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Accountability assessment of regulatory impacts on ozone and $PM_{2.5}$ concentrations using statistical and deterministic pollutant sensitivities

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Abstract Since the 1990 Clean Air Act Amendments, the USA has seen dramatic decreases in air pollutant emissions from a wide variety of source sectors, which have led to changes in pollutant concentrations: both up and down. Multiple stakeholders, including policy-makers, industry, and public health professionals, seek to quantify the benefits of regulations on air pollution and public health, a major focus of air pollution accountability research. Two methods, one empirical, the other based on a chemical transport model (CTM), are used to calculate the sensitivities of ozone (O_3) and particulate matter with diameters less than 2.5 μm ($PM_{2.5}$) to electricity-generating unit (EGU) and mobile source emissions. Both methods are applied to determine impacts of controls on daily concentrations (which are important in assessing acute health responses to air pollution), accounting for nonlinear, meteorologically, and emission-dependent responses of pollutant concentrations. The statistical method separates contributions of nearby EGU, regional EGU, and mobile source emissions on ambient city-center concentrations. Counterfactual emissions, an estimate of emissions under a scenario where no new controls were implemented on local EGU sources after 1995, regional EGUs after 1997, and mobile sources after 1993, are combined with these sensitivities to estimate counterfactual concentrations that represent what daily air quality in Atlanta, GA would have been had controls not been implemented and other emissions-reducing actions not been taken. Regulatory programs are linked with reduced peak summertime O_3, but have had little effect on annual median concentrations at the city-center monitoring site, and led to increases in pollutant levels under less photochemically-active conditions. The empirical method and the CTM method found similar relationships between ozone concentrations and ozone sensitivity to anthropogenic emissions. Compared to the counterfactual between 2010 and 2013, the number of days on which O_3 ($PM_{2.5}$) concentrations exceeded 60 ppb (12.0 $\mu g\, m^{-3}$) was reduced from 396 to 200 (1391 to 222). In 2013, average daily ambient O_3 and $PM_{2.5}$ concentrations were reduced by 1.0 ppb (2 %) and 9.9 $\mu g\, m^{-3}$ (48 %), respectively, and fourth highest maximum daily average 8-h O_3 was reduced by 14 ppb. Comparison of model-derived sensitivities to those derived using empirical methods show coherence, but some important differences, such as the O_3 concentration where the sensitivity to NO_x emissions changes sign.

Keywords Air pollution modeling · Sensitivity · Accountability · Statistical modeling

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Introduction

In response to the 1990 Clean Air Act Amendments and other measures, the US Environmental Protection Agency (US EPA), states, and local agencies have implemented a

number of policies that are designed to decrease emissions of pollutants linked with adverse health outcomes (e.g., USEPA 1999b, 2009, 2005, 2000a; Georgia EPD 2013). Goals of the various programs include bringing areas into compliance with the National Ambient Air Quality Standards (NAAQS) and reducing air toxics emissions (NAP 2004). Of the six criteria pollutants for which NAAQS are promulgated, ozone (O_3) and fine particulate matter have proven the most difficult to reduce below the standards levels. As of July 2016, 214 and 87 full or partial counties were designated in non-attainment for O_3 and particulate matter ($PM_{2.5}$ or PM_{10}), respectively, compared to 26 and 47 counties for lead and sulfur dioxide (SO_2), and none for carbon monoxide or nitrogen dioxide (NO_2) (http://www3.epa.gov/airquality/greenbook/ancl.html, *accessed 13 July, 2016*). These designations are based on the previous O_3 standard (0.75 *ppb*, which was changed to 0.70 *ppb* on 1 October, 2015) since the EPA will not update its non-attainment designations for the new standard until late 2017 (U.S. EPA 2015b).

In assessing the effectiveness of regulatory programs, a number of challenges arise (HEI 2003; van Erp et al. 2008). First, multiple regulations may be implemented at or near the same time across different sectors, making it difficult to disentangle the effects of a specific regulation from others. Second, controls may not have immediate effects, instead yielding an increasing effect over time, e.g., cleaner cars entering the vehicle fleet and replacing older, more polluting cars. Over those longer periods, other long-term changes in weather, land use, and other source emissions may occur. Third, compliance timelines and effectiveness may not be uniform over all targets of a regulation. In regard to stationary sources, operators may take actions such as retrofitting a plant to a new fuel, installing controls, or switching load between different plants for a variety of reasons besides current regulations, including anticipated future regulations, changing demand, and fuel costs (Georgia Power 2007; van Erp et al. 2008). Further, chemical reactions between certain atmospheric pollutants may result in different impacts on ambient concentrations from the reduction of multiple pollutants ($NO_x = NO + NO_2$ and VOCs—volatile organic compounds—in the case of ozone) than to reductions of a single pollutant (e.g., Cohan et al. 2005; Seinfeld and Pandis 2006). Further still, variations in meteorological conditions lead to differences in reaction rates, atmospheric transport, and deposition that affect pollutant concentrations.

The present study uses ambient air pollution concentrations, measured emissions from power plants (also called electricity generating units—EGUs), and modeled mobile source emissions in Atlanta, Georgia from 1999 to 2013 to develop counterfactual time series of ozone and $PM_{2.5}$ that assume no additional policies were implemented after 1995 on local EGUs, 1997 on regional EGUs, and 1993 on mobile sources. Important EGU regulatory programs implemented during this period include (year the program began) the Acid Rain Program (1995), the Clean Air Interstate Rule (CAIR—2008), and the Georgia Multipollutant Rule (2009). Mobile source programs include the Georgia Gasoline Marketing Rule (1999), the Tier 2 Vehicle and Gasoline Sulfur Program (2004), and the Heavy-Duty Highway Rule (2007), USEPA (1999a, 2000b, 2005, 2012a; EPD 2014).

Researchers have calculated ambient pollutants sensitivities to emissions using statistical methods (e.g., Blanchard et al. 2010; Harrington et al. 2012) and using first-principles chemical transport models (CTMs), both by brute force (e.g., Digar and Cohan 2010 and Xie et al. 2011) and direct methods (e.g., Dunker 1981, 1984; Cohan et al. 2005; Liao et al. 2008; Hakami et al. 2004). The Community Multiscale Air Quality Model with the Decoupled Direct Method (CMAQ-DDM) and the Comprehensive Air Quality Model with Extensions (CAMx, which includes DDM) are examples of models that can calculate sensitivities directly.

CTMs such as CMAQ-DDM and CAMx offer the benefit of incorporating detailed physics and chemistry parameterizations in the calculations of concentrations and sensitivities. Studies have shown that results are sensitive to uncertainties in meteorological inputs (Appel et al. 2007; Byun et al. 2007; Gilliland et al. 2008), emissions inputs (Byun et al. 2007; Gilliland et al. 2008; Zhang et al. 2015), and the combination of chemical and physical mechanisms employed in the model (Hanna et al. 2001; Appel et al. 2007; Byun et al. 2007; Gilliland et al. 2008). In spite of these uncertainties, CTMs have been demonstrated to accurately simulate ambient concentrations and capture observed trends under changing emissions in dynamic evaluations (Foley et al. 2015b; Foley et al. 2015a; Zhou et al. 2013).

Statistical models do not explicitly utilize information on the detailed physics and chemistry that influence ambient air pollution concentrations. Instead, prior knowledge and empirical information are used to select an initial set of variables that may be associated with the outcome—in this case, ozone and $PM_{2.5}$ concentrations. Physical and chemical processes are captured through empirical relationships between the response variable and model inputs. Care must be taken in the implementation and interpretation of these models, however, as the models require a number of assumptions to be met for the models to be appropriate and their outputs reliable. For instance, collinearity in statistical model inputs can lead to regression coefficients that may not reflect physical reality and confound in the results. If care is taken to account for such issues, statistical models serve as a valuable tool for investigating the relationships between multiple variables. In general, they are less computationally and time-intensive to utilize than CTMs and are driven

directly by observations, as opposed to air quality models with uncertain inputs and parameters. A further benefit of not explicitly including physical and chemical parameterizations is that the model may be able to approximate relationships that are not accurately captured in chemical transport models.

Statistical models of ozone have been used to examine how certain meteorological conditions contribute to ozone formation (Bloomfield et al. 1996; Camalier et al. 2007; Henneman et al. 2015; Kuebler et al. 2001), model ozone levels in future climate conditions (Chang et al. 2014), investigate geographic differences in relationships between meteorology and ozone concentrations (Davies and Kelly 1992), and to adjust air quality model outputs to better match observations (e.g., Hu et al. 2014; Porter et al. 2015). Harrington et al. (2012) used monthly-averaged $PM_{2.5}$ concentrations and power plant emissions in a linear regression model to investigate the effects of regulations imposed under the 1990 Clean Air Act Amendments on $PM_{2.5}$ concentrations in the USA.

