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Meteorological detrending of primary and secondary pollutant concentrations: Method application and evaluation using long-term (2000–2012) data in Atlanta

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HIGHLIGHTS

- Comparison of meteorological detrending methods in measured data in Atlanta, GA.
- Quantification of variability in air pollution concentrations linked to meteorology.
- Assessment of air pollution reductions linked to specific controls in Atlanta.
- Controls linked to reductions in primary pollutants and $PM_{2.5}$.
- Controls linked to a narrowing ozone distribution and little change in the median.

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ABSTRACT

The effectiveness of air pollution regulations and controls are evaluated based on measured air pollutant concentrations. Air pollution levels, however, are highly sensitive to both emissions and meteorological fluctuations. Therefore, an assessment of the change in air pollutant levels due to emissions controls must account for these meteorological fluctuations. Two empirical methods to quantify the impact of meteorology on pollutant levels are discussed and applied to the 13-year time period between 2000 and 2012 in Atlanta, GA. The methods employ Kolmogorov–Zurbenko filters and linear regressions to detrended pollutant signals into long-term, seasonal, weekly, short-term, and white-noise components. The methods differ in how changes in weekly and holiday emissions are accounted for. Both can provide meteorological adjustments on a daily basis for future use in acute health analyses.

The meteorological impact on daily signals of ozone, NO_x, CO, SO_2, $PM_{2.5}$, and PM species are quantified. Analyses show that the substantial decreases in seasonal averages of NO_x and SO_2 correspond with controls implemented in the metropolitan Atlanta area. Detrending allows for the impacts of some controls to be observed with averaging times of as little as 3 months. Annual average concentrations of NO_x, SO_2, and CO have all fallen by at least 50% since 2000. Reductions in NO_x levels, however, do not lead to uniform reductions in ozone. While average detrended summer average maximum daily average 8 h ozone (MDA8h O_3) levels fell by 4% (2.2 ± 2 ppb) between 2000 and 2012, winter averages have increased by 12% (3.8 ± 1.4 ppb), providing further evidence that high ozone levels are NO_x-limited and lower ozone concentrations are NO_x-inhibited. High ozone days (with MDA8h O_3 greater than 60 ppb) decreased both in number and in magnitude over the study period.

1. Introduction

Policy makers design regulations to reduce the adverse impacts of air pollutant emissions on human health and the environment. The effectiveness of the regulations are assessed based on measured pollutant concentrations and estimates of impacts on

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health endpoints related to exposure to these pollutant concentrations (van Erp et al., 2008). However, given the number of factors that affect a measured air pollution signal (*e.g.* meteorological fluctuations, climate change, population growth, weekend/weekday emissions differences, and the disproportionate effect of sources close to the monitor), it is often difficult to identify and quantify the effects of specific controls in long-term air pollution records. The meteorological impact on ozone and particulate matter with diameter less than 2.5 μm ($PM_{2.5}$) levels is especially important because of their known associations with negative health outcomes and the fact that ambient concentrations of both ozone and certain components of $PM_{2.5}$ are secondary, *i.e.* their formation in the atmosphere is dependent on both meteorological and chemical processes. Although emissions of precursors to ozone (*e.g.* NO_x and volatile organic compounds—VOCs) and $PM_{2.5}$ (*e.g.* primary $PM_{2.5}$, NO_x, SO_2, NH_3, and VOC's) have decreased under programs implemented under the Clean Air Act since 1970, ozone and $PM_{2.5}$ levels continue to exceed EPA standards (Pachon et al., 2012; Vijayaraghavan et al., 2014). In order to assess the impact of controls on measured pollutant signals, it is important to account for the impact of variations in meteorology (Cox and Chu, 1993; Rao and Zurbenko, 1994).

The years between 2000 and 2012 saw the implementation of a number of regulations and associated controls on emissions across the United States. Major programs implemented during this period include the Tier 2 Vehicle and Gasoline Sulfur Program (phased in 2004–2009), the 2007 Heavy-Duty Highway Rule, the Acid Rain Program and associated NO_x State Implementation Plan (SIP) Call and Budget Trading Programs (phased in 1995–2010), and the Clean Air Interstate Rule (CAIR—phased in beginning in 2009) (Hubbell et al., 2009; Morgenstern et al., 2012; USEPA, 1999; USEPA, 2000; USEPA, 2009). In order to better assess the effectiveness of these regulations it is necessary to investigate the primary pollutants whose emissions are affected by the controls directly, the secondary pollutants that are products of atmospheric reactions of the primary pollutants (*e.g.* O_3 and $PM_{2.5}$), and the role that meteorological fluctuations play in affecting trends. To this end, meteorological effects can be quantified and removed from the measured pollutant signal.

A number of methods have been used to remove meteorological impacts on ambient pollutant concentrations (Cox and Chu, 1993; Flaum et al., 1996; Gardner and Dorling, 2000; Kuebler et al., 2001; Rao and Zurbenko, 1994). Rao and Zurbenko (1994) used multiple-pass moving average filters of different averaging windows and varying numbers of passes to isolate short-term fluctuations in ozone concentrations, and found a lag between temperature and ozone. Flaum et al. (1996) extended the analysis further to include dew point temperature, specific humidity, and wind speed. Kuebler et al. (2001) employed a similar filter and regression technique as to Rao and Zurbenko (1994), but also included effects of solar radiation.

Others have used approaches besides filtering and regression techniques to isolate meteorological contributions. Cox and Chu (1993) used data from 43 urban areas to create a distribution of ozone concentrations in urban areas, and accounted for both surface temperature and wind speed effects. Gardner and Dorling (2000) used neural networks and filtering to calculate meteorology-adjusted ozone time series. In general, the neural network method finds a greater contribution of meteorology to ozone concentrations than the filter-regression methods. Brönnimann and Neu (1997) used weekly emissions trends derived from the Swiss emissions inventory and characterized meteorological conditions as “favorable” and “unfavorable” in order to determine concentration levels that can be expected on different days given certain meteorological conditions.

Camalier et al. (2007) used generalized linear models (GLMs) to quantify annualized effects of meteorology on ozone levels in cities across the United States. This method was extended by Blanchard et al. (2010) to investigate the effects of changing levels of ozone precursors on ozone levels in Atlanta and the reason for the relatively small change in ozone levels compared to its precursors. The authors found that daily ozone levels are more sensitive to meteorological fluctuations than to changes in ambient non-methane organic carbon and NO_y.

Within the context of an accountability assessment of how regulatory policies affect acute (daily) health outcomes, it is important to estimate meteorological impacts on observed daily concentrations for use in acute health impact analyses. The methods presented here assess over a decade of observations of multiple gaseous and particulate constituents to aid the assessment of regulatory actions on public health, independent of daily meteorological fluctuations.

