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MODELING AEROSOL FORMATION AND TRANSPORT IN THE PACIFIC NORTHWEST WITH THE COMMUNITY MULTI-SCALE AIR QUALITY (CMAQ) MODELING SYSTEM

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1. INTRODUCTION

The Northwest Regional Modeling Center (NWRMC) demonstration project was undertaken to determine the feasibility of using the Community Multi-scale Air Quality (CMAQ) modeling system to investigate regional haze in the Pacific Northwest. The project was also aimed at establishing the NWRMC as a virtual modeling center accessible to all Northwest air quality stakeholders. This was a collaborative effort between the Environmental Protection Agency (EPA) Region 10, Washington State University (WSU), Washington State Department of Ecology, Idaho Department of Environmental Quality (ID DEQ), and Oregon Department of Environmental Quality (OR DEQ) to address regional haze and visibility problems in the Pacific Northwest. Canada also played a role in the project because of the common border and problems often experienced with data inconsistencies between the countries.

The approach of the NWRMC involved two phases. In the first phase, results from CMAQ were compared to results from CALGRID, as discussed in O'Neill and Lamb (2002), to simulate ozone formation and transport along the I-5 corridor of western Washington and Oregon. The second phase of the project involved expanding to a regional scale domain to investigate aerosol formation and transport. The period of July 1-16, 1996 was selected as the study focus period.

Two emission inventories (EI) were developed for the second phase of this study. For the first EI, anthropogenic emissions were based on the National Emission Trend 1996 (NET96) database (U.S. EPA, 2000), and biogenic emissions were obtained from the BEIS2 (Pierce et al., 1998) biogenic emissions model. For the second EI, anthropogenic emissions were developed from the "ground up" by the NWRMC participants. This emission inventory will hereafter be referred to the NWRMC emission inventory (WA DOE, 2002). As part of the NWRMC effort, Canada provided a point source emission inventory, and biogenic emissions were obtained from the GLOBEIS (Guenther et al., 2000) biogenic emissions model. Seigneur (2001) indicated that significant uncertainties are associated with NH3 and biogenic volatile organic compounds (VOC)'s in emission inventories. This study explores these uncertainties by employing biogenic emissions obtained from two biogenic emissions models and using two anthropogenic emission inventories with order of magnitude differences in ammonia emissions.

The fifth generation Pennsylvania State/NCAR Meteorological Model (MM5) was used to simulate the meteorology for the period of July 3-16, 1996 with two domains; a parent 36 km domain and a 12 km domain encompassing Oregon, Idaho, Washington, and a significant portion of south western Canada. These emission inventories and meteorology were then used to drive the Community Multi-scale Air Quality (CMAQ) modeling system to simulate ozone and aerosol formation and transport in the Pacific Northwest and the associated impacts to visibility and regional haze.

This study is unique because previous particulate matter (PM) studies have focused on urban areas in the Northeastern U.S. and the Los Angeles Basin as summarized by Seigneur (2001). The Pacific Northwest offers a different situation because of the heavily urbanized areas situated in a domain dominated by forests, mountains, and agricultural areas. The complex terrain of the Pacific Northwest is illustrated in Figure 1. Urbanization exists in the forested area between the coast and western ridge of the Cascade Mountains along the Interstate highway 5 (I-5) corridor. The east side of the Cascade Mountains is less heavily populated and lies in the rain shadow of the Cascades, thus conditions are often hot and dry, especially in the summer months. The Columbia Plateau, spanning much of eastern Washington and south into Oregon, is an area rich in agriculture. The Rocky Mountains run North-South through Idaho, Montana, and Canada. The most populated area of Idaho is the Snake River valley looping through southern Idaho from east to west before turning north to make the border with Oregon. These terrain features and population distributions have

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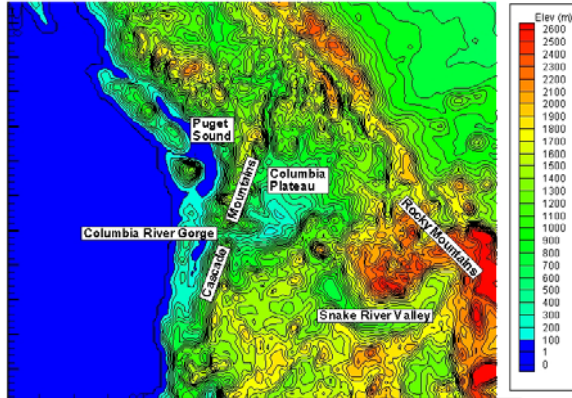implications for pollutant sources and the flow regimes through the domain.

Figure 1. Terrain Elevation of the Pacific Northwest.

The Pacific Northwest is also the home to several Class I wilderness areas. The Interagency Monitoring of PROtected Visual Environments (IMPROVE) program has been developed to study the components of visibility degradation in Class I wilderness areas. Malm et al. (1994), in their analysis of data from the IMPROVE program, indicated that SO4, organic carbon, and fine soil are the largest contributors to PM2.5 mass in the Pacific Northwest. Furthermore, organic aerosols can contribute 40%-45% to light extinction in the PNW.

