9A.14 EFFECTS OF UNCERTAINTY IN METEOROLOGICAL INPUTS ON O3 CONCENTRATION, O3 PRODUCTION EFFICIENCY, AND O3 SENSITIVITY TO EMISSIONS REDUCTIONS IN THE REGIONAL ACID DEPOSITION MODEL

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1. INTRODUCTION

Photochemical models are widely used to determine the level of VOC and NOx emissions reductions required to attain the National Ambient Air Quality Standard for ozone (O3). Modeled O3 concentrations, [O3], are affected by photochemical production of O3, by NOx and VOC precursor emissions, and by meteorological processes that include horizontal transport, vertical mixing, cloud effects on actinic flux, and deposition of O3 and its precursors. There is considerable uncertainty in simulating each of these processes individually and in simulating the effects of interactions or feedbacks between chemical and meteorological processes; thus, it is possible for models to correctly reproduce observed [O3] because of compensating errors in model inputs or model processes. This puts into question how much confidence we can have in model predictions of the effectiveness of VOC and NOx emissions reductions. Despite the importance of meteorology, there has been little experience within the air quality research community in quantifying meteorological impacts on photochemistry, particularly with the use of primitive equation meteorological drivers. In this study, we conduct a series of sensitivity experiments with the Regional Acid Deposition Model (RADM) using changes in meteorological inputs, such as PBL height and cloud effects, to examine their impact on [O3] and on O3 production efficiency. Using each of these modified meteorology scenarios as a "base case" we then evaluate the effect of meteorology on the effectiveness of emissions controls by running additional model simulations with 15% and 50% reductions in NOx or VOC emissions.

2. THE BASE RADM EXPERIMENT

The RADM simulations were conducted over the eastern US for the time period of July 19 to August 12, 1988, at a resolution of 80km on a three-dimensional grid of 35 by 38 horizontal elements and 21 vertical layers. Thirteen layers are in the PBL with the lowest layer extending to approximately 40m above the surface. The physical and chemical processes included in RADM are advection, vertical eddy mixing, cloud effects, dry deposition, emissions, and photochemical production and losses. RADM uses the RADM2 Chemical Mechanism developed by Stockwell et al. (1990). Anthropogenic emissions are from the NAPAP 90 emissions inventories. Biogenic emissions from agriculture and forests are from BEIS2. Clear sky photolysis rates are calculated by the delta-Eddington method. The dry deposition velocity is computed from the resistance-in-series method. The subgrid-scale turbulent mixing processes in the PBL are described by using the local eddy diffusivities (K_z), which are based on boundary layer scaling theory (Byun, 1990 and 1991). For clouds, four primary effects are represented: (1) vertical redistribution, (2) aqueous chemical reactions, (3) scavenging effect (loss), and (4) radiative effect.

3. METEOROLOGICAL INPUT FROM MM5

The meteorological fields used in the RADM simulations are derived from archives generated by hydrostatic MM5 simulations, the fifth-generation Penn State/NCAR Mesoscale Model [Grell et al., 1994]. Three-dimensional fields of horizontal winds (u,v), temperature (T), specific humidity (Q_v) and two-dimensional surface pressure (p_s), column-cumulative precipitation rates, ground temperature, etc., are archived at hourly intervals during MM5 runs. We use a meteorology-chemistry interface program (MCIP) to derive RADM-required fields from MM5 archives. MCIP estimates various surface and PBL parameters (such as friction velocity, surface heat flux, Monin-Obukov length, and PBL mixing height) as well as dry deposition velocities. These fields are used as inputs to RADM and are used in RADM algorithms to determine cloudiness and vertical velocity, and to quantify various subgrid scale transport processes, dry and wet deposition rates, and chemical reaction rates.

4. SENSITIVITY EXPERIMENTS

The sensitivity experiments we conducted are summarized in Table 1. The second column lists the meteorological parameters or processes to be changed for the sensitivity experiment. The third and fourth columns show how the parameters or processes are changed as compared to the base RADM experiment. The last column designates a symbol which will be used in the rest of the paper to refer to each individual

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Table 1. Meteorology Sensitivity Experiments, Their Affected Parameters and Processes, and Designated Symbols.