This paper presents results obtained using meteorological detrending methods used by Kuebler et al. (2001) and a new method designed to improve the ability to isolate only the fluctuations attributed to meteorological variability in a daily concentration signal. The new method uses multiple Kolmogorov–Zurbenko filters to account for the long-term and annual fluctuations, and a multi-linear regression with meteorological terms, interaction terms, monthly terms, and weekday and holiday terms. The methods are applied to ozone, NO_x, SO_2, CO, total $PM_{2.5}$, SO_4^{2-}, organic carbon (OC) and elemental carbon (EC) measured in Atlanta, GA from 2000 to 2012.

2. Data

Air quality and meteorological data, excluding rainfall, are from the SEARCH network's Jefferson Street monitoring station (JST) in downtown Atlanta (Atmospheric Research and Analysis, 2014). Rainfall measurements come from NOAA's National Climatic Data Center station at Hartsfield–Jackson Airport, which is 15 km south of the Jefferson Street Station.

Eleven pollutant species and eight meteorological metrics are used in this study (Tables 1 and 2). The meteorological variables are chosen as those likely having a strong physical–chemical link to the pollutant species. Daily metrics of three gas phase species (ozone, NO_x, and CO) are calculated in two ways each—the daily 8-h and 1-h maxima ozone and daily and morning means of NO_x and CO. Morning means capture commute emissions, while daily means reduce the impact of high concentrations seen due to shallow boundary layer height expected in the early morning (Kuebler et al., 2001). Most meteorological and species records had less than 10% missing days with the exceptions of OC and EC, which switched from being measured every day to 1-in-3 day measurements in 2009. Hourly measurements of incoming solar radiation, temperature, wind speed, and relative humidity were converted to daily metrics (Table 2). Rainfall was used as a binary input.

3. Methods

Observed pollutant species concentrations are a nonlinear function f^* of emissions (E) and meteorology (M), *i.e.*

$$C(t) = f^*[E(t), M(t)] \quad (1)$$

Results from Blanchard et al. (2010) suggest a near–linear relationship between ozone and temperature and relative humidity, and nonlinear relationships for other meteorological variables (Blanchard et al., 2010). Recognizing that most variability in pollutant concentrations is on a seasonal scale, and emissions and

Table 1
Daily pollutant species used in detrending analysis (2000–2012). Hourly measurements from JST are converted to daily metrics (# days = 4745).

Species[a]	Metric	Period	Exclusion criteria[b] (hours)	# Days	% Missing days
$MD8hO_3$	Max of 8-h Mean	12 am–11 pm	–	4681	1.43%
O_3^{Ma}	Daily Max	12 am–11 pm	$\geq 12/24$	4627	2.57%
NO_x	Daily Mean	11 am–7 pm	$\geq 5/9$	4447	6.36%
NO_x^{morn}	Morning Mean	8 am–11 am	$\geq 3/4$	4398	7.39%
CO	Daily Mean	11 am–7 pm	$\geq 5/9$	4609	2.95%
CO^{morn}	Morning Mean	8 am–11 am	$\geq 3/4$	4623	2.65%
SO_2	Daily Max	12 am–11 pm	$\geq 12/24$	4658	1.92%
$PM_{2.5}$	Daily Mean	12 am-11 pm	–	4015	9.41%
SO_4^{2-}	Daily Mean	12 am–11 pm	$\geq 12/24$	4015	1.92%
OC	Daily Mean	12 am–11 pm	–	3384	28.7%
EC	Daily Mean	12 am–11 pm	–	3384	28.7%

[a] The superscript *M* for pollutant abbreviations in the left column signifies the daily maximum, the superscript *morn* signifies a morning mean, and metrics with no superscripts are the average of hourly values within the Period.
[b] If the Exclusion Criterion for each averaging/maximum period is violated (*i.e.* there are more hours missing than allowed), the day is counted as NA. If there is no Exclusion Criterion, the minimum number of measurements needed for that day is one.

their effects on measured pollutant species are primarily cyclical (dominated by daily, weekly, and annual patterns), their contributions to a daily signal can be removed along with the background. The residual fluctuations in the signal can then be attributed to short-term meteorological fluctuations through a multilinear regression. Short-term meteorological fluctuations are assumed to be unaffected by and independent of emissions.

The long-term time series of daily pollutant concentrations (C) can be decomposed into components (Kuebler et al., 2001; Rao and Zurbenko, 1994):

$$ln[C(t)] = C^{LT}(t) + C^{S}(t) + C^{W}(t) + C^{STM}(t) + C^{WN}(t) \quad (2)$$

or

$$C(t) = \exp\left[C^{LT}(t) + C^{S}(t) + C^{W}(t) + C^{STM}(t) + C^{WN}(t)\right] \quad (3)$$

Ozone concentrations in the atmosphere typically approximate a log-normal distribution (Hogrefe and Rao, 2000; Rao et al., 1997). A log transformation is applied prior to detrending for all pollutants to ensure model residuals follow the assumptions of normal distribution and homoscedasticity. A discussion of model residuals and comparing log- and no-log models is included in the Supporting information.

The components of each signal in (2) are defined as follows (time period in parentheses):

LT: long-term (>365 days)
S: seasonal (365 days)
WH: weekly-holiday (7–365 days)
STM: short-term meteorological (<365 days)
WN: white-noise (1 day)

The *LT* component in the species decomposition captures long-term changes in background concentrations, climate, and precursor emissions (in secondary pollutants) (Gardner and Dorling, 2000; Rao and Zurbenko, 1994). While the *WH*, *STM*, and *WN* signals cover overlapping periods, they are each independent. The goal of the detrending is to quantify and remove the *STM* signal and relate it to meteorological fluctuations so that the remaining daily signal is corrected for short term meteorological fluctuations, *i.e.*, deviations from the typical meteorology for the time of year.

A meteorological signal can be decomposed in a similar way as a pollutant species signal, though a log transform is not used for the meteorological variables. A log transform is not necessary because meteorological variables are the covariates in regression models described below, and there are no assumptions of specific distributions that need to be met. The short-term deviations of meteorological signals, ΔM, occur on time periods of less than those accounted for by the long-term and seasonal trends.

$$M(t) = LT^{met}(t) + S^{met}(t) + \Delta M(t) \quad (4)$$

Two methods are assessed for their ability to separate daily deviations from long-term fluctuations. Both methods quantify the effect of individual meteorological variables on species signals. They are designed to create daily adjustments for meteorological contributions to pollutant signals, and account for holiday and weekday effects that vary throughout the year as well as nonlinear effects of temperature and relative humidity on daily pollutant

Table 2
Daily weather variables used in detrending analysis (2000–2012). Hourly measurements from JST are then converted to daily metrics (# days = 4745). Rainfall (RF) is the exception: measurements are from NOAAs National Climatic Data Center station at Hartsfield–Jackson Airport, which is 10 miles south of the Jefferson Street Station.