This work develops detailed statistical models for assessing daily impacts of controls on both ozone and $PM_{2.5}$, and produces counterfactual time series of pollutant concentrations from 1999-2013. The work is unique in relation to others discussed in the introduction in its use of daily emissions and meteorology in statistical models over such a long period of detailed measurements. Daily results are important both for health analyses and for capturing sub-seasonal responses to emissions controls. We apply and compare results from statistical and CTM-derived approaches.

Data and method

Meteorological and ambient air quality measurements

Air quality and meteorological observation datasets used here are described in detail in Henneman et al. (2015), though for the present work the data time span was expanded by 1 year at the beginning and the end to a range of 1999–2013. Ambient concentrations and meteorological data were obtained from the SouthEastern Aerosol Research and Characterization (SEARCH) network's Jefferson Street (JST) monitoring station (33.777 °N, 84.416 °W) in Atlanta, Georgia (Hansen et al. 2003). Hourly data were converted to daily metrics using metric-driven averaging times (Table 1). Maximum daily 8-h average O_3 (MDA8h O_3) and 24-h average $PM_{2.5}$ are used because they are the standard metrics used for regulatory purposes in the USA, and have been widely used in health impact studies (e.g., Pope et al. 2009; Rich et al. 2012; Garcia et al. 2011). JST is located near downtown Atlanta (Fig. 1), and represents urban conditions, which may differ slightly from other locations in the broader Atlanta area. In cases of missing data, meteorological observations from JST were supplemented with measurements from Hartsfield-Jackson International Airport (ATL), which is southwest of the city center. Rainfall data came from the airport monitor.

Emissions from mobile sources and EGUs

Mobile source emissions were modeled using the EPA's MOVES2010b software (USEPA 2012c) for the Atlanta 20-County $PM_{2.5}$ Non-Attainment Area (ANAA—Fig. 1). The ANAA is designated by the US EPA as the area surrounding the city of Atlanta that is in non-attainment of the NAAQS. This area is subject to emissions limits that are more strict than those in the surrounding counties (EPD 2009, 2012). For a discussion of MOVES model setup and inputs, see the supplemental.

MOVES estimates mobile emissions using inputs and internal parameterizations that change month-to-month (e.g., temperature and fuel formulation) and year-to-year (e.g., vehicle population, inspection and maintenance, and vehicle miles traveled). As a consequence, estimated daily emissions often exhibit unrealistic step functions, e.g., between months and years, that should be more gradual. These were corrected using a linear smoothing model that includes linear, squared, and cubed calendar date, weekday/weekend indicators, sine and cosine terms with period of one year, and a time-cosine interaction term (see supplemental).

Total daily NO_x and SO_2 emissions (tons) and load (MWh) from EGUs in the Southeast region (states included: Alabama, Georgia, Mississippi, North Carolina, South Carolina, and Tennessee) were retrieved from the EPA's Air Markets Program Database (USEPA 2013). Under the Acid Rain Program (ARP) beginning in 1995, EPA has required large emitters to report Continuous Emissions Monitoring (CEM) data. This data was separated into two groups: those within the ANAA (Fig. 1) and those outside. Daily emissions from all EGUs in each group were summed to

Table 1 Daily pollutant species used in detrending analysis (2000-2012). Hourly measurements from JST are converted to daily metrics (# days = 4749)

Metric	Averaging description	Period	# Days	% Missing days
$MA8hO_3$	Max of 8h	12am-12am	4681	1.43 %
$PM_{2.5}$	Daily	12am-12am	4015	9.41 %

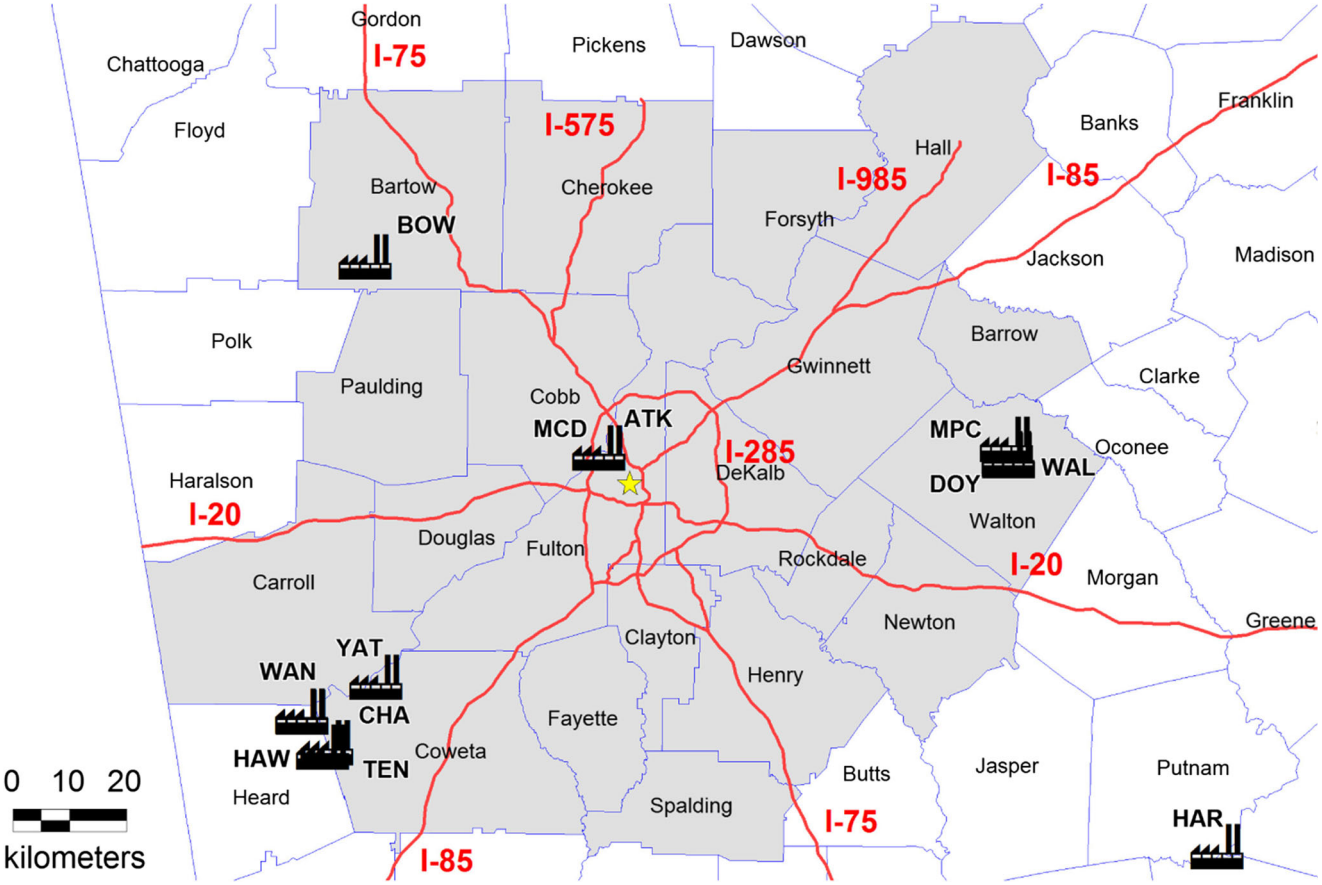

Fig. 1 The 20 county Atlanta $PM_{2.5}$ Non-Attainment Area (shaded). JST is denoted by the star. Power plants are Atkinson (ATK), Bowen (BOW), Chattahoochee Energy Facility (CHA), Doyle Generating Facility (DOY), Harllee Branch (HAR), Hawk Road Energy Facility (HAW), McDonough (MCD), MPC Generating, LLC (MPC), Tenaska Georgi Generating Station (TEN), Walton County Power, LLC (WAL), Wansley (WAN), and Yates (YAT)

represent total EGU emissions. Not all EGUs shown in Fig. 1 were online during the entirety of the study period. The load for the ANAA plants represents the demand on the suite of plants in Fig. 1, and not the demand of the greater Atlanta area, which may be met by importing electricity from other counties or across state lines. Further, electricity may be exported from the area to meet demand elsewhere.