Exp. #	Met Parameter or Physical Process	Value or Definition for Sensitivity Experiment	Value or Definition Base Experiment	Symbol Designated
1	PBL height (H)	50%	100%	0.5H
2	PBL height (H)	150%	100%	1.5H
3	PBL mixing scheme for CBL	Pleim & Chang (1992)	Byun (1990)	ACM
4	Derivation of surface parameters and PBL height estimation in MCIP	Byun (1991); ground temperature	Byun (1990); Holtslag etal(1995)	SFC
5	MM4 meteorology	MM4	MM5	MM4
6	All cloud effects	all not included	all included	noCld
7	radiative effect of clouds	not included	included	noJcld
8	all other cloud effects than radiative	not included	included	noWet

sensitivity experiment for convenience of presentation. The eight experiments listed in Table 1 can be divided into two categories. Category one covers experiments 1 to 5 that are designed to test PBL mixing schemes and parameters, especially the PBL height. Category two includes experiments 6 to 8 designed to test the effect of clouds processes.

The first two sensitivity experiments in Table 1 apply a simple scaling factor, 50% and 150% respectively, to the base PBL height (designated as "H" hereafter) in RADM, while all the other parameters and processes are unchanged. Their designated symbols are "0.5H" and "1.5H", respectively. The third experiment, symbolized as "ACM", utilizes the asymmetric convective model (ACM) of Pleim and Chang (1992) as an alternative to describe the eddy mixing process for the convective boundary layer (CBL) in RADM. The fourth experiment, symbolized as "SFC", estimates various surface parameters (such as friction velocity, surface heat flux, and Monin-Obukov length) in MCIP using the PBL scaling theory developed by Byun (1991) as opposed to the surface-layer scaling theory for the base experiment (Byun, 1990). This SFC configuration has been used in most of our previous RADM simulations using MM4 meteorology. In order to explain the sensitivity results presented in section 5, it is crucial to understand the difference in the details of the estimation algorithms for convective PBL height among the base, ACM, and SFC experiments. While the algorithms in all three experiments are based on vertical profiles of potential temperature (PT) and the Richardson number, the difference lies on the initial temperature assumed for a rising air parcel from near the surface. The base case uses an effective surface PT which is the near-surface air PT plus a temperature excess measuring the strength of convective thermals in CBL (Holtslag et al., 1995). The same algorithm is employed in MM5 and such computed H has a maximum value of about 2 km over eastern US. On the other hand, ACM uses the near-surface air PT, which results in significantly lower H than the base experiment. SFC uses the ground PT, which results in much higher H than the base experiment. Indeed, SFC has a maximum value of about 3 km over eastern US. During the daytime when the ground is heated, the effective surface PT used by the base experiment is higher than the air PT but lower than the ground PT. A buoyant thermal with a higher initial temperature will shoot higher during convection. The fifth sensitivity experiment, designated as "MM4" in Table 1, uses MM4-produced meteorology and an older version of MCIP. This older MCIP has a similar configuration as SFC and therefore its PBL height is significantly higher than the base experiment.

The sixth to eighth experiments are conducted to test how cloud processes affect the model-predicted O3 chemistry. In the sixth experiment, designated as "noCld", all cloud effects are turned off. In the seventh experiment, designated as "noJcld", only cloud radiative effects on photolysis rates are turned off, creating effectively clear sky photolysis. In the eighth sensitivity experiment, designated as "noWet", other cloud effects are turned off while the radiative effect is unchanged. For each of these meteorology sensitivity experiments, two runs are conducted. One is with all emissions and the other is with anthropogenic NOx emissions uniformly reduced by 15%. For the base experiment, a third run is also conducted in which anthropogenic NOx emissions are uniformly reduced by 50%. It will be designated as ".5nox" in the following text. The base run with 15% NOx emissions reduction will be designated as ".85nox".