Species[a]	Metric	Period	Exclusion criteria[b] (hours)	# Days	% Missing days
SR	Total Daily	12 am–11 pm	Any, 7 am–6 pm[c]	4289	9.69%
SR^M	Daily Max	12 am–11 pm	–	4689	1.26%
Tm	Daily Mean	11 am–3 pm	$\geq 3/5$	4664	1.79%
TM	Daily Max	12 am–11 pm	–	4717	0.68%
WS	Daily Mean	11 am–3 pm	$\geq 3/5$	4580	3.56%
WS^{morn}	Morning Mean	7 am–10 am	$\geq 3/4$	4580	3.94%
RH	Morning Mean	8 am–11 am	$\geq 3/4$	4678	1.50%
RF	Daily Factor	12 am–11 pm	–	4745	0%

[a] SR—solar radiation, T—temperature, WS—wind speed, RH—relative humidity, and RF—rainfall. Superscripts *M* and *morn* signify the daily maximum and morning mean. Metrics with no superscripts are the average of hourly values within the Period.
[b] If the Exclusion Criterion for each averaging/maximum period is violated (*i.e.* there are more hours missing than allowed), the day is counted as NA. If there is no Exclusion Criterion, the minimum number of measurements needed for that day is one.
[c] If any solar radiation values are missing during the daytime hours, the day is excluded.

concentrations. While the results reported here are for data from Atlanta, the methods can be applied to any monitoring station with a multi-year meteorological and ambient air pollution record.

3.1. Multiple KZ and Kuebler et al. (2001) methods

The Multiple KZ (MKZ) and Kuebler et al. (2001) (KEA) methods are approaches to perform the detrending described above. Both use the Kolmogorov–Zurbenko (KZ) filter, which is a low-pass moving average filter (Kuebler et al., 2001; Rao and Zurbenko, 1994; Zurbenko, 1991). Zurbenko (1991) defined a $KZ_{m,p}$ as

$$Y_i = \frac{1}{m}\sum_{j=-k}^{k} X_{i+j} \tag{5}$$

where $m = 2k + 1$ (Zurbenko, 1991). m defines the window length, p is the number of passes, and i is the day of interest. KZ filters are widely used for their adaptability to different cutoff frequencies and robustness to missing data (Eskridge et al., 1997). Rao and Zurbenko (Rao and Zurbenko, 1994) investigated different values of m and p depending on the desired cutoff frequency. The filtering in this work was done using version 3.0.0 of the 'kza' filter package in the statistical program R (Close and Zurbenko, 2011; R Core Team, 2012). A benefit of the KZ filter as implemented in R is that it can handle missing data.

Both the MKZ and KEA methods employ KZ filters and linear regressions to separate the long-term signals of daily concentrations into the components listed above. The methods are described in detail in the Supporting information, however, key differences are described here. The models take the forms:

$$ln[C_{MKZ}(t)] = \underline{\widehat{C}}^{LT}_{MKZ}(t) + \underline{\underline{\widehat{C}}}^{S}_{MKZ}(t) + \dot{\widehat{C}}^{WH}_{MKZ}(t) + \dot{\widehat{C}}^{STM}_{MKZ}(t) + \dot{\widehat{C}}^{WN}_{MKZ}(t) \tag{6}$$

$$ln[C_{KEA}(t)] = \underline{\widehat{C}}^{LT}_{KEA}(t) + \underline{\underline{\widehat{C}}}^{S}_{KEA}(t) + \dot{\widehat{C}}^{WH}_{KEA}(t) + \dot{\widehat{C}}^{STM}_{KEA}(t) + \dot{\widehat{C}}^{WN}_{KEA}(t) \tag{7}$$

The hats indicate an estimated value. In both methods, a $KZ_{365,3}$ filter (denoted by the single underlines in (6) and (7)) is used to remove the *LT* signal. The averaged output—on a day-of-year basis—from a second filter, $KZ_{15,5}$ (double underline), represents the *S* component in the MKZ method. The method is named for the use of multiple KZ filters to separate the detrending components. For the KEA method, *S* is calculated by averaging the resulting signal after removing *LT* by date of year, which takes into account holidays that occur on a different date each year (*e.g.* Thanksgiving and Memorial Day—tilde underline). A result of this averaging is that the weekly signal, *WH*, is combined with the *S* signal in the KEA method.

LT and *S* are subtracted from the observations in the MKZ method, and *LT*, *S*, and *WH* are subtracted from the observations in the KEA method. The remaining signals (*WH*, *STM* and *WN* in the MKZ method and *STM* and *WN* in the KEA method) are estimated in regressions (single dots underneath variables in the MKZ formulas, double dots in the KEA). The covariates included in the MKZ regression are (see Table 2 for variable definitions): ΔT^m, $(\Delta T^m)^2$, $(\Delta T^m)^3$, ΔWS, ΔRH, RF, $T^{M*}\Delta T^m$, $T^{M*}\Delta RH$, ΔT^m_{-1}, ΔWS_{-1}, ΔRH_{-1}, RF_{-1}, ΔT^m_{-2}, ΔWS_{-2}, ΔRH_{-2}, RF_{-2}, weekday indicators, weekday indicators times maximum temperature, month indicators, and holiday indicators. Subscripts represent number of lagged days. In the KEA regression, only the meteorology covariates are used (*i.e.* none of the indicator variables). Values are provided for the full set of variables are described in the supporting information, and for the regression coefficients in Tables S1 and S2 in the Supplemental Material. The regressions are different in that weekday and holiday effects are modeled explicitly in the MKZ method, while they are not in the KEA method, and $\Delta M(t)$ are estimated slightly differently (see supplementary material).

4. Results

4.1. Method evaluation and comparison

Correlations (Table 3) show that the MKZ model captures as much or more of the observed variability than the KEA method for all but one of the pollutants (SO_4^{2-}). Most pollutants have slopes and R^2 values of greater than 0.40 across all models. SO_2 is the only exception—neither of the models produce an R^2 greater than 0.30 for SO_2. The concentration of SO_2 that is measured at JST is highly dependent on an electricity generating unit (EGU) plume passing over the monitoring site. This is especially true since most mobile emissions of SO_2 decreased dramatically after rules came into effect that reduced the allowable sulfur content in gasoline and diesel beginning in 1999. This has in turn reduced the variability in the SO_2 signal over time, leading to a problem with fit in both methods.

A holdout analysis is a type of cross-validation used to investigate the robustness of a model to missing data. 30 holdout tests were performed by training the model on the data with 10% of the observations removed. The sampling was performed independently for each holdout test, meaning it is possible data points were held out for multiple tests. For each test, the remaining 90% of the data was used to estimate each component of the detrending (*i.e. LT*, *S*, *WH*, *STM*, and *WN*). The *LT* and *S* components are estimated using the KZ filter, which can handle missing data, and still assigns an estimate to missing days. In the KEA method, the *WH* component is estimated along with *S*, and *STM* is estimated using the best fit of the linear model. In the MKZ method, the *WH* and *STM* components are both estimated using fits of a linear regression. The linear regressions are fit using the 90% remaining observations, and regression parameters are used to estimate the withheld 10%. The predicted values are compared with observed using the root mean square deviation (Table S2 in Supplementary Information).