Emissions from different locations within the ANAA have different effects on the measured O_3 and $PM_{2.5}$ concentrations at JST, and these effects vary across days due

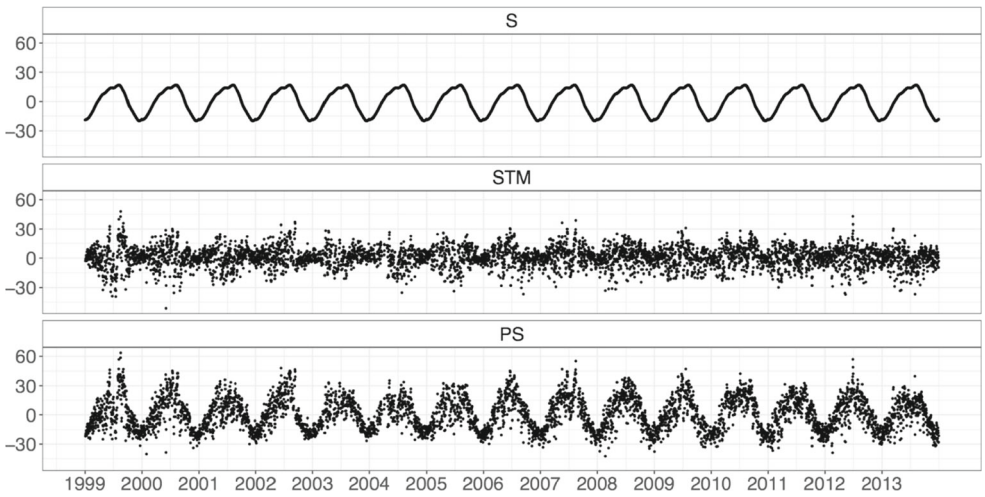

Fig. 2 PS^* (*bottom*) and its two components—seasonal fluctuation (S) and short-term meteorological variation (STM) as calculated by Henneman et al. (2015). Units are *ppb*

to meteorology (e.g., changing wind direction). Sources inside of the ANAA may have different effects on pollutant concentrations in Atlanta than sources that are farther away (Muller et al. 2009) and including regional emissions separately accounts for this.

Empirical estimates of pollutant sensitivities to source emissions

The empirical sensitivity method developed for the present study has two goals: (1) determine the sensitivity of air pollutant concentrations to emissions changes and (2) use these sensitivities to calculate daily air pollutant concentrations for a counterfactual scenario that assumes no new controls were installed after 1993 in mobile sources and 1995 in EGU sources. Sensitivities were calculated using iteratively weighted least squares regression analysis between observed concentrations and estimated emissions for both ozone and $PM_{2.5}$. Statistical analyses in this work were performed using version 3.2.0 of the statistical software R; the regressions were fit with the 'glm' command (R Core Team 2015).

Empirical ozone sensitivities

Covariates for the empirical ozone model were selected based on results from published literature of pollutant sensitivity analyses (e.g., Cohan et al. 2005; Liao et al. 2008; Seinfeld and Pandis 2006; Xing et al. 2011; Blanchard and Hidy 2005). The original list of covariates included NO_x emissions from EGUs (both within the ANAA and regional, denoted reg), and NO_x and VOC emissions from mobile sources, as well as an interaction between these two. Other covariates included NO_x concentration, mean daily windspeed (WS), temperature ($Temp$), and relative humidity (RH), and daily rainfall (RF) as a factor (0−1) variable. All four meteorological variables were centered by subtracting their mean to ease the interpretation of model parameters. The sensitivity of the ozone concentrations to emissions is dependent on the level of photochemical activity, which is often characterized by the ozone level in the atmosphere (indeed, the EPA uses the ozone standard as an indicator for atmospheric photochemical oxidants US EPA 2015a). O_3 serves as a proxy for how much OH is available to oxidize a variety of atmospheric constituents, including those that eventually condense to form secondary PM (such as VOCs for secondary organic aerosols and NO_2 for nitrates). Since ozone is the response in the model, however, raw ozone observations cannot be used as a covariate in the model. To account for this, a measure of emissions-independent atmospheric photochemical oxidative state (PS^*) was applied as an effect modifier with multiple emissions covariates.

PS^* was estimated using components from a meteorological detrending method developed to investigate daily impacts of meteorological fluctuations on pollutant concentrations and described in detail in Henneman et al. (2015). In brief, filtering and linear regressions were used to separate different time scales of fluctuations, including long term (period > 1 year), seasonal (period = 1 year), weekly, and short-term meteorological (period < 3 months) contributions. To calculate PS^*, we summed the seasonal (S, which does not vary between years and is synonymous with annual fluctuation) and short-term meteorological (STM) O_3 trends. S was estimated using a Kolmogorov-Zurbenko filter, a low-pass moving average filter, and averaging the output by date-of-year. STM, an estimate of the impact of daily variability in meteorological variables on ozone, was estimated with a regression of daily fluctuations in multiple metrics: solar radiation (total and daily max) temperature (mean and daily maximum), wind speed (morning and daily means), relative humidity, rainfall, and 1- and 2-day lags of each of these. The sum of S and STM yields a daily metric for the photo-oxidative potential in the atmosphere (Fig. 2). The resulting metric, PS^*, is higher in the summer than the winter, on warmer days, on drier days, and on days with higher wind speed (likely because surface-level NO, which titrates O_3, is carried away from the city on these days). PS^* was centered by subtracting the mean, so that the average contribution to ozone levels is zero, and was used as an interaction between EGU NO_x, mobile NO_x and VOC, and the interaction between mobile NO_x and VOC emissions.

To reduce over-fitting, covariates were removed one at a time from the original list in order of decreasing significance, as measured by the p-values of the parameter error statistics. In general, only regression coefficients significant at the 0.05 level were retained. One exception is the coefficient associated with ANAA EGU NO_x emissions (p = 0.20), which was included in the final model because of its known chemical relevance. Modeled mobile emissions exhibit high co-linearity between emitted species (VOC, CO, NO_x) across the time series. Including multiple species in a statistical model, therefore, presents a problem. Mobile CO emissions contribute to ozone formation (Seinfeld and Pandis 2006), but were excluded from the analysis because of co-linearity with modeled NO_x and VOC emissions, and the chemical action of CO on O_3 formation is similar to that of VOCs. The mobile VOC emissions term was removed because of the co-linearity between modeled mobile NO_x and VOC emissions; however, VOC emissions appear in the model in interaction terms. Model iterations used in the model selection process are provided in the supplemental. The following is the final model:

$$\begin{aligned}[O_3] = {} & \beta_0 + (\beta_1)E^{NO_x}_{EGU} + (\beta_3 + \beta_2 PS^*)E^{NO_x}_{EGU,reg} \\ & + (\beta_4 + \beta_5 PS^* + \beta_6 E^{VOC}_{MOB})E^{NO_x}_{MOB} + \beta_7 WS \\ & + \beta_8 Temp + \beta_9 RH + \beta_{10} RF + \varepsilon_{O_3} \end{aligned} \quad (1)$$

where $[O_3]$ is MD8hO_3, E_j^k is daily emissions, and ε_{O_3} are the model residuals.

Empirical $PM_{2.5}$ sensitivities

The model of $PM_{2.5}$ sensitivities to EGU and mobile emissions is similar to that of ozone. The variable list originally included ANAA and regional EGU NO_x and SO_2 emissions, mobile NO_x, $PM_{2.5}$, SO_2, and VOC emissions, and interaction terms. Each of these emissions terms was included in an interaction with the PS^* as well. Temperature, relative humidity, and the daily temperature and relative humidity-dependent dissociation constant for nitrate (Mozurkewich 1993) were also included. Model selection proceeded for the $PM_{2.5}$ model in a way similar to that for the O_3 model, i.e., covariates were removed one-by-one in order of decreasing significance. The final formulation is:

$$\begin{aligned}[PM_{2.5}] = {} & \beta_0 + (\beta_1 PS^*)E_{EGU}^{NO_x} + (\beta_2 PS^*)E_{EGU,reg}^{NO_x} \\ & + (\beta_3 PS^*)E_{EGU}^{SO_2} + (\beta_4 + \beta_5 PS^*)E_{EGU,reg}^{SO_2} \\ & + (\beta_6)E_{MOB}^{PM_{2.5}} + (\beta_7 PS^*)E_{MOB}^{VOC} + \beta_8 WS \\ & + \beta_9 Temp + \beta_{10} RH + \beta_{11} RF + \varepsilon_{PM_{2.5}} \end{aligned} \quad (2)$$

The statistical models (1 and 2) were used to address the two goals stated previously (to determine the sensitivity of air pollutant concentrations to emissions changes and to use these sensitivities to calculate daily counterfactual concentrations). The models relate observations to emissions on the same day. Multi-day impacts are captured to an extent using the meteorology variables and PS^*, but are difficult to estimate directly because of the correlation between consecutive days in emissions. Sensitivities of pollutants are represented by the βs in each equation. These, when multiplied by their respective covariate, yield the contribution to the concentration by each model input.

