SENSITIVITY ASSESSMENT OF OZONE AND FINE PARTICULATE MATTER TO EMISSIONS UNDER INFLUENCE OF FUTURE CLIMATE AND EMISSIONS CHANGES

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1. INTRODUCTION

Global climate change can have a potential impact to regional air quality. However, uncertainties in climate change give rise to difficulty in predicting how regional air quality will be affected. In addition, although it is not apparent, climate change and its uncertainties can affect the response of air quality to emissions to a certain degree. To answer the latter question is important because it helps indicate the long-term ability of control strategies on emissions and how they should be maintained or adjusted to account for climate change. In this modeling study, sensitivity assessment of ozone (O3) and fine particulate matter (FPM) (here, sulfate and nitrate) concentrations to emissions under the combined influence of climate and emissions changes was conducted for the continental US during the June episodes of control years 2000, 2001, and 2002 (to be referred to as control case) and those three of mid-century years 2049, 2050, and 2051 (to be referred to as future case). In doing so, the Decoupled Direct Method (DDM) incorporated in the CMAQ model (CMAS, 2005) was employed. The regional-scale meteorology was adopted from the simulation work by Leung et al. (2005), which was based on applying the MM5 model (MM5, 2005) to downscaling outputs generated by the NASA's GISS global climate model (GCM) driven by the SRES A1B emissions scenario of the Inter-governmental Panel on Climate Change (IPCC, 2005). The SRES A1B scenario describes a future world of rapid economic growth and global population that peaks in mid-century and declines thereafter, rapid introduction of more efficient technologies, and balanced usage between fossil fuels and other energy sources. Anthropogenic emissions inventory (EI) was projected (or forecast) for the future case. Methods used and results from the study are discussed below, focusing primarily on future air quality changes. The outline of future plan is also given.

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2. APPROACH

2.1 Emissions Inventory Development

Considering data availability, spatial-temporal extent and resolution, and modeling consistency, we have adopted the 2001 CAIR EI (US EPA, 2005) as the EI for the control case. To seek for an EI projection method suitable for developing the future EI, a number of existing regional- and global-scale emissions projection efforts were reviewed, and the approach finally applied consists of two main steps: near future projection (from 2001 to 2020) and distant future projection (from 2020 to mid-century). The first step followed closely the 2020 CAIR EI of the US EPA while the second step was carried out based on the results suggested by the Netherlands Environmental Assessment Agency's IMAGE model (IMAGE, 2005). Some advantages of the IMAGE model are that 1) it is readily available and covers a relatively long time horizon (up to year 2100), 2) it represents an integrated model that includes

interactions among various components (e.g. society, biosphere, and climate system) to assess issues such as climate change, and 3) it takes into account the SRES emissions scenarios of the IPCC. The BELD3 land use database (US EPA, 2005) was used in estimating biogenic emissions, and it was held the same for both control and future cases due to no clear scientific basis available for forecasting distant-future land use changes. So far, the first phase of the EI work has been completed, in which anthropogenic emissions in the US were projected while those in Canada and Mexico were not (i.e. held constant). Their projection is currently underway. A new spatial surrogate profile file was created to be consistent with the new EPA surrogates and the grid configuration as well as the map projection selected for the air quality modeling using the MIMS Spatial Allocator (CEP-UNC, 2005).

2.2 Modeling

A 147x111 grid domain was used, covering the continental US and parts of Canada and Mexico, with a grid size of 36 km. Emissions data were processed by the SMOKE model (CMAS, 2005) to generate emissions fields. Air quality modeling was implemented using the CMAQ model for the chosen six summer episodes (i.e. June of 2000-2002 and 2049-2051). As said previously, the regional-scale meteorology was from Leung et al. (2005), in which the meteorological conditions of present and future (~mid-century) periods (10-yr each) were compared, suggesting various changes in mean surface air temperature, mean precipitation, mean downward solar radiation, etc. spatially and seasonally. Fig. 1 shows the average surface air temperatures over the US and its five regions, which are West (WS), Plains (PL), Midwest (MW), Northeast (NE), and Southeast (SE), in the control case and their changes (in parentheses) in the future case. As seen, every region has an increase in temperature (~1.5-2.0 deg. C), and the SE is the warmest region. Table 1 gives a summary of changes in total NOX, SO2, NH3, VOC, biogenic VOC emissions (averaged over the episodes of each case), showing the significant reduction of both NOX and SO2 due primarily to the fact that the EI projection conducted in the study attempts to follow the underlying assumptions of the SRES A1B scenario. The warmer (modeled) future climate also plays a crucial role in inducing more VOC emissions from biogenic sources (~21% for the US) since VOC emissions are typically temperature-dependent. In the NE, there is only a slight change in total VOC emissions. For NH3 emissions, their changes are positive but relatively small (<10%).