The purpose of this paper is to present results describing aerosol formation in the PNW and compare CMAQ model output of aerosol species and visibility parameters with observational data from the IMPROVE network.

2.   RESULTS

As quantitative measures of model performance the following statistical measures were applied: normalized gross error, normalized bias, index of agreement, and correlation coefficient. The normalized gross error is calculated by:

$$Error = \frac{1}{N}\sum_{i=1}^{N}\frac{|p_i - o_i|}{o_i} \tag{1}$$

where p_i and o_i are the predicted and observed concentrations respectively for measurement i. N is the total number of measurements. The normalized bias is calculated by:

$$Bias = \frac{1}{N}\sum_{i=1}^{N}\frac{p_i - o_i}{o_i} \tag{2}$$

where a normalized gross error of 35% and a bias less than +/- 15% are suggested by EPA as indicative of acceptable ozone modeling performance.

The index of agreement is calculated by:

$$I = 1 - \frac{\sum_{i=1}^{N}(p_i - o_i)^2}{\sum_{i=1}^{N}(|p_i - \bar{o}| + |o_i - \bar{o}|)^2} \tag{3}$$

where $\bar{o}$ denotes the average observed ozone concentration and a value of 1 indicates perfect agreement between predicted and observed values. These various measures were applied to obtain a relative indication of how the two CMAQ solutions performed predicting aerosol specie concentrations, total aerosol mass, visibility parameters, and ozone concentrations.

Contour plots of PM2.5, across the PNW domain for the NWRMC EI solution and NET96 EI solution, are shown in Figure 2 for July 14, 1996 at 7 am PDT; a time when PM2.5 concentration maximums occurred. In the NWRMC EI solution (Figure 2a), the PM2.5 concentrations were below 20 μg/m^3 for much of the period, then concentrations increased to approximately 35 μg/m^3 toward the end of the modeled period in the Columbia River Gorge region, Central Idaho, and the north-slope of the Olympic Mountains in the Puget Sound region. The results for the NET96 EI solution (Figure 2b) were similar in trend to the NWRMC EI solution but PM2.5 concentrations were significantly lower with concentrations mostly below 10 μg/m^3 east of the Cascade Mountains. West of the Cascade Mountains, in the NET96 EI solution, the PM2.5 concentrations were mostly below 20 μg/m^3 with the exception of the Portland region where concentrations were up to 45 μg/m^3.

The highest PM2.5 concentrations occurred in three regions of the domain: the north slope of the Olympic Mountains, the Columbia River Gorge region, and the mountains of Central Idaho. These PM2.5 concentrations are attributable to organic aerosols, where in the Olympic and Columbia River Gorge regions the organics are anthropogenic and biogenic in origin, while in the mountains of Idaho the organics were biogenic in origin.

Statistical results comparing total PM2.5 mass from the two CMAQ simulations with IMPROVE observations are given in Table 1. All twelve of the IMPROVE sites had PM2.5 data. The CMAQ results were averaged for the 24 hour period corresponding to the IMPROVE sampling period. At the IMPROVE sites, the NWRMC EI solution tended to over-predict PM2.5 mass with a normalized bias of 93%, while the NET96 EI solution tended to under-predict PM2.5 with a normalized bias of –15%. Normalized gross error was 49% for the NET96 EI solution and 98% for the NWRMC EI solution for PM2.5. These solutions, while exhibiting a wide range of performance, are generally within the bounds of solutions reported by Seigneur (2001), Pagowski et al. (2002) and Chandrasekar et al. (2002).
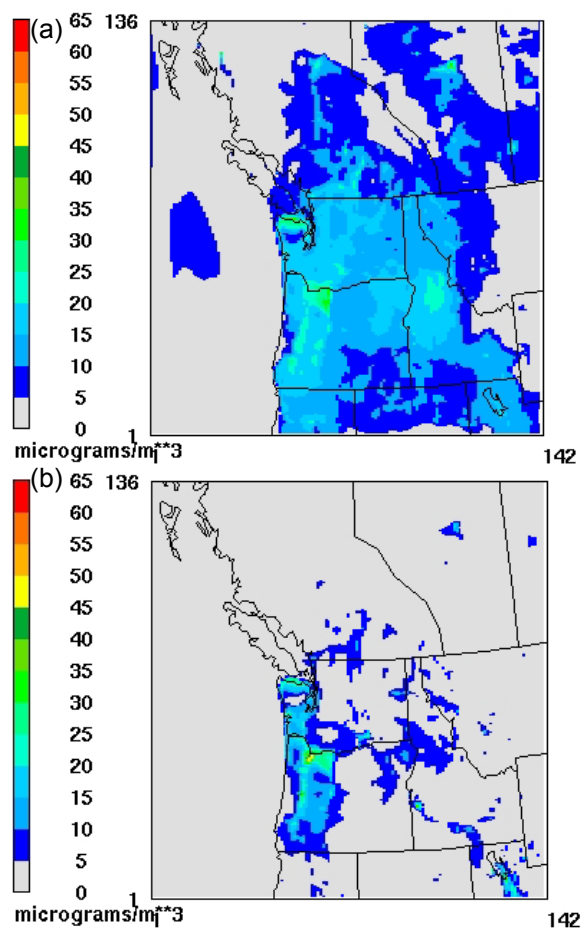
Figure 2. PM2.5 concentrations for July 14, 1996 at 7 am PDT from the (a) NWRMC EI and (b) NET96 EI solutions.