Estimated emissions were replaced with counterfactual emissions—described below—to estimate counterfactual concentrations. All other model inputs, including PS^* and ε remain unchanged, since these are independent of emissions. The supplemental includes an assessment of the relationships between model residuals and inputs. Both the O_3 and $PM_{2.5}$ models capture the variability at the middle-and lower quantiles, but the models have some difficulty fully capturing very high observations. These very polluted days are due to factors that are difficult to control for in a statistical model, such as specific combinations of meteorological factors or impacts of wildfire plumes.

Chemical transport model quantification of ozone sensitivities with CMAQ

It is of interest to compare empirical sensitivities based on measured ambient concentrations to alternative source apportionment techniques. The CMAQ model (Byun and Schere 2006) provides a detailed characterization of physics and chemistry governing the transport, removal and formation of air pollutants in the ambient air. In this study, results from a previous study (Liao et al. 2008) and newer results using CMAQ runs with updated parameters and smaller grid size were compared to the results obtained from the statistical O_3 model. CMAQ-DDM version 4.3 with SAPRC-99 chemical mechanism was applied in 2001 on a 36 km grid, and version 5.0.2 with CB05 chemical mechanism was applied on a 12 -km grid. Such models have previously been used to simulate responses (or sensitivities) of ambient O_3 concentrations to changes in emissions of their precursors (Cohan et al. 2005; Dunker 1981; Yang et al. 1997). The 36 km CMAQ run was driven using results from the Fifth-Generation NCAR/Penn State Mesoscale Meteorological Model (MM5) (Grell et al. 1994; Seaman 2000), and the 12 km CMAQ used the Weather Research Forecast (WRF) model version 3.6.1. Values for the 36 km run were modeled for a 2001 climatological year (the meteorology was developed by down-scaling from a climate model), and the 12 km results and empirical values that are plotted are also from this year. Both processed precursor emissions using the Sparse Matrix Operator Kernel Emissions (SMOKE) (Houyoux et al. 2000).

We chose 2001 for the modeling episode both because it came before many of the large changes in emissions and because the we have modeling results for both 36 and 12 km resolutions. Detrending results showed that meteorological variability had a slight negative effect (3.2ppb) on summertime ozone concentrations, and little impact in the winter (Henneman et al. 2015).

CMAQ-DDM directly calculates the semi-normalized first-order (or linear) sensitivities of both gas- and condensed-phase pollutants to precursor emissions (Cohan et al. 2005; Napelenok et al. 2006), i.e., the semi-normalized first-order sensitivity ($S_{i,j}$) of pollutant concentration i (C_i) to source emissions j (E_j) is determined, effectively, as Yang et al. (1997):

$$S_{i,j} = \frac{\delta C_i}{\delta \alpha_j} \quad (3)$$

where α_j is the relative level of the emissions from source j base calculation, and has a nominal value of 1 (Cohan et al. 2005; Hakami et al. 2004). The sensitivities, as presented here, have the same units as the corresponding pollutant concentrations. These sensitivities are local and represent

how pollutant concentrations respond to precursor emission changes if the systems were linear. It is recognized that the system is not linear, but numerous studies suggest the first-order (linear) response is accurate up to domain-wide emission changes of the order of 30–50 % (depending on species) (Cohan et al. 2005; Hakami et al. 2003, 2004). The modeled concentrations and sensitivities for the 36 km run were first published in Liao et al. (2008).

Estimating emissions changes

Estimates of changes in emissions due to regulatory programs must take into account a number of factors that depend on the emission source. For EGUs, population growth, plant efficiency improvements, control installation dates, economic growth and decline, fuel type, etc. must be considered. On the other hand, vehicle fleet age and turnover, fuel type, and population are the important variables to consider for mobile source emissions.

Counterfactual emissions were calculated using a method similar to that used by Gégo et al. (2007). Their method takes into account all controls over the time period while correcting for changes in demand due to population growth or decline, economic trends, etc. An annual emissions rate was calculated for the base year (BY) as the average ratio of daily emissions (tons (US)) to daily load (kilowatt hours—kWh).

$$ER_{BY} = \left\langle \frac{E(d)}{L(d)} \right\rangle_{BY} \tag{4}$$

where L denotes load and d indexes day. ER_{BY} was assumed to remain constant for the counterfactual scenario of no controls. Annual counterfactual emissions were calculated by multiplying each day's load by ER_{BY}:

$$E^{count}(d) = ER_{BY} * L(d, y) \tag{5}$$

where y is each year between BY and 2013. Complete CEM data was available for the ANAA starting in 1995, but data from multiple plants in the region were missing in 1995 and 1996, so 1997 is used as the BY for regional emissions.

This approach takes into account factors that cause changes in the emissions rate, e.g., controls, improvements in transmission efficiency, changing fuel costs that incentivize switching fuel type, etc. The model assumes that the application of controls did not differentially change plant dispatch. Another way to think about this is that the electricity demand in Atlanta is represented by the load carried by all of the plants in the area combined, which may not be the case if electricity is imported from or exported to plants in surrounding regions. This assumption is addressed by including regional emissions as a separate term in each regression equation, however, the limitation means that all deviations from observed concentrations calculated in the counterfactual concentrations below cannot be linked exclusively to specific controls. Comparison of counterfactual concentrations and dates when specific controls were installed allow for interpretation of emissions changes as attributable to specific regulatory actions and controls (details are discussed in Section "Counterfactual emissions").

Counterfactual mobile emissions were estimated using the "Rate of Progress" option in MOVES2010b, which models a scenario with no Clean Air Act Amendments by applying 1993 emission rates to all vehicles after this year (1993 is the default option for this scenario in MOVES) (USEPA 2012b). "Rate of Progress" still uses the same changes in vehicle fleet composition, vehicle miles traveled, and fuel formulations, but assumes 1993 emissions factors for new vehicles.

Results

Counterfactual emissions estimates

Differences between actual and counterfactual EGU NO_x emissions (Fig. 3) show the largest reductions occur in 2002 and 2009, aligning with the beginning of summertime-specific NO_x controls (early 2000s) and a shift to year-round controls (late 2000s). The largest SO_2 emissions reductions occurred in 2009—when two large coal plants in the region had completed the installation of their flu gas desulphurization (FGD) technologies—and 2012—when much of the electricity load in Atlanta switched from coal to natural gas.

Both mobile NO_x and $PM_{2.5}$ emissions (Fig. 3) show decreases between 2000 and 2012, corroborating the findings of Vijayaraghavan et al. (2014). Mobile source SO_2 emissions decrease dramatically (by 80 % between 2004 and 2006) after fuels with reduced sulfur content were required year-round beginning in 2004. Other emitted species (e.g., VOCs, primary PM species) used in this study besides those plotted in Fig. 3 generally follow the trend of NO_x emissions. Counterfactual estimates show slightly increasing NO_x and $PM_{2.5}$ emissions that do not vary much between years and follow the estimated VMT.

Empirical model evaluations

The regression analysis led to a statistical model for O_3 (Table 3) with an R^2 and root mean square error (RMSE) of 0.67 and 11 ppb, respectively. The related values for the $PM_{2.5}$ model (Table 4) are 0.42 and 5.7 $\mu g\, m^{-3}$. Mean observed O_3 and $PM_{2.5}$ over this time period were 41 ppb

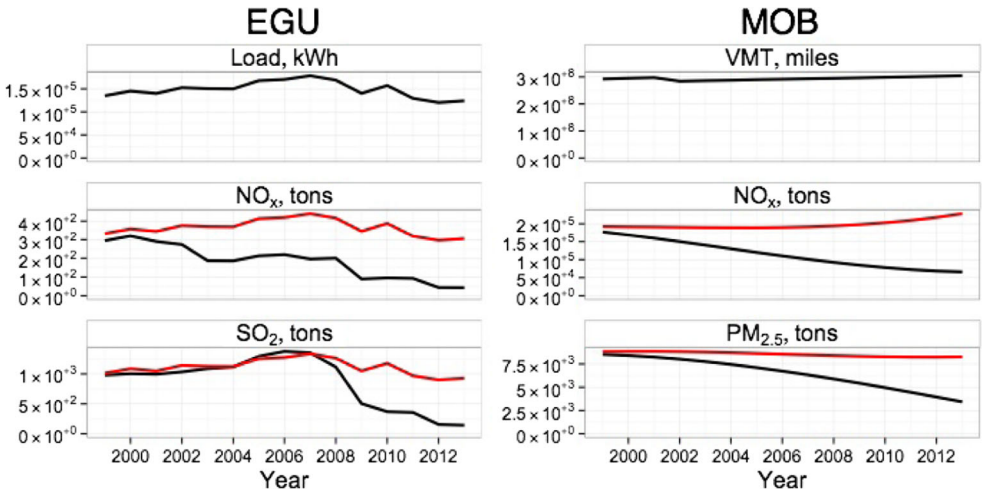

Fig. 3 *Left*, actual (*black*) and counterfactual (*red*) emissions from EGUs. *Right*, actual and counterfactual emissions from mobile sources (MOB). The top graph in each column (load for EGUs and VMT (Vehicle Miles Traveled) for MOB) are the measure used for demand for each source category

and 14 $\mu g\, m^{-3}$, respectively. While the RMSE values are somewhat large, it is more revealing to compare the regression parameter magnitudes to their standard errors. Standard errors are generally small compared to their coefficients in both the O_3 and $PM_{2.5}$ models, which suggests that the model covariates are predictive of changes in the response.

