

**Emissions-Scenario Simulations of Potential Sulphur-Content
Reductions for Heavy Fuel Oils and Light Fuel Oils Using the Acid
Deposition and Oxidant Model**

Contract No. KM155-01-0225

Prepared for:

Meteorological Service of Canada
2121 Trans-Canada Highway
Dorval, Québec, H9P 1J3

Scientific Authorities:

Dr. Michael D. Moran
Air Quality Research Branch
Meteorological Service of
Canada
Environment Canada
4905 Dufferin Street
Toronto, Ontario, M3H 5T4

Mr. Michel Jean
Operations Branch
Meteorological Service of
Canada
Environment Canada
2121 Trans-Canada Highway
Dorval, Québec, H9P 1J3

Ms. Lyne Monastesse
Senior Program Engineer
Oil, Gas and Energy Branch
Environment Canada
Place Vincent Massey
351 St. Joseph Blvd.
Hull, Québec, K1A 0H3

Prepared by:

Dr. Jacek W. Kaminski
ARM Consultants
2161 Yonge Street, Suite 808
Toronto, Ontario, M4S 3A6

January 31, 2002

Table of Contents

1. INTRODUCTION	5
2. SCENARIO DESCRIPTIONS	8
3. SCENARIO IMPLEMENTATION	11
4. ADOM SIMULATION	15
5. ADOM EPISODE AGGREGATION AND POST-PROCESSING	16
6. RESULTS.....	19
7. DISCUSSION AND ANALYSIS.....	26
8. CONCLUSIONS.....	27
9. ACKNOWLEDGMENTS	27
10. REFERENCES	28
APPENDIX A – SCENARIO SCALING FACTORS.....	31
APPENDIX B – POINT-SOURCE-SPECIFIC EMISSIONS VALUES	31
APPENDIX C – SO₂ EMISSION LEVELS FROM HFO AND LFO	33
APPENDIX D – LITERATURE REVIEW AND GUIDE TO ADOM LEGACY.....	35

List of Tables

Table 1. Total SO ₂ emissions within ADOM domain for 1989 base case and sixteen future-year emission scenarios.....	10
Table 2. Eastern Canadian land area in exceedance of wet SO ₄ critical load for ADOM CCUSA2 and HLFO SO ₂ emission scenarios.....	26
Table 3. Eastern Canadian land area (in 1,000 km ²) in exceedance of wet SO ₄ critical load for ADOM CCUSA2 and HLFO SO ₂ emission scenarios for different thresholds.....	26
Table A1. Scenario scaling factors used in ADOM scenario implementation for Scenario HLFO.....	31
Table C1. SO ₂ emission levels based on production of HFO and LFO per region.	33
Table C2. SO ₂ emission levels based on consumption of HFO and LFO per region.	33

List of Figures

Figure 1. Plot of the jurisdictional mask used for emissions scaling.	12
Figure 2. Plots of SO ₂ emissions fields in units of Ktonnes yr ⁻¹ : (a) Scenario HLFO; (b) Scenario CCUSA2.	13
Figure 3. Plot of difference between 2010 scenario (“CCUSA2”) and Scenario HLFO SO ₂ emissions fields (Ktonnes SO ₂ yr ⁻¹).	14
Figure 4. Plot of wet-sulphate-deposition critical-load field for Canada at 5% lake-sacrifice level in units of kg SO ₄ ha ⁻¹ yr ⁻¹ (from Environment Canada [1997] and based on RMCC [1990]).	18
Figure 5. Plots of calibrated wet SO ₄ annual deposition patterns in units of kg SO ₄ ha ⁻¹ yr ⁻¹ for (a) HLFO and (b) CCUSA2 ADOM SO ₂ emissions scenarios.	20
Figure 6. Plot of difference between wet SO ₄ annual deposition fields (units of kg SO ₄ ha ⁻¹ yr ⁻¹) for CCUSA2 and HLFO SO ₂ emission scenarios.	20
Figure 7. Same as Figure 6, but for percentage difference (%).	21
Figure 8. Plots of wet SO ₄ deposition critical-load exceedance fields (units of kg SO ₄ ha ⁻¹ yr ⁻¹) (a) HLFO and (b) CCUSA2 ADOM SO ₂ emissions scenarios.	22
Figure 9. Plot of difference of wet SO ₄ deposition critical-load exceedance fields (units of kg SO ₄ ha ⁻¹ yr ⁻¹) for CCUSA2 and HLFO SO ₂ emission scenarios.	22
Figure 10. Plots of ambient near-surface SO ₄ annual air-concentration patterns in units of μg SO ₄ m ⁻³ for (a) HLFO and (b) CCUSA2 ADOM SO ₂ emissions scenarios.	23
Figure 11. Plot of difference between ambient SO ₄ annual fields (units of μg m ⁻³) for the CCUSA2 and HLFO SO ₂ emission scenarios.	24
Figure 12. Plot of percentage difference (%) between ambient SO ₄ annual fields for the CCUSA2 and HLFO SO ₂ emission scenarios. The map was enlarged over the impacted area.	25

1. Introduction

In spite of significant reductions of emissions of acidifying gases in Canada and the United States in recent years due to control programs in both countries, the impacts of acid deposition in eastern Canada have not been eliminated or reduced to acceptable levels. Environment Canada has identified a requirement to carry out a new emission-scenario simulation with the Acid Deposition and Oxidant Model (ADOM) to determine the impact of a reduction of the sulphur content in heavy fuel oils and light fuel oils across eastern Canada.