The DDM module for the CMAQ model used here is based on Cohan et al. (2003) and Napelenok et al. (2005). It is capable of directly calculating the sensitivity of any gaseous and aerosol concentrations to emissions. Here, let C_i and E_j be the concentration of species i and the base (or nominal) value of emission j, respectively. By defining

$$\Delta E_j = \varepsilon_j E_j , \qquad (1)$$

where ε_j is the perturbation factor on E_j, the sensitivity of C_i to ε_j (denoted by S_{ij}) can be expressed as

$$S_{ij} = \partial C_i / \partial \varepsilon_j . \qquad (2)$$

Eq. (2) is the definition of sensitivity used in the context of this work. By the above relationships, the units of both C_i and S_{ij} are the same.

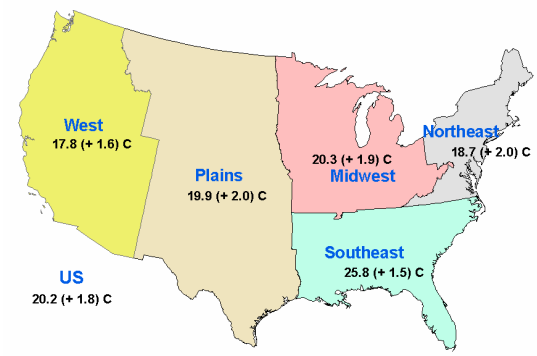

Fig. 1. Average surface air temperatures for different US regions in the control case and their changes (in parentheses) in the future case

3. RESULTS AND DISCUSSION

In the following, we will limit the presentation and discussion to the modeling results of O3, fine sulfate aerosol (i.e. ASO4 = ASO4I + ASO4J), and fine nitrate aerosol (i.e. ANO3 = ANO3I + ANO3J) for the continental US, the NE, and the SE. In Fig. 2a, O3 (averaged over the episodes of each case) decreases for the SE and the US, with a relatively larger decrease (~20%) for the SE. In Table 2, O3 appears more sensitive to NOX

Table 1. Changes in total NOX, SO2, VOC, NH3, and biogenic (B) VOC emissions between the control and future cases (based on the first phase of the EI development work)

Region	Emission	Change (%)
WS	NOX	-51.8
	SO2	-49.3
	NH3	+4.1
	VOC	+9.0
	VOC (B)	+19.0
PL	NOX	-41.1
	SO2	-67.1
	NH3	+2.3
	VOC	+16.2
	VOC (B)	+27.0
MW	NOX	-52.7
	SO2	-60.1
	NH3	+5.1
	VOC	+8.1
	VOC (B)	+28.9
NE	NOX	-65.1
	SO2	-66.6
	NH3	+9.9
	VOC	-0.72
	VOC (B)	+29.6
SE	NOX	-62.8
	SO2	-60.5
	NH3	+4.6
	VOC	+7.0
	VOC (B)	+15.0
US	NOX	-54.1
	SO2	-62.2
	NH3	+4.5
	VOC	+8.6
	VOC (B)	+21.4

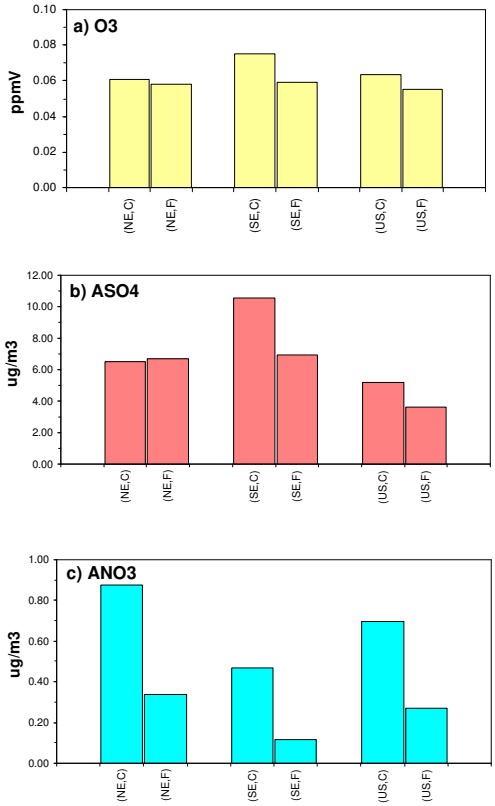

Fig. 2. O3, ASO4, and ANO3 levels (averaged over the episodes of each case) for NE, SE, and US (C: Control, F: Future)

than to VOC, and its sensitivity to both NOX and VOC in the control case does not differ much from that in the future case. Thus, it is fair to say that the substantial NOX reduction in the future case (Table 1) is the main reason of the O3 decreases. Note that the units of sensitivity are the same as those of concentration (i.e. ppmV for O3 and ug/m3 for ASO4 and ANO3). Notice that the levels of O3 in both cases differ only slightly for the NE. This may be attributed, in part, to keeping the NOX EI of Canada constant for both control and future cases. Since the NE is close to Canada, emissions from Canada are more likely to affect the air quality of this region.