Average contributions (Tables 3 and 4), calculated by multiplying each regression coefficient by the average of the corresponding covariate, are a measure of the relative importance of each term in the regression. Plots of the daily contribution summed by source category (Fig. 4) show that each source contribution varies by seasons. The intercepts for O_3 (42 ppb) and $PM_{2.5}$ (5.0 $\mu g\, m^{-3}$) are estimates of the average background concentrations in Atlanta over the study period that would occur without local and regional emissions from mobile and EGU sources, but would include long-range transport and the impact of other sources, though do not include all nonlinear responses to emissions.

CMAQ-DDM model evaluation

CMAQ-modeled ozone concentrations from each model are from single grid cells that cover downtown Atlanta (including JST). Concentrations from both models exhibit similar annual and daily variability as observed concentrations (Fig. S-6). Evaluation statistics for all days and days with observed O_3 over 60 ppb (Table 2) are somewhat higher than typical statistics reported in the meta-analysis of CTM results published by Simon et al. (2012). However, the current evaluation (i.e., at a single monitor) is stringent in comparison to others, which typically use many monitors at multiple locations. Between the two model runs, the 36-km model was biased higher than the 12 km, and correlation was higher for the 12 km. The overall high bias of the 36-km model improves the results for days over 60 ppb, though the correlation on these days is lower than for the 12 km model.

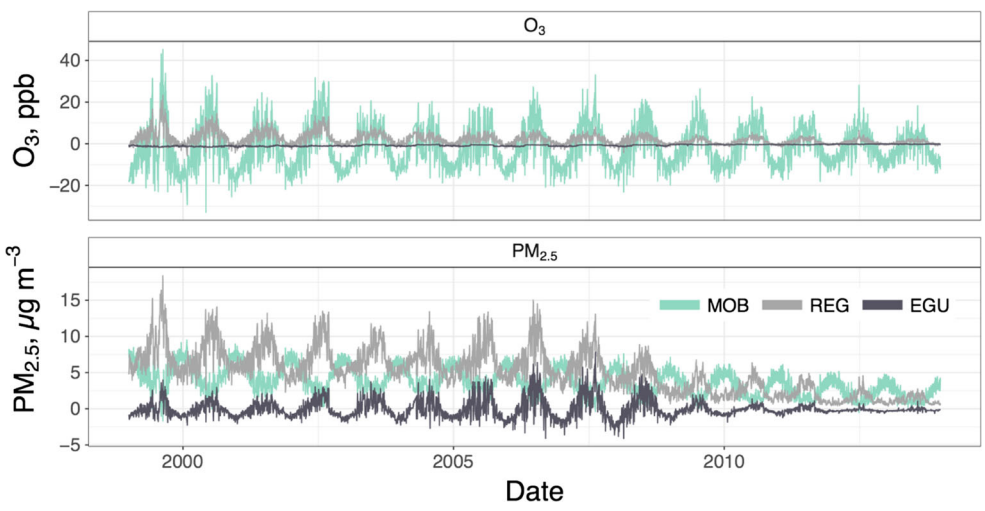

Fig. 4 Sensitivities of daily O_3 (1) and $PM_{2.5}$ (2) concentrations to mobile (MOB), local EGU, and regional EGU (REG) emissions

Table 2 Evaluation statistics for 2 versions of CMAQ, including the number of comparisons (N), normalized mean bias (NMB), normalized mean error (NME), mean bias (MB), and correlation (r)

CMAQ model	*N*	NMB	NME	MB	*r*
36-km	356	38.24	48.78	16.18	0.54
12-km	361	−0.82	27.28	−0.35	0.70
36-km (>60)	80	6.60	23.76	4.79	0.14
12-km (>60)	80	−16.83	19.71	−12.23	0.43

The evaluation is for 2001 for both all available days and days with observed O_3 greater than 60 ppb

Sensitivities of O_3 to EGU and mobile emissions in 2001 peak in the summertime and are negative in the winter and fall (Fig. S-6). In the winter and spring, sensitivities produced by the CMAQ models agree more with each other than the empirical, and, in the summer the 12-km model results agree more with the empirical results. 36-km sensitivities are biased high compared to the other models in the summertime.

While a direct comparison between observed and measured concentrations and sensitivities is important for putting results in perspective with other studies, this study focuses attention in the discussion on the comparison between sensitivities relative to O_3 levels. The goal of this analysis is to assess model intermediates and, relating to model outputs, somewhat reduces the impact of bias and differences in model inputs between model setups.

Counterfactual concentration estimates

Year-specific box plots of the actual and counterfactual ozone time series (Fig. 5) show that median ozone values are relatively insensitive to emissions changes in EGUs and mobile sources (the observed median is 39.5 *ppb* and counterfactual median is 38.8 *ppb*). The bulk of the difference between actual and counterfactual appears in the highest and lowest ozone concentrations. As emissions have decreased, variability in annual ozone distributions has shrunk. Counterfactual emissions would have led to both more low-ozone days (days with MA8hO_3 below 30 *ppb*) and high ozone days. For example, 99 days were observed with MA8hO_3 below 30 *ppb* at JST in 2013, and the counterfactual estimates it would have been 134 days without controls. Four days were observed above 70 ppb (the recently promulgated O_3 NAAQS), whereas the counterfactual predicts 27. For 75 *ppb*, the similar values are one and 16 days, respectively. Differences between observed and counterfactuals are small at the beginning of the time series, and increase as the differences between actual and counterfactual emissions grow.

Discussion

Counterfactual emissions

Differences between actual and counterfactual emissions (Fig. 3) align well with known regulations and the resulting controls. Mobile emissions decrease in a near-linear fashion as old vehicles are replaced with new, cleaner ones, while VMT in the region has grown slowly. EGU emissions change more abruptly as controls are installed, plants change fuel types, and load is shifted between facilities. Information on when controls were installed and new plants were brought online were used to analyze changes in emissions. The information in this section has been taken from information available through EPA's Air Markets Database (USEPA 2013). The discussion focuses on sources within the ANAA (Fig. 1).

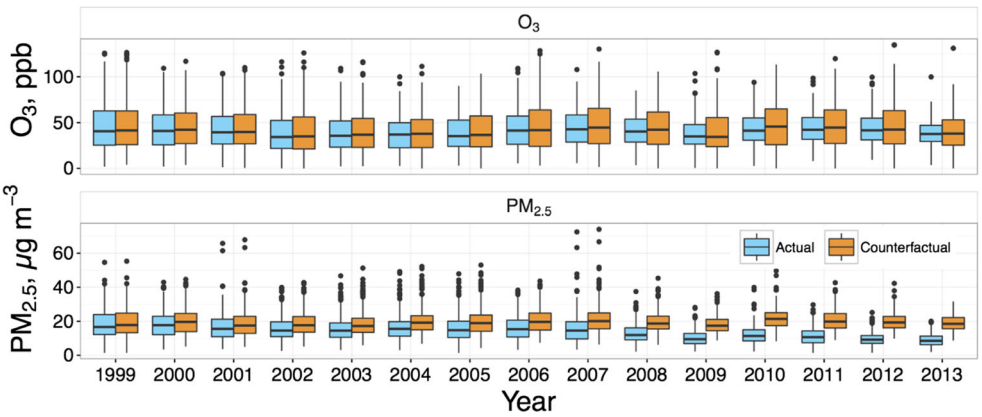

Fig. 5 Annual box plot of the actual and counterfactual ozone and $PM_{2.5}$. *Center lines* are the median, the *boxes* are the first and third quartiles, and *whiskers* extend to values within 1.5 times the interquartile range

Of the 12 EGUs in the ANAA (Fig. 1), some are larger base load plants and others are smaller peaking plants. One plant near downtown Atlanta, McDonough, was converted from coal to natural gas between 2011 and 2012, and another, Wansley, began operation of 10 natural gas units between 2002 and 2012 while still keeping its coal units available. Similarly, all six of the smaller plants that have gone online since 1999 (Chattahoochee Energy Facility, Doyle Generating Facility, Hawk Road Energy Facility, MPC Generating Facility, Tenaska Georgia Generating Station, and Walton County Power Facility) use natural gas. Plant Atkinson, a small plant near downtown, was run on natural gas between 1995 and 2006, when it was retired. The remaining plants (Bowen, Harllee Branch, and Yates) ran primarily on coal between 1995 and 2012. In the ANAA plants, 3 % of the total load was generated by units with natural gas as the primary fuel in 2008 compared with 21 % in 2012 (USEPA 2013), a change that is attributable to both regulations and reduced natural gas prices, and was planned many years in advance (Georgia Power 2007).