The average root mean square deviation (RMSD) was lower for 7 of the 11 pollutant metrics for the MKZ method, and for only one (*OC*) in the KEA method (p-value < 0.05). Differences in the rest of the pollutants were not statistically significant. The RMSD for each species describes the *WN* component of the detrending, so a smaller RMSD means more of the variability is captured by the model.

Monitor design values are used to assess attainment under the National Ambient Air Quality Standards (NAAQS). For ozone, the

Table 3
Slopes, correlation coefficients (R^2 values—in parentheses), and root mean square deviations (RMSD) for the fit of the KEA and MKZ methods.

	Slope [*ppb ppb*$^{-1}$]		R^2		RMSD [*ppb*]	
	KEA	MKZ	KEA	MKZ	KEA	MKZ
O_3	0.72	0.71	0.62	0.66	11.3	10.7
O_3^M	0.72	0.72	0.67	0.71	11.7	11.0
NO_x	0.52	0.53	0.58	0.64	9.60	9.11
NO_x^{morn}	0.33	0.37	0.44	0.50	36.1	34.4
CO	0.44	0.44	0.53	0.53	73.9	73.9
CO^{morn}	0.28	0.29	0.44	0.46	274	271
SO_2	0.28	0.21	0.19	0.23	13.2	12.9
$PM_{2.5}$	0.46	0.47	0.49	0.49	4.85	4.82
SO_4^{2-}	0.41	0.38	0.50	0.48	1.92	1.97
OC	0.38	0.38	0.43	0.45	1.37	1.36
EC	0.38	0.39	0.45	0.48	0.57	0.56

design value is defined as the three-year average of the annual fourth highest MDA8h ozone. For $PM_{2.5}$, the design value is defined as the three-year average of the fourth highest 24hr average $PM_{2.5}$. The observed design values for O_3 plotted in Fig. 2 show a decreasing trend. After detrending with both the KEA and MKZ methods, however, design values show a greater decreasing slope. Results from the two models are similar, and provide evidence that meteorological fluctuations contributed to higher design values in most of the years in the current study. While meteorology can both increase and suppress ozone levels, most of the highest days (which contribute to the design value) are on hot summer days with meteorological conditions that are conducive to ozone formation. When this effect is removed in the detrending, the design values decrease (Fig. 2).

Design values for detrended $PM_{2.5}$ from 2003 to 2007 are less than the observed. For the other years, both detrending methods are similar to the observed.

Results from both models show that average annual ozone concentrations did not change much from 2000 to 2012 in Atlanta (Fig. 3), although design values of both ozone and $PM_{2.5}$ have decreased substantially (Fig. 2). Results of the models are similar based on the R^2 values (Table 3) and the ability to calculate daily adjustments. Because of the higher R^2 values and results of the holdout analysis, the MKZ method was identified as the most appropriate model. Therefore, a majority of the following discussion addresses results are from this model.

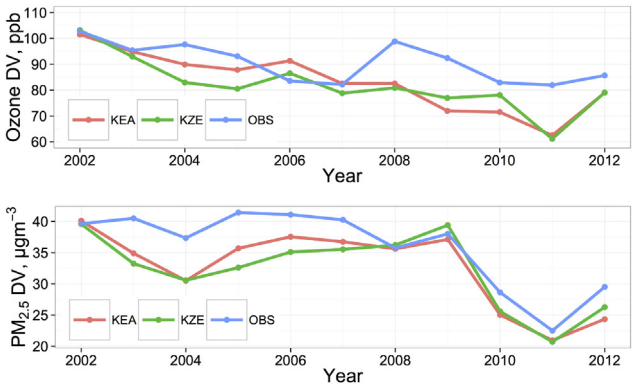

Fig. 2. Monitor design values for ozone and $PM_{2.5}$. For ozone, the design value is defined as the three-year average of the annual fourth highest MDA8h ozone. For $PM_{2.5}$, the design value is defined as the three-year average of the fourth highest 24-hr average $PM_{2.5}$.

4.2. Effect of meteorology on ozone levels

The magnitude of the daily meteorological contribution to ozone levels are specific to different climate regions (Camalier et al., 2007; Gégo et al., 2007; Rao et al., 1995), and, for Atlanta, approximate a normal distribution (Fig. S1a in Supplementary Material). Fig. S1b in Supplementary Material displays the monthly means and standard