Project Objective

The primary objective of this project was to investigate the impact of implementing European sulphur limits on heavy fuel oils (HFOs) and light fuel oils (LFOs). Sixteen emissions scenarios had already been run with ADOM in previous modeling projects. Results from the current model simulation combined with earlier ADOM results (see Environment Canada [1997], AETG [1997], and ARM [2000]) are expected to provide additional insight into the extent to which controls of sulphur content in HFO and LFO in Canada could contribute to achievement of critical-load target for aquatic systems across eastern Canada and to reduce ambient fine-particulate loading. More specifically, changes in annual wet SO_4 deposition, annual near-surface SO_2 and SO_4 air concentrations, and critical-load exceedances will be determined. In the context of this study, concentration of sulphates will be used as a surrogate for $\text{PM}_{2.5}$ over eastern Canada.

Project Execution

The project was carried out in three main steps, comprised of (a) scenario preparation; (b) model execution; and (c) results aggregation and analysis.

Emission-scenario preparation

This task was coordinated by the Scientific Authorities (SA) with initial participation by personnel from the Oil, Gas and Energy Branch of Environment Canada to ensure that all required software and quality-control (QC) protocols and the descriptions of emissions scenarios were in place. In order to implement the required emission reduction option (scenario) in ADOM, FORTRAN code modifications were introduced to the ADOM emission input/output (I/O) subroutine "GETEMS" (to implement jurisdiction-specific base-case and scenario scaling masks) and to emission I/O subroutine "RATEMS" (to implement changes to

individual point-source emission rates). These code changes are described in more detail in Section 3, and ADOM itself is described in more detail in Section 4.

The impact of these coding changes was verified by the SA. Quality control was performed both by inspecting the modified computer code and by examining changes in total SO₂ emissions between the ADOM domain-mass-budget output files for the new scenario and the reference scenario CCUSA2 (see section 2 for definition) from the previous study carried out by Environment Canada.

ADOM simulations

As outlined in Sections 4 and 5, in order to estimate annual fields, ADOM time integrations were carried out for each of 33 three-day aggregation episodes for the new emissions scenario. Predicted flux, concentration, and domain-mass-budget files were extracted from the primary model output files and archived on the Meteorological Service of Canada (MSC) Central File Server (CFS) in Montréal in the same way as was done for previous ADOM SO₂ future-year emission-scenario runs (see Environment Canada, 1997). To ensure consistency and comparability between the new scenario run and the previous ADOM emissions-scenario runs, the ADOM domain, spatial and temporal resolution, and aggregation episode set used in the present project were identical to those used for the earlier scenario runs. The model source code used was also identical, with the exception of the modifications needed to subroutines “GETEMS” and “RATEMS” to implement the new scenario.

Aggregation of ADOM results and other ADOM post-processing and analysis

This task was carried out with the initial participation of the SA to ensure that the required software and QC protocols were in place. Post-processing of hourly ADOM output flux and surface concentration fields was performed to generate daily totals, then three-day episode totals, and then annual fields using episode-aggregation weights (see Section 5 and Environment Canada [1997] for details).

The environmental measures used to evaluate the impact of the emissions control scenario were the ADOM-predicted future-year sulphate deposition fields and the annual sulphate air concentration field. Wet sulphate (SO₄) deposition field predicted by ADOM for the current SO₂ emissions scenario was compared against the eastern Canadian sulphate-deposition critical-load field, and areas with critical-load exceedances, that is, areas with deposition above critical load, were deemed to be in excess of ecological carrying capacity. The ambient sulphate concentration field from the current scenario was compared to the results from the CCUSA2 scenario which was selected as a reference scenario for the current study.

Report Organization

The new SO₂ future-year emissions scenario for eastern North America that has been investigated with ADOM is described in Section 2 and its implementation in ADOM is described in Section 3. Some details about the ADOM model and ADOM simulations are provided in Section 4. Section 5 describes the post-processing of the ADOM simulation results, including the use of episode aggregation to estimate annual fields from a set of short-term model results and the critical-load-exceedance calculations. Results from the new ADOM SO₂ emissions scenario is then presented in Section 6, and these results are discussed and analyzed further in Section 7. Finally, some conclusions are given in Section 8.