The load on the plants increased by 4 % between 1995 and 2012, with a peak in 2007 (Fig. 3). Most plants have added low NO_x burners, and some (Bowen, Chattahoochee Energy Facility, and Wansley) have installed selective catalytic reduction (SCR) NO_x controls. Annual NO_x emissions by the 12 local plants fell by 85 % between 1995 and 2012. At first, regulations led to the SCRs being operated only during the summer months (May-September). Beginning in 2009, NO_x controls on the largest plants in the region were operated year-round.

Years with the greatest increase in the difference between actual and counterfactual EGU NO_x emissions (Fig. 3) are 2002–2003 and 2008–2009. Between 2002 and 2003, plants Bowen and Wansley completed installation of SCR NO_x controls, the Chattahoochee Energy Facility, which was built with SCR technology, went online, and Harllee Branch installed low NO_x burners.

Plants Bowen and Wansley installed FGD technologies on their coal units in 2008 and 2009. These controls, along with the switch to natural gas, have contributed to a decrease in SO_2 emissions by EGUs in Atlanta of 81 % between 1995 and 2012. The years between 2008 and 2010 saw the greatest decrease in SO_2 emissions compared to the counterfactual. Nearly all of this decrease can be traced to the installation of FGD controls at plant Bowen and Wansley. Further reductions were achieved by relying less on coal-fired plants, including the conversion of Plant McDonough from coal to natural gas in 2012.

In Georgia, because the cost of controls cannot be recovered by raising electricity rates unless the control is deemed necessary under existing law, it is assumed that all controls can be attributed to regulatory actions, and these could potentially be future anticipated regulations (Georgia Power 2007). Fuel switches, plant commissioning and retirement schedules, and electricity trading between utilities are governed by complex relationships that include current/projected fuel prices, varying costs of producing electricity at different plants, projected demand, and anticipated future regulatory actions. Therefore, while the estimated emissions reductions are tied to controls, not all emissions changes calculated can be attributed exclusively to regulatory actions.

Magnitudes of EGU and mobile sensitivities

Parameters from the models in Eqs. 1 and 2 (Tables 3 and 4) provide the relative importance of each source-pollutant contribution to concentrations. It is important to recognize that modeled mobile emissions are highly correlated across species. Therefore, it is difficult to separate the effects of all species of interest in a statistical model. For the O_3 model, VOC emissions were included, but only in interactions terms because of this cross-species co-linearity. Mobile source carbon monoxide (CO) emissions, which contribute to O_3 formulation along with VOCs and NO_x, are highly correlated with VOC emissions, so the sensitivity to VOC emissions includes the impacts from CO. Contributions of mobile emissions to total O_3 and $PM_{2.5}$ concentrations are best interpreted as a sum of the component emissions (Fig. 4).

Modeled mobile contributions to ozone concentrations are dominated by the interactions between NO_x emissions and PS^* and NO_x and VOC emissions and PS^*. Average contributions of both of these is zero because the terms are normalized to zero, but the magnitudes of the maxima and minima are large compared to the other terms, i.e., NO_x emissions lead to high ozone on photochemically-active days, but reductions when the meteorology is not conducive to O_3 formation. The negative coefficient on the interaction term that includes VOC emissions and PS^* shows the importance of VOC-limited conditions when increased VOC emissions lead to increases in otherwise low ozone concentrations, i.e., during radical-limited periods there is a positive sensitivity to VOC emissions and negative sensitivity to NO_x emissions. This happens during most of the non-summer season as well as on lower O_3 days during the summer.

For 13 monitors within 74 km of JST, correlations (Pearson R) were at least 0.77 for MDA8h O_3 from 2002-2010, and measurements show consistent annual trends at urban, suburban and rural sites (Fig. S-1). NO_x concentrations, however, show greater spatial variability; therefore, concentration sensitivities to emissions estimated at JST may differ across the region.

Table 3 Regression coefficients summary for Eq. 1 for O_3. Values on the left hand side of the table represent values from the regression, and values on the right hand ride summarize the total empirical sensitivities of ozone to emissions. The regression was performed on 4030 observations

Covariate	Coefficient	Unit	Estimate	Std. Error	Average contribution [ppb]	Minimum contribution [ppb]	Maximum contribution [ppb]
Intercept	β_0	ppb	42	$5x10^{-1}$	42	–	–
$E^{NO_x}_{EGU}$	β_1	$ppb\,ton^{-1}$	$-4.1x10^{-3}$	$3x10^{-3}$	−0.74	−2	0.02
$E^{NO_x}_{EGU,reg}$	β_2	$ppb\,ton^{-1}$	$1.7x10^{-3}$	$8x10^{-4}$	2.1	0.46	5.3
$E^{NO_x}_{EGU,reg} * PS^*$	β_3	ton^{-1}	$1.1x10^{-4}$	$4x10^{-5}$	0.0	−9.7	19
$E^{NO_x}_{MOB}$	β_4	$ppb\,ton^{-1}$	$-4.2x10^{-3}$	$3x10^{-3}$	−1.3	−2.3	−0.41
$E^{NO_x}_{MOB} * PS^*$	β_5	ton^{-1}	$3.4x10^{-3}$	$2x10^{-4}$	0.0	−58	110
$E^{NO_x}_{MOB} * E^{MOB}_{VOC} * PS^*$	β_6	ton^{-2}	$-8.3x10^{-6}$	$1x10^{-6}$	0.0	−66	33
WS	β_7	$^1 ppb \,\|\|m\,s^{-1}\|\|^{-1}$	$-3.3x10^{-1}$	$2x10^{-1}$	0.0	−1.8	0.44
$Temp$	β_8	$^1 ppb \,\|\|°C\|\|^{-1}$	$3.5x10^{0}$	$3x10^{-1}$	0.0	−11	7.5
RH	β_9	$^1 ppb \,\|\|\%\|\|^{-1}$	$-1.7x10^{0}$	$2x10^{-1}$	0.0	−3.9	4.0
RF	β_{10}	$^{1,2} ppb \,\|\|Y\|\|^{-1}$	$8.6x10^{-2}$	$2x10^{-1}$	0.0	−0.06	0.13

[1] $\|\|unit\|\|$ denotes scaled and normalized by subtracting the mean and dividing by standard deviation

[2] Y denotes Rainfall is a factor (1-0) variable

The sensitivities of ozone to emissions (Fig. 4) suggest that mobile emissions have a greater effect on ozone levels at JST than EGU emissions. As annual ozone distributions have shrunk since 2000, the sensitivities have also decreased in magnitude. Tong et al. (2006), Muller et al. (2009) and others have found ground-level NO_x emissions have a much greater impact on ozone concentrations than stack emissions from outside of the city.