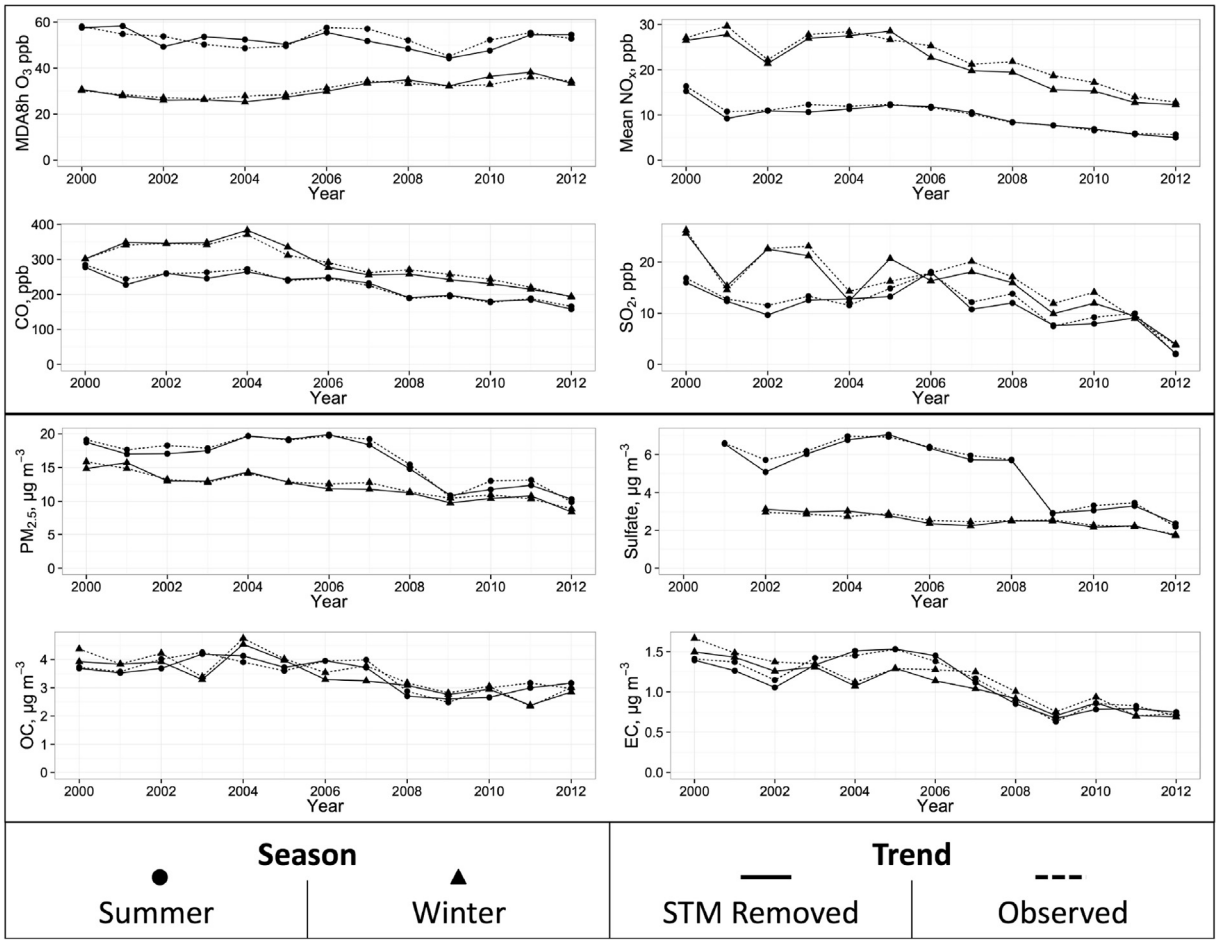

Fig. 1. The raw and meteorologically-detrended summer and winter means. Winter averages are taken from January 1st to March 31st, and Summer averages are taken from July 1st to September 30th. Gaseous species are above and PM species are below.

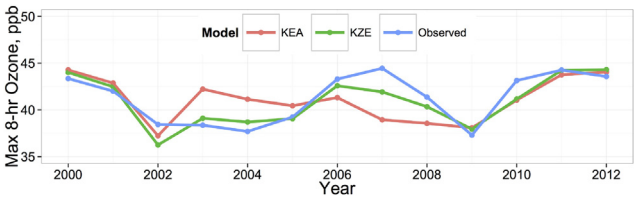

Fig. 3. The annual average adjusted max of 8-h max ozone levels for Atlanta for the KEA and MKZ, methods compared with the observed.

deviations of meteorological contribution to ozone levels. In general, meteorological contributions increase in the summer and decrease in the winter. However, the width of the distribution also changes, as the summer sees a wider range of effects than the winter. Two exceptions to this trend are May and July, which both average a near-zero effect and have standard deviations that are smaller than April, June, and August. The cause of this may be traced to the negative association of relative humidity with ozone.

The average contribution of short-term meteorology is 0.9 ppb (near zero because positive and negative contributions balance each other out over the length of the 13-year time series), but the model finds contributions as low as −57.2 ppb and as high as 44.0 ppb (Table 4). The mean absolute contribution of daily short-term meteorology is 8.4 ppb. Days with high and low relative humidity show opposite average effects on ozone levels. Though the distributions of high and low relative humidity days have different means, the standard deviations are similar. Therefore, changes in temperature yield a widening of the response distribution, and changes in relative humidity yield a change in the mean direction of the response distribution.

4.3. Meteorologically detrended species metrics

Meteorologically detrended and raw summer and winter means have decreased in Atlanta for all pollutants in this analysis over the study period (Fig. 1). The MKZ detrending process has the effect of reducing the means and variances in the observations of all pollutants, though changes in the variances are more substantial than changes in the mean (Fig. S2 in Supplemental Material) This is an expected result due to the added variability that meteorological fluctuations add to observed concentrations of pollutants.

Detrended winter averages for ozone increase by almost 20 ppb between 2005 and 2011 (Fig. 1). Summer values show more variability between years. Decreases in summer averages are seen from 2001 to 2002, 2004–2005, and 2006–2009. However, these decreases are each followed immediately by one or two years of increases in ozone levels. While the overall trend in ozone between 2000 and 2012 shows a slight decrease, it is not nearly as distinct as in the primary pollutants or $PM_{2.5}$.

Table 4
The mean (μ), standard deviation (σ), minimum, and maximum of the meteorological contribution of different subsets of the Jefferson Street data. "high" and "low" splits are made at the mean value for each variable. MAC is the mean absolute contribution of meteorology, or the average absolute STM for each data transect. The differences in the means are statistically significant (as determined by a student's t-test) for temperature and relative humidity subsection. Units are ppb.

	μ	σ	min	max	MAC
All	0.9	10.3	−57.3	44.0	8.4
Temperature					
high	1.5	12.3	−57.3	44.0	9.8
low	0.2	8.7	−43.2	28.6	6.5
Relative Humidity					
high	−2.0	9.7	−44.9	35.8	7.5
low	3.2	11.2	−57.4	44.0	9.1

Both SO_2 and SO_4^{2-} show large decreases in winter concentrations between 2000 and 2001, then remain stagnant until 2008. Concentrations then decrease steadily until 2012, with an exception in 2010. Overall, changes in SO_4^{2-} concentrations mirror changes in SO_2 concentrations with a slightly dampened response.

Winter $PM_{2.5}$ shows a near-steady decrease since 2001. Summer values, on the other hand, show little change before 2006 followed by a large drop between 2007 and 2009. Average values in the summer of 2012 were 11 μgm^{-3} less than average values in 2000.

4.4. Atmospheric response to emissions changes

In order to identify changes in air quality due to emissions changes, fluctuations from both short-term meteorology and seasonal variations must be accounted for. Seasonal variations tend to dominate the variability in concentration signals, and inhibit the ability to observe short-term changes. To this end, the seasonal (S) portion of the detrended concentration signal is removed. The remaining signal is then averaged over different time periods (1, 2, 3, 4, 6, and 12 months). For each averaging time, adjacent mean concentrations are compared with a students t-test, using the Bonferroni method to correct each p-value. This correction accounts for the increased error associated with performing a large number of significance tests (Hastie et al., 2009). All averaging periods are shown for NO_x (Fig. 4), along with annual averages for the remaining pollutants (Fig. 5). See Supplementary material (Figs. S3–S6) for plots of all averaging periods for SO_2, SO_4^{2-}, $PM_{2.5}$, and O_3.

Meteorological detrending increases the ability to observe a statistically significant change in the observed NO_x, SO_2, and SO_4^{2-} concentrations at shorter averaging periods. Results of the signal processing for SO_2 (Fig. 4) show that averaging times of 1 and 2 months are still too short to observe meaningful statistically significant (*i.e.* p-value < 0.05) changes in concentration. Statistically significant peaks in the 1- and 2- month averaging times may be the product of spikes caused by local disturbances. At averaging times of 3 months and greater, however, differences in the meteorologically detrended concentrations become statistically significant and relatable to known emissions changes. At 3 months and higher, both the detrended and raw signals show significant decreases from 2000 to 2001 and 2011 to 2012. Raw concentrations show a significant increase in SO_2 between 2009 and 2010 for averaging periods of 3 months and greater. After detrending, this increase only appears in the 3-month average. At six months averaging periods and longer, no increases are significant.