5. RESULTS

Results for an urban grid cell in New York City (NYC) are given in Tables 2 and 4 and for a rural grid cell in Scotia, Pennsylvania, in Tables 3 and 5. The NYC cell is characterized by high emissions of NO that titrate O3 causing low [O3] near the surface, with higher [O3] levels above the surface layer. The Scotia cell is a rural, NOx-limited cell in which [O3] levels and local photochemistry are largely determined by the transport of O3 and it's precursors into the cell. The results in Tables 2 through 4 are 7-hour averages from 10am to 4pm local time during the period of July 20 to August 12. Tables 2 and 3 shows results for the base case scenario, the base case with 15% and 50% NOx emissions reductions, and for the eight meteorological sensitivity experiments.

5.1 *Ozone Concentration*

Table 2. Daytime Average [O3] and Local O3 Production Efficiency at NYC for the Base, NOx Emissions Reductions, and Uncertainty in Meteorology Parameters and Processes. Average Concentrations Are Computed from 10am to 4pm Local Time for the 11 highest [O3] days of the base case at New York City.

	BASE	.85nox	.5nox	0.5H	1.5H	ACM	SFC	MM4	noCld	noJcld	noWet
O3 (> 100ppb)	126	126	115	141	113	129	117	106	139	133	132
P(Ox)	160	160	136	189	142	162	144	140	167	161	166
P(Ox) / P(HNO3)	7.1	8.4	15.0	6.6	7.3	6.6	7.4	7.2	7.0	7.0	7.1

Table 3. The same as table 2 but for Scotia, Pennsylvania, average values for 4 highest [O3] days of the base case

	BASE	.85nox	.5nox	0.5H	1.5H	ACM	SFC	MM4	noCld	noJcld	noWet
[O3] (> 100ppb)	121	115	98	130	112	123	114	111	125	122	124
P(Ox)	58	55	38	72	50	54	50	49	55	54	58
P(Ox) / P(HNO3)	47	51	81	52	48	38	45	43	46	46	47

The first row in Tables 2 and 3 shows the average [O3] for days when the Base Case average [O3] exceeds 100 ppbv (11 days at NYC and 4 days at Scotia). At Scotia, average [O3] decreases from 121 ppbv to 115 pbbv for the 15% NOx emissions reduction and to 98 ppbv for the 50% NOx emissions reduction, reflecting the NOx-limited characteristics of Scotia. In NYC, however, average [O3] is unchanged for the 15% NOx reduction and decreases only 11 ppbv for the 50% NOx reduction case, which is only half as much as at Scotia. These results suggest that marginally controlling NOx has little effect in this NYC cell, although it may benefit downwind locations.

The PBL-related meteorology sensitivity experiments (0.5H, 1.5H, ACM, SFC, MM4) show more significant effects at NYC than at Scotia, with the difference of [O3] from the base experiment ranging from -20 to 15 ppb at NYC versus -10 to 9 ppb at Scotia. In the experiments in which we removed cloud processes (noCld, noJcld, noWet), the NYC [O3] increased by 6 to 12 ppbv, and in Scotia [O3] increased by only 1 to 4 ppb. These results demonstrate that variations in the representation of PBL height, the eddy mixing process, and cloud processes significantly affect [O3] predictions at NYC, with smaller but still notable effects at Scotia.

For NYC, the [O3] reductions due to greater vertical mixing as in 0.5H, SFC, and MM4 are much larger than the change due to 15% NOx emission reduction; in fact, they are comparable to the 11 ppb reduction achieved by the 50% NOx emissions reduction. Thus, we estimate that the meteorological uncertainties in PBL height have comparable impacts on [O3] as a 30 to 50% uncertainty in emissions has. This suggests that it is as important to quantify and reduce meteorological uncertainty as emissions uncertainty.