Similar findings are obtained for ASO4. The decreases of ASO4 for the SE (~35%) and the US (~30%) can be explained by the substantial SO2 reduction. ASO4 is positively sensitive to both SO2 and NOX. Its sensitivity to the former is clearly larger, particularly for the SE (8.3 ug/m3 and 5.1 ug/m3 in the control and future cases, respectively). For the NE, a slight increase in ASO4 is found, which may also be attributed, in part, to the fact that the SO2 EI of Canada was held constant for both control and future cases.

From Table 2, the sensitivity of ANO3 to emissions is positive for NOX and NH3 but negative for SO2 because sulfate preferentially scavenges ammonia, leaving less to combine with nitric acid to form ammonium nitrate aerosol. The magnitudes of sensitivity to NOX and SO2 are comparable for the NE and SE, but become larger for NOX for the US. From Fig. 2c, ANO3 decreases significantly in every region (~60% for the NE, ~75% for the SE, and ~60% for the US). By comparing Figs. 2b and 2c, it is seen that the contribution (in terms of mass) of ANO3 to FPM is much smaller than that of ASO4 for the episodes chosen in this study. Since the changes in NH3 emissions are relatively small, it can be said that the decreases of ANO3 were caused primarily by the NOX reduction. Nevertheless, this does not give a full explanation particularly to the role of SO2 changes to such decreases, thus needing further investigation. As seen, the combination of climate and emissions changes can affect the

sensitivity of to emissions to a degree but, mostly, not significantly.

The study presented here can be viewed as a limited assessment of the sensitivity of O3 and FPM to emissions. It is also of practical interest to assess the impacts of climate change alone (i.e. direct impacts) on both regional air quality and its sensitivity to emissions by maintaining emissions fields the same for both control and future cases, as in Manomaiphiboon et al. (2004), and then comparing results so that the roles of climate and emissions in air quality are better identified. As seen, sensitivity analysis is one of useful tools for investigating the interplay between air quality and emissions that can be changed by direct reduction and/or meteorology.

Table 2. Comparison of sensitivity of O3, ASO4, and ANO3 to emissions (averaged over the episodes of each case) for NE, SE, and US

Region	Sensitivity	Control	Future	Diff
NE	O3 to NOX	0.011	0.017	0.006
	O3 to VOC	0.003	-0.002	-0.005
	ASO4 to SO2	4.779	4.468	-0.311
	ASO4 to NOX	0.504	0.741	0.237
	ASO4 to NH3	0.040	0.123	0.083
	ANO3 to SO2	-0.672	-0.358	0.313
	ANO3 to NOX	0.691	0.419	-0.272
	ANO3 to NH3	1.244	0.562	-0.681
SE	O3 to NOX	0.024	0.022	-0.002
	O3 to VOC	0.000	-0.004	-0.005
	ASO4 to SO2	8.325	5.062	-3.262
	ASO4 to NOX	0.939	0.691	-0.247
	ASO4 to NH3	0.043	0.155	0.111
	ANO3 to SO2	-0.547	-0.162	0.386
	ANO3 to NOX	0.591	0.162	-0.429
	ANO3 to NH3	0.802	0.243	-0.559
US	O3 to NOX	0.015	0.015	0.000
	O3 to VOC	0.001	-0.002	-0.003
	ASO4 to SO2	3.970	2.455	-1.515
	ASO4 to NOX	0.398	0.296	-0.102
	ASO4 to NH3	0.078	0.190	0.111
	ANO3 to SO2	-0.546	-0.187	0.359
	ANO3 to NOX	0.707	0.367	-0.340
	ANO3 to NH3	0.901	0.306	-0.595

4. FUTURE WORK

Many aspects of the current study are being enhanced and extended: 1) Development of EI projection for the US, Canada, and Mexico, 2) Performing longer simulations, 3) Examining GCM-MM5/CMAQ performance, e.g. using some techniques discussed in Hogrefe et al. (2004). The evaluation process is useful because it helps identify the suitability of simulated climate and suggests how long simulations should cover such that the underlying climate conditions are captured.

In addition, since there exist large uncertainties in predicting future climate conditions, it is of further interest to examine how such uncertainties could impact our estimation of future regional air quality and implications related to the control strategies to reduce O3 and FPM precursors. To do so, we consider to use the uncertainty ranges of meteorological variables of interest, which are suggested and quantified by recent simulations (Webster et al., 2002 & 2003) using the Integrated Global System Model (IGSM) (Prinn et al., 1999), and then perturb future meteorological fields by some representative uncertainty values from those ranges through and within the MM5 model.

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