All EGU SO_2 emissions terms are of particular interest in the $PM_{2.5}$ model due to the importance of sulfate in Atlanta. The sum of the mean contributions of these terms is 5.2 $\mu g\,m^{-3}$, which corresponds to 36 % of the aver-

Table 4 Regression coefficients summary for Eq. 2 for $PM_{2.5}$. Values on the left hand side of the table represent values from the regression, and values on the right hand side summarize the total empirical sensitivities of $PM_{2.5}$ to emissions. The regression was performed on 3616 observations

Covariate	Coefficient	Unit	Estimate	Std. Error	Average Contribution [$\mu g\,m^{-3}$]	Minimum Contribution [$\mu g\,m^{-3}$]	Maximum Contribution [$\mu g\,m^{-3}$]
Intercept	β_0	$\mu g\,m^{-3}$	5.5	$3x10^{-1}$	5.5	–	–
$E^{NO_x}_{EGU} * PS^*$	β_1	$\mu g\,m^{-3}\,ppb^{-1}\,ton^{-1}$	$-1.9x10^{-4}$	$1x10^{-4}$	0.0	−4.7	2.7
$E^{NO_x}_{EGU,reg} * PS^*$	β_2	$\mu g\,m^{-3}\,ppb^{-1}\,ton^{-1}$	$-5.3x10^{-5}$	$4x10^{-5}$	0.0	−9.4	4.9
$E^{SO_2}_{EGU} * PS^*$	β_3	$\mu g\,m^{-3}\,ppb^{-1}\,ton^{-1}$	$1.1x10^{-4}$	$3x10^{-5}$	0.0	−6.8	9.3
$E^{SO_2}_{EGU,reg}$	β_4	$\mu g\,m^{-3}\,ton^{-1}$	$1.5x10^{-3}$	$1x10^{-4}$	5.2	0.69	11
$E^{SO_2}_{EGU,reg} * PS^*$	β_5	$\mu g\,m^{-3}\,ppb^{-1}\,ton^{-1}$	$4.3x10^{-5}$	$2x10^{-5}$	0.0	−10	17
$E^{PM_{2.5}}_{MOB}$	β_5	$\mu g\,m^{-3}\,ton^{-1}$	$2.1x10^{-1}$	$3x10^{-2}$	3.7	0.81	5.8
$E^{VOC}_{MOB} * PS^*$	β_6	$\mu g\,m^{-3}\,ppb^{-1}\,ton^{-1}$	$4.3x10^{-4}$	$1x10^{-4}$	0.0	−6.4	3.7
WS	β_7	$^1 ppb \,\|\|m\,s^{-1}\|\|^{-1}$	$-1.7x10^{0}$	$1x10^{-1}$	0.0	−9.1	3.6
$Temp$	β_8	$^1 ppb \,\|\|°C\|\|^{-1}$	$1.3x10^{0}$	$2x10^{-1}$	0.0	−3.8	2.7
RH	β_9	$^1 ppb \,\|\|\%\|\|^{-1}$	$-2.0x10^{1}$	$1x10^{-1}$	0.0	−0.45	0.28
RF	β_{10}	$^{1,2} ppb \,\|\|Y\|\|^{-1}$	$-6.5x10^{1}$	$1x10^{-1}$	0.0	−1.0	1.4

[1] $\|\|unit\|\|$ denotes scaled and normalized by subtracting the mean and dividing by standard deviation

[2] Y denotes Rainfall is a factor (1-0) variable

age total $PM_{2.5}$ across all days ($14.2\,\mu g\,m^{-3}$). Over the same time period, sulfate, a secondary particulate species mainly attributable to atmospheric processing of EGU SO_2 emissions, made up 27 % of measured $PM_{2.5}$ at JST. Ammonium particulate matter is strongly associated with sulfate (sulfuric acid will react with ammonia gas), and recent results find that sulfate can enhance biogenic secondary organic aerosol formation (Marais et al. 2016; Weber et al. 2016), explaining when the sensitivity to SO_2 emissions is greater than the measured sulfate.

Mobile sources are estimated to have contributed an average of $3.7\,\mu g\,m^{-3}$ (26 %). The measured species important to the total over this time period are organic carbon (28 %), elemental carbon (11 %), ammonium (11 %), and nitrate (6 %). Each of these remaining species is associated with mobile emissions, and may be a portion of the 26 % they contribute. As in the ozone model, it is most appropriate to interpret the contributions from mobile emissions as the sum of their parts instead of by individual species. While major contributors (primary $PM_{2.5}$ and NO_x) are included in the model, their co-linearity with other species emissions means that the total captured is likely the joint effect of all mobile emissions emissions. For example, primary $PM_{2.5}$ emissions are highly co-linear with VOC emissions (VOC emissions are still included in the model in an interaction with PS^*).

Two terms of interest in the $PM_{2.5}$ model are the interactions between EGU and REG NO_x emissions and PS^*. These terms are negative, meaning that increasing emissions correspond with increasing $PM_{2.5}$ levels in the wintertime, and a negative contribution in the summer. Positive contributions in the winter correspond with increased nitrate levels, a secondary species that forms when NO_x is oxidized to HNO_3, which then reacts with NH_3 to form ammonium nitrate. Brock et al. (2002) showed that young NO_x plumes decrease the conversion of SO_2 to sulfate because of decreased radicals due to NO_x titration, but cautioned that this effect is generally assumed small and uncertain. The current study finds evidence of this phenomenon, and attributes a mean daily reduction of $1\,\mu g\,m^{-3}$ in summers (May-September) across the time period, though ins importance has decreased over time as both NO_x and SO_2 emissions have been reduced. Total $PM_{2.5}$ sensitivities to EGU emissions are highest in the summer, which corresponds with both increased SO_2 emissions and increased photochemical activity that contributes to elevated sulfate concentrations. Mobile sens itivities exhibit much less annual variability than EGUs, and contributions from both source categories have decreased over time as emissions have decreased.

Counterfactual concentrations

The largest changes between observed and counterfactual concentrations occur after 2009, when control programs had at or near their greatest impact on emissions. Between 2010 and 2013, Atlanta experienced many fewer high-O_3 and $PM_{2.5}$ days than it would have without regulations (Table 5). Values for comparison in Table 5 were chosen based on standards of regulatory importance. For example, 12.0 and $15.0\,\mu g\,m^{-3}$ are the primary and secondary annual mean NAAQS for $PM_{2.5}$, and $35\,\mu g\,m^{-3}$ is the primary and secondary 24-hr NAAQS. The O_3 NAAQS was recently changed from 75 ppb to 70 ppb, though the lower end of the proposed range was 60 ppb (U.S. EPA 2014). Results here show that regulatory programs have had important influence on the concentrations of regulatory importance in Atlanta.

The Georgia Department of Natural Resources reports 46 exceedances of the (http://www.air.dnr.state.ga.us/). The highest reported MDA8hr in that year in the ANAA was 139 ppb, which is 20 ppb higher than the highest value measured at JST that year. The maximum MDA8hr value observed at JST is 131 ppb (in the summer of 1998—the year that the station began recording ozone data). There are 4 days in which the counterfactual ozone exceeds 130 ppb, all of which occur in 2007 and later (Fig. 5). The shapes of the annual distributions of counterfactual ozone in the later years resemble those of observed ozone early in the time period. Decreased emissions have had the effect of decreasing median concentrations and distribution widths for $PM_{2.5}$. In 2013, the observed median $PM_{2.5}$ is $8.9\,\mu g\,m^{-3}$, compared with a counterfactual of $18.9\,\mu g\,m^{-3}$. The largest reductions occurred in 2009 and onwards, coinciding with the years of greatest reductions in SO_2 emissions from EGUs.

Table 5 The number of days on which observed and counterfactual O_3 and $PM_{2.5}$ were greater than concentrations of regulatory importance from 2010 to 2013, when regulatory policies were at or near their greatest impact

	Observed	Counterfactual
$O_3 > 60\,ppb$	200	396
$O_3 > 70\,ppb$	75	223
$O_3 > 75\,ppb$	42	164
$PM_{2.5} > 12.0\,\mu g\,m^{-3}$	456	1391
$PM_{2.5} > 15.0\,\mu g\,m^{-3}$	222	1164
$PM_{2.5} > 35\,\mu g\,m^{-3}$	2	7

For both O_3 and $PM_{2.5}$, EGU and REG NO_x sensitivities are positive in the summertime and negative in the wintertime. Therefore, summertime emissions reductions—for instance, NO_x emissions reductions occurring in the mid-2000s—typically reduced concentrations of both pollutants. Winter sensitivities, however, are negative, meaning wintertime NO_x controls, in wider use in the late 2000s, likely increased concentrations. For $PM_{2.5}$, this effect is small compared to the effect of reduced SO_2 emissions.

Comparison to CMAQ-calculated sensitivities

As reported here, CMAQ-modeled ozone sensitivities represent the first order change in ozone expected from a 100 % reduction in anthropogenic emissions. Sensitivities show the contribution of the sources to the measured concentration calculated in the modeled grid cell of interest, which, for this work, is the grid cell that corresponds to JST. CMAQ-modeled sensitivities provide a point of comparison for empirical sensitivities calculated using a different, independent method.