Detrending of NO_x concentrations (Fig. S3 in Supplementary Material) also shows differences from the measured concentrations. Again, averaging times of 2 months and less capture too much of the noise to show statistically significant differences. First, changes in 2001 and 2007 are significant for averaging times as low as 3 months. These changes are only significant in averaging times of 6 months or greater when meteorological fluctuations are not accounted for. Annual averages show significant changes in the detrended signal in 2007 and 2008 that are not present in the raw signal. Meteorological detrending reduces the standard deviation of the daily NO_x signal (over the 13 year time period) from 15.8 ppb to 12.8 ppb.

$PM_{2.5}$ and ozone are inherently more difficult to analyze using these signal processing techniques. Part of this difficulty arises from the fact that a large portion of $PM_{2.5}$ and all ozone are secondary pollutants whose primary constituents are emitted from a number of sources. When the seasonal signal is removed from the $PM_{2.5}$ and ozone signals, many statistically significant changes exist that are not easily linked to known emissions changes (Figs. S5 and S6 in Supplementary Material). Annual averages for $PM_{2.5}$ and ozone

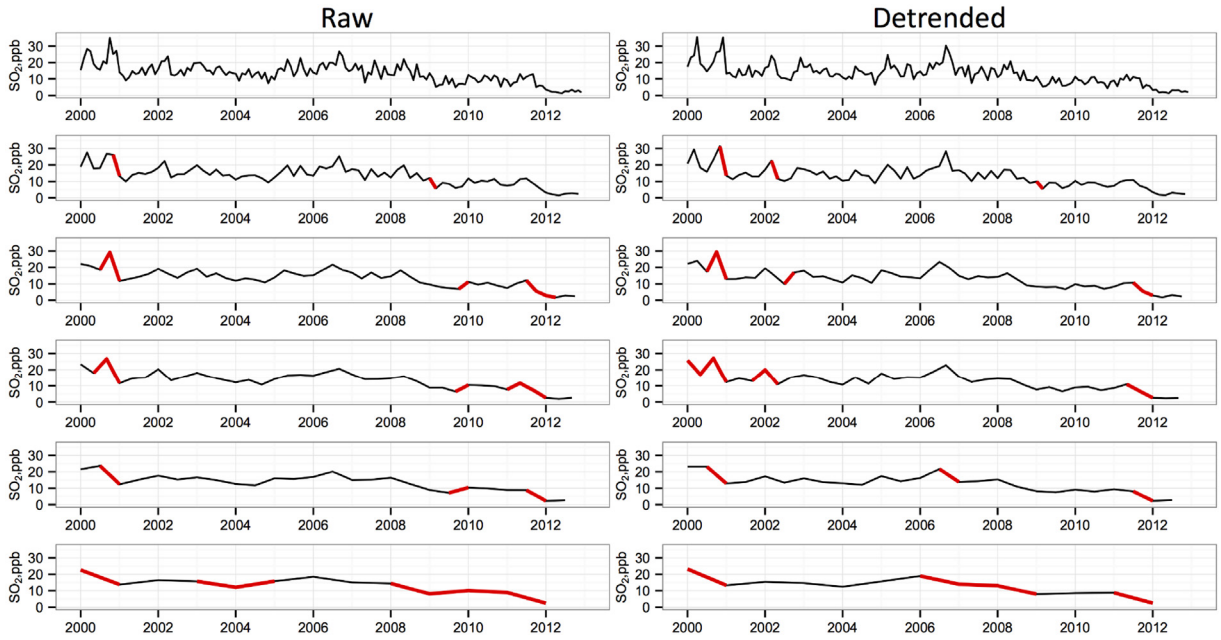

Fig. 4. Averaged SO_2 concentrations at different averaging periods. Raw concentrations are on the left, and meteorologically detrended concentrations are on the right. The averaging times are (top to bottom): 1, 2, 3, 4, 6, and 12 months. Bold red lines indicate a statistically significant ($p < 0.05$) change in the average concentration. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

are exceptions. Statistically significant changes in $PM_{2.5}$ occurred in years from 2007 to 2009 and 2011 to 2012. These years align with significant decreases in sulfate concentrations, which makes sense since sulfate is a major constituent of $PM_{2.5}$ in the southeast. Removing the seasonal signal from the ozone concentrations yields a signal that changes phase (*i.e.* the signal peaks in the summers at the beginning of the data record but changes to the winter at the end) and in the middle of the data record (Fig. S6 in Supplementary Material). This is due to the narrowing distribution of ozone levels (Fig. 1). The result of this is that averaging periods below 12 months have little meaning, since the change in phase drives the majority of the change in average concentrations at smaller periods.

5. Discussion

5.1. Other methods considered

Initially, attempts were made to use band-pass filtering in frequency space (by employing a Fast Fourier Transform (FFT)) to isolate fluctuations at each of the periods in (2). Results of the FFT

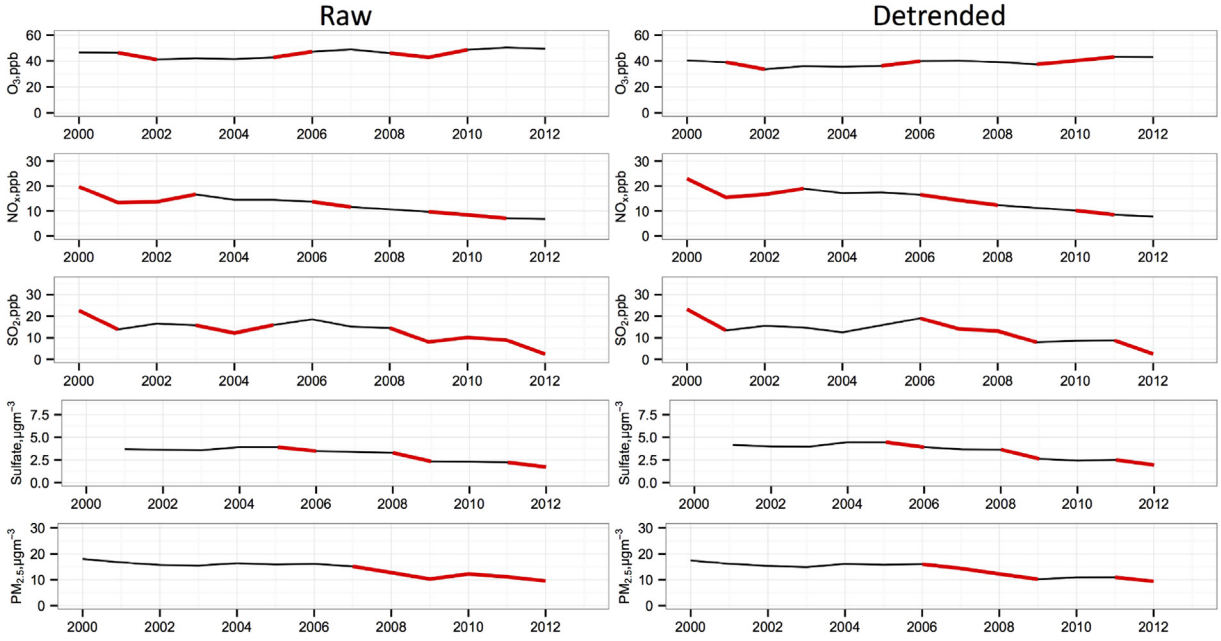

Fig. 5. Averaged annual concentrations of 5 pollutants. Raw concentrations are on the left, and meteorologically detrended concentrations are on the right. Bold red lines indicate a statistically significant ($p < 0.05$) change in the annual average concentration. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