5.2 *Ozone Production Efficiency*

To simulate O3 correctly in regional scale models, it is necessary to accurately simulate the O3 production efficiency per HNO3 produced, P(O3)/P(HNO3) or P(Ox)/P(HNO3) (Tonnesen and Dennis, 1997). Here Ox is defined as O3+NO2+O1D+O3P+PAN+2NO3+3N2O5 and P() represents the net chemical production rate. In Tables 2 and 3 we show the effect of meteorological inputs on the O3 production efficiency as well as P(Ox), as integrated over the time period 10 AM to 4 PM for the eight highest [O3] days.

Values of P(Ox)/P(HNO3) less than 9 suggest radical-limited conditions (Tonnesen and Dennis, 1997). For NYC, P(Ox)/P(HNO3) is less than 8 ppb/ppb for the base case and all of the meteorology sensitivity experiments; this indicates that all of these are radical-limited. P(Ox) increased and O3 production efficiency decreased in the meteorology experiments with reduced vertical mixing (0.5H, ACM). This indicates that reduced vertical mixing causes the system to become relatively more radical-limited, while increased vertical mixing causes the system to shift to relatively more NOx-limited conditions. Results for Scotia are more ambiguous. Table 3 does show that P(Ox)/P(HNO3) is greater than 40 ppb/ppb for all of the meteorology experiments. This indicates that conditions are very NOx-limited. In contrast to NYC, the O3 production efficiency increased in Scotia for the 0.5H experiment suggesting that reduced vertical mixing caused this rural cell to become more NOx-limited. It is likely that this occurs because NOx was converted to HNO3 more rapidly in the upwind urban areas in 0.5H, so less NOx and PAN were transported to Scotia. These results show that variations in meteorological inputs do affect O3 production efficiency and O3 sensitivity to NOx.

5.3 *Response to NOx Emissions Reduction*

This section examines how the meteorological uncertainties change the system's response to O3 precursors' control. We only present results for NOx emissions reduction here, in Tables 4 and 5. We compare the percent change of daytime average O3 concentration due to the 15% NOx emissions reduction for the base and each sensitivity experiment: $([O3]_{nox}$ - [O3])/[O3]*100, where $[O3]_{nox}$ is for the corresponding 15% NOx reduction runs. Some selected results for the 50% NOx reduction are also included in the last three columns. The results are subdivided, according to the O3 concentration distribution, into an upper third, a middle third, and a lower third partition, of 8 days each.

Table 4. Response of Daytime Average Ozone Percent Change for 15% NOx Emissions Reductions to Uncertainty in Meteorology Parameters and Processes at NYC. The last row shows the 24 day average [O3] of 96 ppbv and the average change in [O3] for all 24 days for each meteorology experiment. Rows 1 to 3 show the averages sorted into three bins for high, intermediate, and low [O3].

Class (ppbv)	O3 (ppbv)	ACM	0.5H	BASE	1.5H	SFC	MM4	noCld	noJcld	noWet	0.5H (50% NOx reduction)	BASE (50% NOx reduction)	1.5H (50% NOx reduction)
> 115	133	2.1	2.6	0.0	-0.9	-1.0	-0.3	-0.5	-0.5	0.1	-2.4	-9.1	-11.0
75-115	96	3.7	4.4	1.8	0.3	0.6	0.9	2.2	1.5	2.0	3.1	-3.4	- 7.1
< 75	59	8.4	10.2	5.8	3.0	3.3	3.7	5.2	5.9	5.6	30.7	14.1	4.3
ALL	96	4.7	5.7	2.5	0.8	1.0	1.4	2.3	2.5	2.8	10.5	0.5	- 4.6

Table 5. The Same As Table 4 but at Scotia, Pennsylvania.