CMAQ-modeled and empirically-calculated sensitivities to all anthropogenic NO_x emissions have a positive relationship with O_3 concentrations (Fig. 6). Empirical sensitivities show more variability and a greater magnitude across the same range than CMAQ-modeled sensitivities from both grid resolutions. From a regulatory perspective, the ozone concentration that corresponds to a sensitivity of zero is of interest—at ozone concentrations above this point, NO_x controls reduce ozone; at concentrations below this point, controls increase ozone. The three models estimate a range of 16.8 *ppb* for this value (42.9 (95 % CI 41.3–44.5), 57.9 (56.5–59.3), and 59.7 (57.2–62.6) *ppb* for empirical, 36 km CMAQ, and 12 km CMAQ, respectively). The slope of the empirical sensitivities (0.63 $ppb\ ppb^{-1}$) is slightly greater than that for CMAQ (0.48 $ppb\ ppb^{-1}$), leading to a closer agreement of the two models at higher O_3 concentrations. The use of a climatologic year will also influence the cross-over points.

Fewer modeled O_3 high (concentration greater than 80 *ppb*) days leads to a closer agreement between the empirical model and the 12 km CMAQ results than the 36 km CMAQ results. Recent evidence has shown that models may overestimate mobile NO_x emissions by as much as 50 % (Anderson et al. 2014; Goldberg et al. 2016; Souri et al. 2016; Travis et al. 2016), which would generally decrease modeled ozone concentrations and sensitivities in the city center. This phenomenon may help explain the general underestimate of sensitivities in the CMAQ results.

Differences between empirical and CMAQ-modeled sensitivities are due, in part, to differences in emissions inputs used in the models. Although the empirical sensitivities are estimated using emissions only in the Atlanta area, long-term regional emissions trends are similar due to

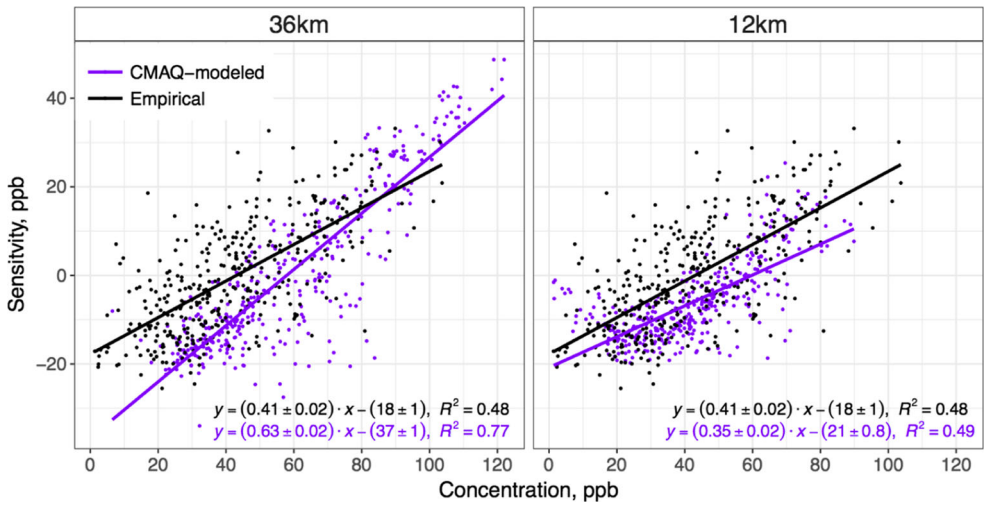

Fig. 6 2001 ozone sensitivities to anthropogenic NO_x emissions calculated by the empirical (1) method and CMAQ (3) for both the 36 km and 12 km runs. Sensitivities greater than zero occur in conditions where NO_x emissions increase ozone, and sensitivities less than zero indicate conditions where NO_x emissions reduce ozone. The x-axis intercepts of the lines are 42.9 (95 % CI 41.3–44.5) for the empirical model, 57.9 (56.5–59.3) for 36 km CMAQ, and 59.7 (57.2–62.6) for 12 km CMAQ. *Numbers in parentheses* in the equations are the standard error of the regression coefficients

national regulatory actions impacting nearby states during similar timeframes. Besides local contributions, empirical sensitivities are impacted by regional emissions, and correlations between the two may impact the results. CMAQ sensitivities are estimated for emissions within the entire modeling domain.

Cohan et al. (2005) investigated the magnitudes of first- and second-order CMAQ-DDM sensitivities of ozone in Atlanta to NO_x and VOC emissions. They found that second-order sensitivities are of similar magnitude to first order sensitivities on high-ozone days, which may further explain the difference between empirical and CMAQ-estimated sensitivities in the present study—CMAQ is used to calculate only first-order sensitivities, while the statistical model captures higher order sensitivities because of the PS^* and nonlinear term.

Method limitations

While this work shows benefits of statistical and numerical air quality modeling, both approaches have limitations. Statistical modeling is subject to bias as a result of correlated input variables, confounding, model selection, and data errors, including estimated emissions. The current study, in particular, is limited by modeled mobile emissions; each of the species is highly correlated with the others, and daily variability is dominated by the approximation of typical weekday/weekend differences. The high level of correlation between species limits the interpretation of ambient sensitivities to mobile emissions—the combined effects from a specific source are more relatable to known physical processes than individual source-species terms in the model.

The use of PS^*, an approximation for emissions-independent photochemical oxidative state, allows for the estimation of emissions-meteorology relationships. However, some meteorological conditions, such as days with wind from specific sources, are not fully captured by this method. Attempts to split daily emissions inputs by wind direction did not improve the predictive ability, and produced results inconsistent with established atmospheric relationships.

Counterfactual emissions assume that changes in electricity production by ANAA EGUs are independent of regulations, and that deviations from the historical emissions-load relationship can be attributed to regulatory actions. Both assumptions are limitations on the method, but comparisons of observed changes in this relationship and regulatory implementation dates provide evidence that the assumptions are reasonable.

Daily counterfactual concentrations, which maintain the autocorrelation of observed ambient concentrations, are limited by the variability in measurements, particularly in the $PM_{2.5}$ model. At the beginning of the time series, observations showed high scatter around the annual mean, which the statistical model is proficient at estimating. The scatter is smaller towards the end of the time series, therefore, counterfactual estimates in the later years lack the very high days observed in the early part of the time series (Fig. 5). Further, in Atlanta, high $PM_{2.5}$ days can be associated with wild land fires, which are not studies here.

Conclusion

We presented a detailed accountability assessment of regulatory impacts on O_3 and $PM_{2.5}$ concentrations in Atlanta, GA between 1999 and 2013. The atypical approach addressed challenges typical in accountability programs. Applying emissions factors to create counterfactual concentration and relating these to a detailed regulatory assessment increased confidence in the changes attributed to regulations, even as regulations were implemented incrementally over time.

The empirical method that employs statistical modeling to develop daily sensitivities and counterfactuals of measured pollutants to changes in emissions. Empirical relationships in the model are reliant on PS^*, a daily metric for the photochemical state of the atmosphere that varies with temperature, relative humidity, wind speed, and rainfall. The model was applied to a central monitoring site in Atlanta, GA using estimated EGU and mobile emissions from the entire metro Atlanta area. Empirical sensitivities agree with sensitivities estimated using CMAQ-DDM, a CTM that explicitly accounts for atmospheric chemistry and physics. The comparison between model intermediates across platforms provides evidence that the empirical relationships appropriately capture emissions relationships with observed ambient concentrations.

For this monitoring location, sensitivities of ozone to mobile NO_x and VOC emissions dominate those of EGU NO_x emissions in magnitude, a result that may be different in locations outside of the city center. Minimum and maximum annual contributions of both mobile and EGU emissions have decreased over time with the implementation of controls.

Mobile emissions dominate the contribution to $PM_{2.5}$ concentrations in the winter, and EGU SO_2 emissions dominate in the summer. EGU contributions to measured $PM_{2.5}$ concentrations show a large seasonal pattern, show a large seasonal pattern, which has decreased significantly with the implementation of SO_2 emissions controls. Average EGU contributions are similar to average sulfate concentrations in Atlanta, and regional and local EGU's contribute similar amounts to sulfate on average.

Statistical models of this type are relatively straightforward to apply, and daily counterfactuals estimated using these models are appropriate for use in accountability studies investigating public health responses to air quality regulations. This approach could give policy-makers a ready estimate of impacts of past or proposed controls. Then, in areas with the greatest interest (or to investigate specific effects or certain controls), policy-makers could employ more sophisticated CTMs to model changes.

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