the impacts of 14 hypothetical control strategies; Figure 3 shows how results vary as greater amounts of Atlanta emission reductions are applied. We focus on the extent to which the probabilistic methods influence the rankings of strategy impacts, to explore the importance of these methods to control strategy prioritization. The control options that achieve the 1.5 ppb O_3 reduction target under deterministic modeling (underlined) exhibit a range of likelihood for achieving this target when parametric uncertainties are considered (Table 3). The deterministic rankings of control strategy impact, indicated by the listing order in Table 3 and the ranking-scale in Figure 3(a), are largely preserved in the probabilistic modeling but with notable differences. Maximal Atlanta-only controls (C7) yield more O_3 reduction than SCRs at three distant power plants (C5) in the deterministic modeling (2.3 ppb vs 1.7 ppb) but a smaller likelihood of achieving the fixed target (L_{fixed} = 71.7% vs 77.9%). Meanwhile, strategies C1 (Atlanta-only partial control) and C2 (two power plants) are reversed in the deterministic and probabilistic rankings, and strategy C8 (four power plants) fares very differently between the rankings.

These ranking reversals occur in part because the parametric uncertainty analysis methods applied here show regional NO_X controls to have more uncertain O_3 impact than power plant-only controls (as indicated by the 90% confidence intervals for O_3 reduction in Table 3) for three reasons. First, the tonnage reduced is assumed to be perfectly known for the power plants (whose baseline emissions are well-established by continuous emission monitors (*40*)) but to vary with uncertainty in domain-wide NO_X for the regional controls, which are set on a percentage basis. Second, power plant controls have a consistently positive impact on O_3 reduction at a faraway monitor because aged, diluted NO_X plumes produce O_3 under a wide range of input parameter conditions. By contrast, local emissions can have a titrating or inhibiting effect on urban O_3 under certain input perturbations, especially if domain-wide NO_X emissions are much larger than originally modeled (Figure S-1). Finally, the likelihood calculations considered uncertainty in model parameters but not in meteorology and used results averaged over all high O_3 days of the episode. Distant power plant plumes have greater day-to-day variability in impacts (indicated by standard deviation in column 4 of Table 3) than regional sources because fluctuating wind fields determine whether the plume reach the monitor. For example, the C5 strategy controlling three distant power plants exhibits more than twice the day-to-day variability of C7, which controls only local Atlanta emissions. Longer episodes with classification and regression tree analysis (*41*) could be used to ensure that a representative range of high O_3 meteorological conditions have been modeled.

Likelihood to Achieve Flexible Target. The impacts of the control packages are reassessed for a *flexible* air pollutant reduction target, corresponding to eq 6 and Figure S-1, to reflect the fact that meteorological variability and other factors may make the needed amount of improvement uncertain. The results in Table 3 and Figure 3 show that when the reduction target is not accurately known, the chances of attainment are less responsive to the amount of emission control. For example, strengthening Atlanta NO_X controls from 6% to 12% (strategies C1 and C7) increases the L_{fixed} by 52 percentage points but increases $L_{flexible}$ by only 25 percentage points (Table 3). Similar trends can be seen in the flatter lines of Figure 3c than Figure 3b. This occurs because a *flexible* reduction target blurs the distinction between strategies that achieve just more or just less than 1.5 ppb of reduction. However, the results approach the *fixed* target results as the σ used to define $T_{flexible}$ is narrowed (Table S-3).

The likelihood rankings remain largely consistent under the *flexible* and *fixed* reduction targets but with some exceptions (Table 3 and Figure 3). For example, strategy C8 (four power plant controls) ranks second under the *fixed* reduction target but only sixth under the *flexible* reduction target. The relatively narrow uncertainty of power plant control impacts, modeled to occur for reasons explained