Class (ppbv)	O3 (ppbv)	ACM	0.5H	BASE	1.5H	SFC	MM4	noCld	noJcld	noWet	0.5H (50% NOx reduction)	BASE (50% NOx reduction)	1.5H (50% NOx reduction)
> 90	108	-4.1	-4.5	-4.1	-4.0	-4.1	-3.9	-4.1	-4.1	-4.1	-18.7	-17.5	-17.0
64 - 90	74	-3.7	-3.9	-3.8	-3.7	-3.8	-3.6	-4.2	-4.0	-3.8	-16.3	-15.6	-15.2
< 64	60	-3.5	-3.6	-3.4	-3.3	-3.4	-3.5	-4.3	-3.9	-3.8	-14.2	-13.6	-13.0
ALL	81	-3.8	-4.0	-3.8	-3.7	-3.8	-3.7	-4.2	-4.0	-3.9	-16.4	-15.6	-15.1

For the Base Case at NYC (column 5 in Table 4), the average percent change for the upper third partition is 0.0%, and the system on high [O3] days is on or near the ozone ridge line (The ozone ridge line is discussed in Tonnesen and Dennis, 1997). The percent change increases to 1.8% and 5.8%, respectively, for the middle and lower partitions, indicating that the system is more radical-limited on lower [O3] days.

The results indicate that lowering the mixing height moves the system towards being more radical limited. This is consistent with values of the indicators in Table 2, in which P(Ox)/P(HNO3) was lower for 0.5H and ACM (both 6.6) than for the base case (7.1). The shift is similar for the upper and middle third partitions of the concentration distribution. For example, the ACM percent change is 2.1 (2.1 minus 0.0) percentage points larger than Base for O3 > 115 ppb and 1.9 (3.7 minus 1.8) percentages points larger for 75 ppb < O3 < 115 ppb. Figure 1 presents the percent change in the Base Case for a 15% NOx reduction compared to the percent change for the ACM case, showing that the shift is systematic. All of the days are shifted towards greater radical limitation by approximately the same percentage points, with a slight trend towards a larger shift as the Base case percent change increases. The intercept in Figure 1 (1.9) can be compared to the 2.1 and 1.9 percentage point shifts noted above. Sillman et al.(1995) saw a similar effect for Atlanta when they decreased the local eddy diffusivity, Kz.

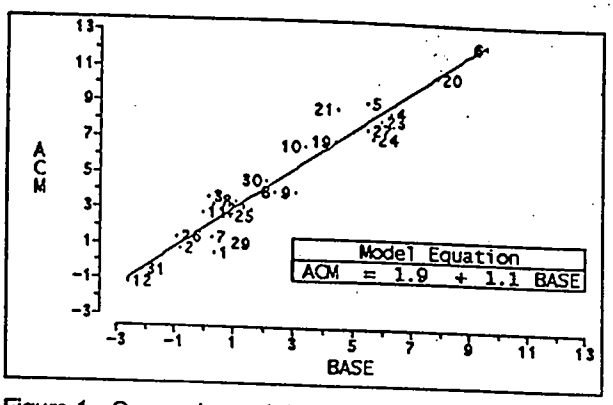

Figure 1. Comparison of the percent change of daytime average [O3] due to a 15% NOx reduction in the ACM case versus the Base case. The dates are shown beside each data point. The "Model Equation" in the embedded box is for the linear fitting line to the data points.

Comparing the 1.5H, SFC, and MM4 results with the base case indicates that lifting the mixing height moves the system towards NOx limitation. Figure 2 compares the percent change in the SFC Case to the percent change for the Base case, showing that the shift is also systematic. There is a slight trend towards a larger shift as the Base percent change increases. The intercept in Figure 2 (-1.0) can be compared to the -1.0 percentage point shift of SFC from BASE for the O3 > 115 ppb partition as shown in Table 4 (-1.0 minus 0.0).

We note that the magnitude of the uncertainty as we have defined it is unbalanced relative to the volume available for mixing. The difference between the 0.5H and Base changes the volume by 100%, whereas the

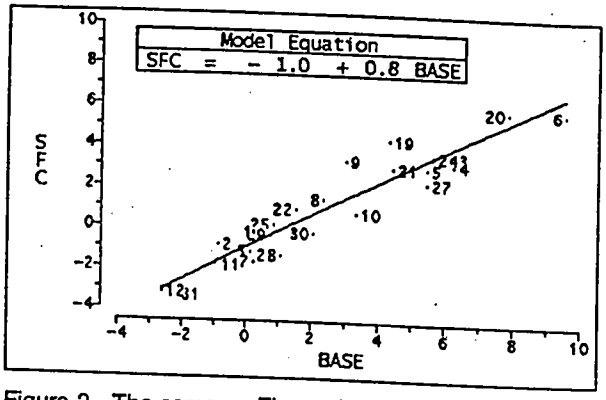

Figure 2. The same as Figure 1 except for comparison between the SFC case and the Base case.