algorithm, however, are highly dependent on the length of the dataset, which reduces the ability to generalize the method to other locations with more or less data. Further, the algorithm does not readily handle missing data, which necessitates filling the missing days with averages or linearly interpolating between days. Given the special treatment required to perform FFT filtering, this method was viewed as less attractive, similar to the results reported in Eskridge et al. (1997).

A fourth method that was taken into consideration in the analysis is reported in Camalier et al. (2007). This method was developed to meteorologically detrend 8-h ozone estimates, and results for cities across the United States are reported at http://www.epa.gov/airtrends/weather.html. However, because the model was developed with the intent of calculating annual adjustments to summer averages, and not day-to-day adjustments, the method was assessed, but is not used further here. Results reported on the EPA website are compared with results from the MKZ and KEA methods.

The MKZ and KEA methods produce detrended annual summer means (Fig. S7 in Supplementary Material) that are comparable with those reported by the EPA at http://www.epa.gov/airtrends/weather2012/atlaGA.jpeg, accessed 27 May, 2015. Actual values were not available, so the EPA values were estimated via visual inspection. The MKZ and KEA methods estimate an average meteorological impact on summer mean ozone of the same sign in 9 out of 13 years of this study. In three of the years that are not adjusted in the same direction (2000, 2003, and 2009), all three methods estimate small adjustments (2–5 ppb), so the differences may be due to outlier days or variations attributable to different monitor locations.

5.2. Comparison with emissions estimates

Reasons for the reductions in primary pollutants can be attributed to a number of factors, including economic activity, fuel costs, and regulatory programs. A number of local and national programs were instituted during this time period. These programs include those targeting point sources (*e.g.* Phase 2 of the Acid Rain Program; the NO_x Budget Trading Program, Georgia was not included in this program, but neighboring states were; the Clean Air Interstate Rule, and the Georgia Multipollutant Control Rule for Electricity Utility Steam Generating Units) and mobile sources (*e.g.* Tier 2 Vehicle and Gasoline Sulfur Program, the 2007 Heavy-Duty Highway Rule, the Mobile Source Air Toxics Rule, and the Georgia Gasoline Marketing Rule) (Georgia Department of Natural Resources Environmental Protection Division Air Protection Branch (2013); Hubbell et al., 2009; Morgenstern et al., 2012). Pachon et al (2012) showed a linear relationship between estimated mobile emissions levels from 1999 to 2007 and measured CO, NO_x, and elemental carbon concentrations (Pachon et al., 2012).

Seasonal variability in NO_x emissions between summers and winters from 2003 to 2008 provide an interesting point of comparison to detrended concentrations. These seasonal emissions controls were required by Georgia in an effort to reduce summer ozone levels. Starting in 2009, the state required year-round controls on NO_x. A comparison of Fig. S8 in Supplemental Material with NO_x concentrations in Fig. 1 shows that EGU summer controls for NO_x had little effect on the detrended summer NO_x concentrations, which increase each year between 2003 and 2005. Decreases in detrended summer and winter NO_x concentrations are observed after 2005. This provides evidence that emissions from local EGUs have a smaller influence on NO_x concentrations in downtown Atlanta than other sources.

The detrended ozone levels, apparently in contrast to its reduced precursor concentrations, do not decrease nearly as much (4% or 2.2 ± 2 ppb) in the summer between 2000 and 2012). Winter averages increased over this time period by 12% (3.8 ± 2 ppb). These results corroborate recent findings in Blanchard et al. (2010) that the ozone response in Atlanta to NO_x levels is less in magnitude than other factors, such as meteorology and ambient levels of non-methane organic carbons (NMOCs).

Many of the regulatory policies established during this period have targeted the highest ozone days. In particular, from 2003 to 2008, seasonal NO_x controls significantly reduced EGU NO_x emissions during the summer ozone season (May–September, Fig. S8 in Supplemental Material). From 2009 on, these controls were kept in place all year. This change in seasonal emissions is used to assess if detrended ozone concentrations can be used to evaluate the impact of these controls on high ozone levels. Days with detrended concentrations greater than 60 ppb were analyzed separately in an attempt to assess how well the impact of those decreases can be identified. Annual averages of concentrations on these days exhibited a decreasing trend over the time period on average, however, inter-annual variability made it difficult to identify if the seasonal EGU emissions had an appreciable impact. Longer-term (3+ year) averages are more stable and do show a marked decrease from the period without seasonal controls to the first four years after seasonal controls were put in place (Table 5). The first averaging period (2000–2002) is before widespread use of seasonal NO_x controls on EGUs in the region, the second (2003–2005) and third (2006–2008) are when seasonal controls were instituted, and the fourth (2009–2012) is when the controls were employed year-round. Average high-ozone day concentrations decrease across the first three periods, and increase slightly in the most recent period. The number of days that saw ozone concentrations greater than 60 ppb decreased significantly over the final three periods. 60 ppb was chosen as a cutoff because of its relevance to current discussions of ozone standards in the United States. The results are reflected in distributions of all ozone levels (Fig. 6); the distribution narrows as new controls are implemented. Days of very high or very low ozone are increasingly rare as controls are installed across a number of sectors.

5.3. Atmospheric response to emissions changes

Because of the range of time scales that are important in atmospheric dynamics, it can be difficult to quantify the effect of a specific control strategy on ambient concentrations. A number of factors regarding the nature of how controls are put in place can also affect the ability to attribute changes in concentration levels at a central monitor to emissions changes. First, regulatory programs are implemented in phases, *e.g.* Phase I (1995) and Phase II (2000) of the Acid Rain Program. Second, more controls are generally required on new emitters (*e.g.* automobiles, EGUs, gas stations, etc.), and the effects are not fully realized until the fleet turns over. Third, multiple regulatory programs are implemented across many domains simultaneously, making it difficult to determine which control is attributed to the change in ambient concentration. Fourth, local sources contribute disproportionally more to the variability in a measured concentration at a central monitor (*e.g.* JST). The time scales analysis shown in the results and discussed

Table 5
Averages concentration of O_3 on days it exceeded 60 ppb and average number of days per year in 4 different periods.