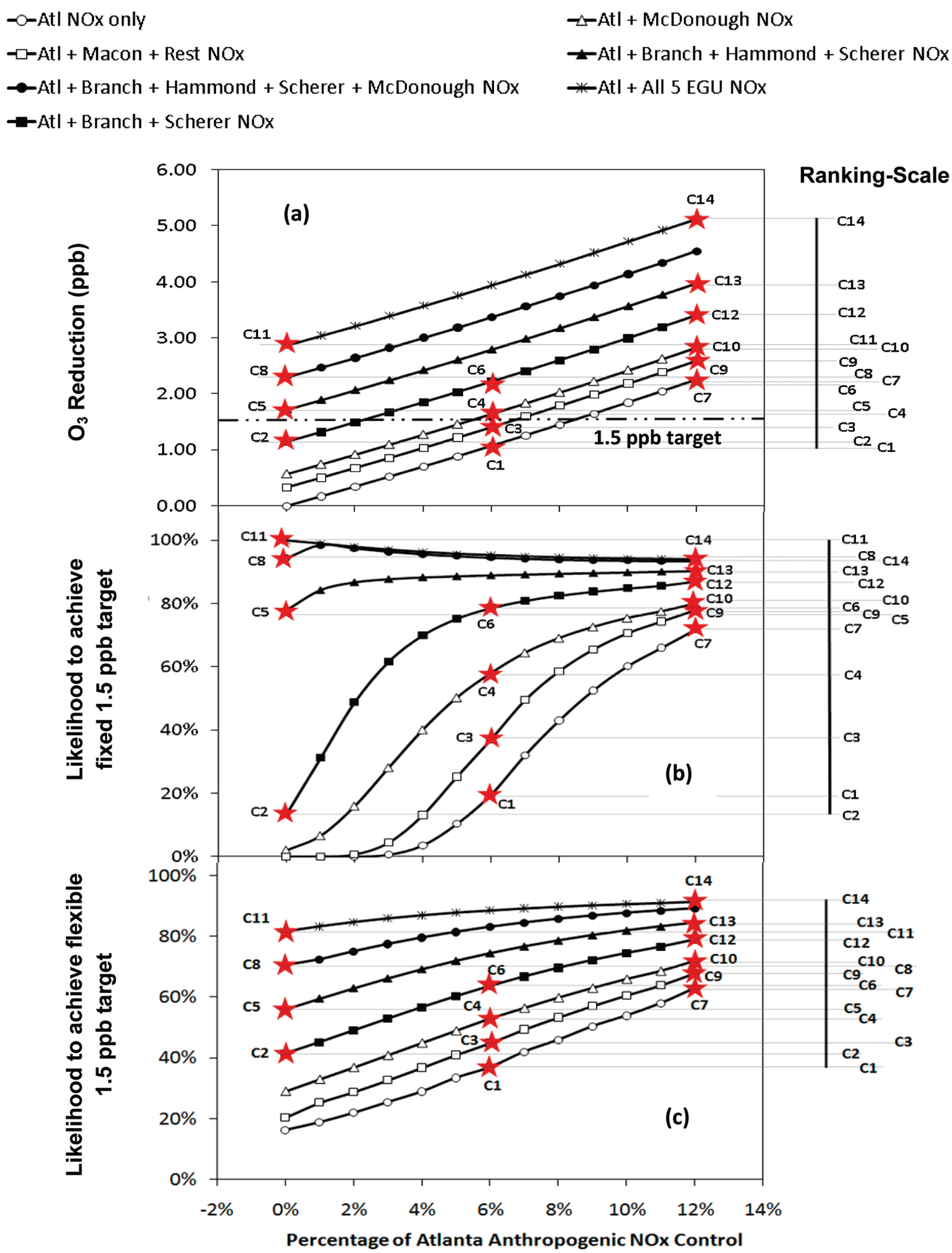

FIGURE 3. Predicted future O_3 design values (a) and likelihood of achieving a fixed (1.5 ppb) (b) or flexible (1.5 ± 3.0 ppb, 95% CI) (c) reduction target at Confederate Avenue monitor as a function of the percentage of Atlanta NO_X that is controlled under various scenarios for reducing NO_X emissions from other sources. Red stars denote control strategies detailed in Table 3.

above, is more helpful in achieving a *fixed* than a *flexible* reduction target, provided that the mean impact is above 1.5 ppb.

Relevance of Results. The approaches introduced here enable probabilistic prediction of the likelihood that a control package will be sufficient to achieve a *fixed* or *flexible* air quality improvement target in the presence of parametric uncertainties in the photochemical model. Both targets may usefully inform environmental decision-making, depending on how the policy issue is framed. The *fixed* target is apt if the needed amount of additional ozone reduction is clearly defined; for example, if regulatory approval of an attainment plan depends on demonstrating an additional increment of ozone abatement. A *flexible* target, meanwhile, is more attuned to predicting the likelihood of future attainment at monitors, which increases with the amount of control but is also influenced by external factors such as meteorological variation. Although the *flexible* target may obscure the distinctions between relative efficacies of control strategies, it avoids unrealistic expectations that a state's control choices could be so determinative of future attainment at monitors.

Results from these approaches could be linked with control cost estimates to maximize the likelihood of attainment, subject to practical or budgetary constraints, or may supplement deterministic approaches to inform the prioritization of control strategies (*42*). Actual selection of control measures depends upon a whole host of practical, economic, and political considerations, but our approaches could usefully inform strategy selection. Probabilistic approaches may also be used as additional 'weight of evidence' analyses in attainment demonstrations. However, probabilistic approaches are unlikely to supplant deterministic bright-line tests as the primary arbiter of attainment plan sufficiency because to do so would require subjective judgments about which model uncertainties to consider, the form of the target function, and what likelihood of attainment is sufficient.

Although only 8-h O_3 attainment was considered here, this method can also be applied for assessing control strategies for other pollutants. Application to particulate matter (PM) would need to account for differences in model performance among PM species and use an alternative method to compute sensitivity coefficients, since high-order DDM is currently unavailable for PM in CMAQ.

This analysis represents an important yet incomplete step toward comprehensive likelihood assessment because it considered uncertainties only in the photochemical model parameters and in the reduction target. The specific *flexible* target considered here is just one of many ways that such a target could be formulated. Structural uncertainties in the photochemical model, uncertainties in the meteorological inputs, and the representativeness of the meteorological episode were overlooked. Additional important uncertainties include control measure effectiveness (i.e., the percent or tons of emissions actually reduced by the abatement measures) and the accuracy of predicted baseline emission trends (e.g., due to economic and population growth, vehicle fleet turnover, etc.). Future work could incorporate these uncertainties into the likelihood assessments and explore alternate formulations of the target functions.

Acknowledgments

Although the research described in this article has been funded by U.S. EPA through Science to Achieve Results (STAR) grant #R833665 to D. Cohan, it has not been subjected to the Agency's required peer and policy review and therefore does not necessarily reflect the views of the Agency and no official endorsement should be inferred. While the research described in the article contains contributions from researchers at the Georgia Department of Natural Resources, it has not been subject to the Department's required peer and policy review and therefore does not necessarily reflect the views of the Department and no official endorsement should be inferred.

Supporting Information Available

Control measures and their efficiencies as obtained from AirControlNET (Table S-1), additional point-source and federal measures explored (Table S-2), variation of attainment likelihood of O_3 control strategies due to varying uncertainty of T_{fixed}, plots illustrating the probability distribution of O_3 reduction along with the likelihood to achieve a flexible reduction target due to a representative control scenario, and response of O_3 reductions to uncertainty in domain-wide NO_X. This material is available free of charge via the Internet at http://pubs.acs.org.

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ES102581E