Statistical results comparing organic aerosol concentrations predicted by the two CMAQ simulations with IMPROVE observations of organic carbon (OC) are given in Table 2. Seven of the 12 IMPROVE sites had OC data available during the July 3-16, 1996 period. Overall, the NWRMC EI solution tended to over-predict OC with a normalized bias of 44% while the NET96 EI solution performed well with a normalized bias of –2%. The NET96 EI solution also performed better than the NWRMC EI solution for normalized gross error, but the NWRMC EI solution had the best correlation and index of agreement statistics.

Statistical results comparing the extinction coefficient predicted by the two CMAQ simulations with IMPROVE observations are given in Table 1. Six of the 12 IMPROVE sites had visibility data available during the July 3-16, 1996 period. The NWRMC EI solution performed better than the NET96 EI solution for all statistics. Both cases have a negative bias indicating that they under-predict visibility degradation.

Table 1. Statistical Comparison of the CMAQ NET96 EI Solution and the CMAQ NWRMC EI Solution with the 12 IMPROVE Sites (combined) for PM2.5.

	IMPROVE	NET96	NWRMC
Average (ng/m^3)	5200	4400	9300
Maximum (ng/m^3)	13400	17000	16800
Minimum (ng/m^3)	1900	1300	3700
Std Dev (ng/m^3)	2400	3500	3300
r^2		0.32	0.43
I		0.54	0.50
Bias *(%)*		-15	93
Error *(%)*		49	98

Table 2. Statistical Comparison of the CMAQ NET96 EI Solution and the CMAQ NWRMC EI Solution with the 12 IMPROVE Sites (combined) for Organic Carbon (OC).

	IMPROVE	NET96	NWRMC
Average (ng/m^3)	2200	2000	3000
Maximum (ng/m^3)	6700	4200	6500
Minimum (ng/m^3)	0	910	1300
Std Dev (ng/m^3)	1600	840	1400
r^2		0.57	0.62
I		0.69	0.81
Bias *(%)*		-2	44
Error *(%)*		35	54

Table 3. Statistical Comparison of the CMAQ NET96 EI Solution and the CMAQ NWRMC EI Solution with the 12 IMPROVE Sites (combined) for Extinction Coefficient.

	IMPROVE	NET96	NWRMC
Average *(1/Mm)*	31	11	29
Maximum *(1/Mm)*	64	38	49
Minimum *(1/Mm)*	12	1	15
Std Dev *(1/Mm)*	13	9	10
r^2		0.48	0.62
I		0.47	0.76
Bias *(%)*		-62	-1
Error *(%)*		62	29

3. SUMMARY AND RECOMMENDATIONS

This is the first step in a comprehensive performance evaluation of CMAQ for the Pacific Northwest. The NWRMC EI solution tended to over-predict PM2.5 concentrations with a normalized bias of 93% while the NET96 EI solution slightly under-predicted with a bias of –15%. Siegneur (2001) reported errors in the range of 50% for PM2.5, and the NET96 EI solution was in this range. Contour plots of PM2.5 for the NWRMC and NET96 EI solutions illustrated the differences between the two sets of results. At 7 am PDT on July 14, 1996, when PM2.5 concentrations reached a diurnal peak across the domain, the NWRMC EI solution maintained PM2.5 levels at or above 10 $\mu g/m^3$ while the NET96 EI solution predicted PM2.5 concentrations below 5 $\mu g/m^3$ for much of the domain east of the Cascade Mountains. Only in the western regions of Oregon and Washington were the PM2.5 contour plots similar.

Three areas of improvement and study are recommended. First additional model development should be undertaken to: improve the treatment of organics in the chemical mechanisms, incorporate NaCl chemistry, transfer mass from the accumulation mode to course mode for NH4, OC, and NO3 (Yeatman, 2001), and include crustal species (Seigneur, 2001).

Secondly, additional emission inventory development is necessary. The NWRMC provides an excellent basis from which EI enhancements can be made. Two CMAQ EI simulations were undertaken in this study with order-of-magnitude differences in the NH3 emissions and large differences in the biogenic emissions.

Finally, model sensitivity studies are necessary. NOX/VOC ratios should be investigated in relation to the interdependencies of PM and ozone formation. The NWRMC EI solution over-estimates PM2.5 yet underestimates visibility, therefore adding more PM2.5 to the model results would not necessarily be the correct way to improve the visibility prediction. This needs to be investigated – Is there a better way to calculate visibility? Could organics be making a larger impact on visibility degradation? Malm et al. (1994) accounts for the hygroscopicity of organics in the calculation of the extinction coefficient. Application of the source footprint modeling system (O'Neill et al., 2002) is also necessary to investigate the upwind source area of aerosol precursor emissions.

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