Period	Average [ppb]	# days
2000–2002	72.4	96
2003–2005	70.2	110
2006–2008	66.9	57
2009–2012	68.6	45

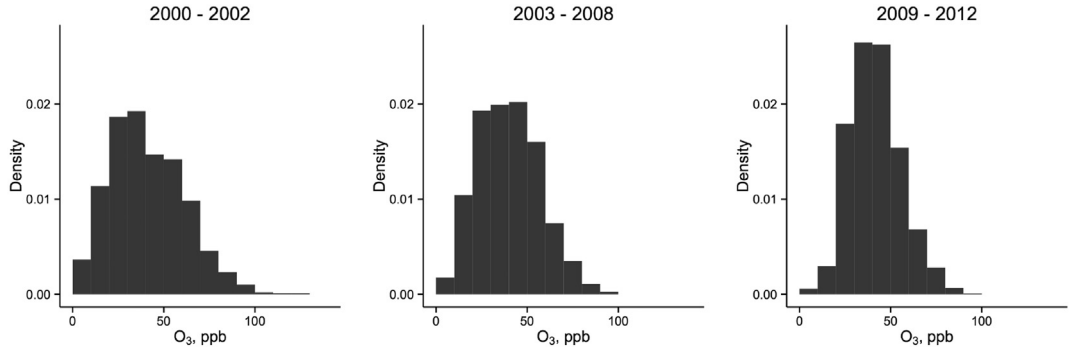

Fig. 6. Normalized densities of concentrations during 3 subsets of the study period.

further here seeks to attribute changes in ambient concentrations to actions that have reduced emissions.

Annual mean detrended SO_2 concentrations show three periods of significant decreases (2000–2001, 2006–2009, and 2011–2012). In the 6 month averaging, the change between 2006 and 2007 is significant. This is the same year that total sales of No. 2 diesel fuel sold by the prime supplier in Georgia changed from 91% Low Sulfur diesel (500 ppm S) and 0% Ultralow Sulfur diesel (15 ppm S) to 35% and 58%. The remaining diesel sold was High Sulfur diesel (5000 ppm S). By 2012, 100% of the diesel sold in Georgia was Ultralow Sulfur diesel (U.S Energy Information Administration, 2014). EGU SO_2 emissions fell between 2008 and 2009. This also contributes to the significant differences in ambient concentrations observed between these years (Fig. 5). The change between 2011 and 2012 is statistically significant for both detrended and raw concentrations for an averaging period of 3 months and greater. Part of this decline can be attributed to the conversion of plant McDonough (a power plant located 10 km northwest of JST) to natural gas between these two years.

One of the significant changes in the NO_x record (Fig. S3 in Supplementary Material) is the change between 2000 and 2001, which is detected at an averaging time of 3 months and greater. NO_x emissions from Plant McDonough were reduced by 20% between 2000 and 2001. Over the total 20-county Atlanta $PM_{2.5}$ nonattainment area, EGU NO_x emissions were reduced by 9.6%. These changes, combined with emissions reductions in other sectors (such as mobile), contributed to a change in both annual average raw and detrended NO_x concentrations between 2000 and 2001 of 32%.

Annual detrended averages of sulfate display significant changes in the same years as SO_2 (Fig. 5). This is expected because sulfate is formed in the atmosphere from SO_2. However, since sulfate must undergo chemical transformations, it is considered a regional pollutant, meaning local sources are not expected to have as much of an impact on sulfate than SO_2. The fact that the two align provides some evidence that strategies to reduce SO_2 emissions in Atlanta influence sulfate concentration, but is also a consequence of the fact that similar regulations have been implemented across the entire region.

The lowest annual average detrended ozone level was observed in 2002, and the two highest values were in 2011 and 2012 (Fig. 5). Statistically significant increases between 2009 and 2011 are present even as NO_x concentrations fell. Annual average ozone levels are increasing because winter concentrations are rising faster than summer levels are falling (Fig. 1). The dramatic rise in winter concentrations provides evidence that winter ozone levels are NO_x inhibited, *i.e.* decreasing NO_x emissions yield higher ozone concentrations. High levels of ozone in the summer show the opposite effect.

6. Summary/conclusions

Meteorological detrending is applied to remove the effect of meteorological fluctuations on measured gaseous and particulate matter pollutants. The daily impact can be substantial, with a mean absolute contribution of 8.4 ppb, with range of −57.2 to 44.0 ppb for ozone. The KZE method builds on previous methods (including the KEA method) to quantify the impact of short-term meteorology on air pollution concentrations. Both methods yield similar results to each other when assessing design values at JST for ozone and $PM_{2.5}$. This investigation shows that the new method is able to do as well or better than the KEA method for all pollutants except SO_4^{2-} in the model fit, and OC in the holdout analysis (Fig. 4). Detrended summertime ozone averages align well with previous results reported by the EPA using the method detailed in Camalier et al. (2007).

In NO_x, SO_2, $PM_{2.5}$, and SO_4^{2-} time series with the seasonal variation removed, statistically significant changes in annual averaged detrended concentrations coincide well with changes in emissions from EGU's and mobile sources. Annual average detrended SO_2 shows statistically significant changes when sulfur concentration in gasoline and diesel was reduced from 2006 to 2008, and when the fuel used at a nearby EGU (Plant McDonough) was changed from coal to natural gas in 2012. Attempts were made to parse out statistically significant changes in NO_x concentrations that reflected summer NO_x controls on Atlanta-area EGU's from 2003 to 2009, but these could not be observed. This is due, in part, to the smaller impact local EGU emissions have on measured concentrations in the city center compared to the impact of mobile NO_x sources.

While seasonal averaging (Fig. 4 and Figs. S3 and S4 in Supplementary Material) allows for the identification of many of the significant changes in atmospheric concentrations of NO_x, SO_2, and sulfate, these averaging times are not as useful for ozone and $PM_{2.5}$. Further, in contrast to the primary pollutants and $PM_{2.5}$, statistically significant changes in ozone concentrations did not coincide with any of the emissions changes mentioned above. Ozone is the only pollutant that does not show a major decrease in mean levels over the 13-year study period. Detrended average summer concentrations decreased 4% (2.2 ± 2 ppb) while winter concentrations increased by 12% (3.8 ± 1.4 ppb) between 2000 and 2012. These results suggest that summer peak ozone levels are *NO_x*-limited, while lower ozone levels are radical-limited (NO_x inhibited). The detrending method developed can be used to adjust for the meteorological impacts on a day-by-day basis for use in acute health studies. The MKZ method provides the benefit of daily detrending that also produces annual averages that agree with previously published detrending methods.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2015.08.007.

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