difference between the Base and 1.5H changes the volume by 50%. We see this asymmetry reflected in the size of the difference in ozone concentration percent change due to the 15% NOx emissions reduction between the 0.5H and 1.5H cases from the Base case.

The results above suggest that meteorology uncertainties have significant effects on model-predicted [O3] response to NOx and VOC emissions reductions at NYC. Therefore, it is important to reduce meteorology's uncertainty to improve confidence in models applications used to design [O3] control strategies. To simply get the ozone concentration prediction right by tuning the meteorology may change the system's [O3] sensitivity to emissions which in turn could lead to non-ideal control strategy decisions.

We also examined the effect of the mixing height uncertainty on the 50% NOx reduction. As shown in Table 4 for NYC, the effect of the meteorological uncertainty scales somewhat less than proportionately to the size of the emissions reduction. For example, for [O3] > 115ppb, the size of the shift for 0.5H is 6.7 (-2.4 minus -9.1) and 2.5 (2.6 minus 0.0) percentage points for 50% and 15% NOx reductions, respectively. The shift increases by a factor of 2.6 when the emissions reduction increased by a factor of 3.3. The size of the shift averaged over all 24 days for 0.5H is 10 percentage points (10.5% minus 0.5%). This change is comparable to the change in emissions reduction effectiveness noted by Sistla et al. (1996).

At Scotia, PA, a rural site, we see little influence on the control strategy effectiveness due to the meteorological uncertainties (Table 5). For the cases associated with changes in mixing height, we believe this is due to the opposing effects that happened to cancel in our particular experiments. We explain this for the 0.5H case. With 0.5H we would expect the [NOx] to increase due to a smaller volume and reduce the production efficiency, changing the control strategy effectiveness. We also expect the effect of the sensitivity in the urban areas to be transported into the rural areas. In Table 2 for NYC, we see that P(Ox) increased 18% from the Base to 0.5H. But the production efficiency, P(Ox)/P(HNO3) decreased by 13%. This means that the P(HNO3) increased by 36%. Hence, NOx in the urban areas is being terminated faster, reducing the NOx concentrations being transported to the rural areas. The reduction in transported NOx counters the smaller volume into which the NOx is going, masking its effect and resulting in an apparently small sensitivity effect.

Our results suggest that the meteorological uncertainties in PBL processes and parameters create a systematic shift in control strategy response that is independent of the state of the system relative to the ozone ridge line. On the other hand, the response of peak ozone to the meteorological uncertainty is dependent on the state of the system relative to the ozone ridge line. Thus, one cannot logically deduce the effect of the meteorological uncertainty on the control strategy response based on an examination of the [O3] response to the meteorological uncertainty.

6. CONCLUSIONS

These results demonstrate that an accurate representation of PBL height, eddy mixing, and cloud processes in the model is extremely important for [O3] predictions and [O3] sensitivity. Therefore, meteorological uncertainty is equally important to be quantified and improved as emissions uncertainty.

The results suggest that meteorology uncertainty does convert into uncertainty of model-predicted effectiveness of emissions control at NYC and even changes the system's sensitivity regime. This conclusion holds even on those days when meteorology uncertainty does not result in differences in [O3]. In other words, the effect of meteorology uncertainties on base [O3] does not provide any insight into how the system's response to emissions control will be changed. Therefore, meteorology uncertainties and their consequences on model results must be carefully addressed before we can rely on numerical model's prediction as a basis in designing control strategy. It also tells us that merely to get O3 prediction right does not necessarily get the system's sensitivity right; independent test and validation for both must be done, and both are equally important for model evaluation.

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