2. Scenario Descriptions

In terms of domain-scale and distribution of SO₂ emissions on the ADOM domain the CCUSA2 scenario was selected as a reference scenario for the current study. The CCUSA2 scenario corresponds to the actual legislated levels expected in 2010 with voluntary overcompliance from some major point sources (further described in Environment Canada, 1997, Section 3.4.1.1).

The starting point for the Canadian SO₂ emissions for the CCUSA2 scenario run with ADOM were the provincial SO₂ emission caps and voluntary overcompliance agreed to under the Eastern Canada Acid Rain Control Program (e.g., Tables 1 and 2 of Environment Canada, 1998). The starting point for the U.S. SO₂ emissions used for the CCUSA2 scenario were the Phase 2 caps and limits set under Title IV of the 1990 U.S. Clean Air Act Amendments, assuming no geographic redistribution through emissions trading.

Emission fields used in the CCUSA2 scenario were then modified for the new scenario (designated as HLFO) in order to achieve the required reduction of 180 Ktonne yr⁻¹ of SO₂ from eastern Canadian sources, which represents approximately a 9% reduction from the Canadian portion of the ADOM model domain as compared to the CCUSA2 scenario. However, this is a relatively small reduction overall – approximately 1% of the total emissions over the model domain as compared to the CCUSA2 scenario (see Table 1).

Canadian and U.S. SO₂ emission totals on the ADOM domain (see Figure 1) for the new HLFO scenario, together with those for all other emission reduction scenarios prepared using ADOM, are given in Table 1. The previous ADOM scenarios are described in Environment Canada (1997), AETG (1997), and ARM (2000) reports.

SO₂ emission levels from HFO and LFO combustion used to define the current scenario are summarized in Appendix C. From Table C2, the estimated reductions in SO₂ emissions

associated with the HFO S-content limit in the Maritimes, Quebec, Ontario, and the Prairies are 108, 21, 29, and 0 kT y⁻¹, respectively. The corresponding estimated reductions in SO₂ emissions associated with the LFO S-content limit in the Maritimes, Quebec, Ontario, and the Prairies are 3, 7, 5, and 0 kT y⁻¹, respectively. Overall, emissions would decrease by 174 kT y⁻¹ in eastern Canada: 64% in the Maritimes, 17% in Quebec, 19% in Ontario, and a negligible amount in the Prairies.

The consumption-based analyses to estimate the SO₂ emission reductions shown in Table C2 were carried out on a regional basis using numbers supplied by Statistics Canada. In order to apportion these reductions to the provincial level, it was necessary to obtain additional information about the use of HFO and LFO in Canada by province.

One of the SAs (M. Moran) carried out an analysis of HFO/LFO usage in Canada by province and source category using the 1990 Canadian national criteria-air-contaminant (CAC) inventory (see Deslauriers, 1996). He found the inventory to contain emissions from HFO combustion for 13 point-source categories and three area-source categories and emissions from LFO combustion for 15 point-source categories and three area-source categories. In 1990, Newfoundland and Labrador accounted for 22% of the 184 kT y⁻¹ of SO₂ emissions associated with HFO and LFO combustion in the Maritimes, Prince Edward Island accounted for 2%, Nova Scotia accounted for 20%, and New Brunswick accounted for 56%. This geographic distribution was used to pro-rate the reduction in SO₂ emissions given in Table C2 for the Maritimes to these four individual provinces.

Based on Table C2 and this provincial apportionment for the Maritimes, the HFO/LFO emissions scenario for ADOM assumed that SO₂ emissions should be reduced by 24.5 kT y⁻¹ in Newfoundland and Labrador, by 24.5 kT y⁻¹ in Nova Scotia and PEI, by 62.3 kT y⁻¹ in New Brunswick, by 28.8 kT y⁻¹ in Quebec, and by 33.6 kT y⁻¹ in Ontario, and 0 kT y⁻¹ in Manitoba and Saskatchewan, for a total of 173.7 kT y⁻¹ across the ADOM domain (see Figure 1). The implementation of these emission changes in ADOM is discussed in the next section.

Table 1. Total SO₂ emissions within ADOM domain for 1989 base case and sixteen future-year emission scenarios.

Scenario Name	Nominal Year	SO ₂ Emissions (Ktonnes yr ⁻¹)			Fraction of BASE	Strategy	Comments
		Canada	U.S.	Total			
BASE	1989	2,688	17,511	20,199	1.00	Current	Base case, effectively 1988-90 average
CCONLY	1994	1,939	17,862	19,801	0.98	Sectoral	Cdn controls (CC) + overcompliance, new US srcs
CCUSA1	1997	1,939	14,865	16,804	0.83	Sectoral	US CAAA Phase 1 cuts, Canada same as 1994
CCUSA2	2010	1,939	12,446	14,385	0.71	Sectoral	US CAAA Phase 2 cuts, Canada same as 1994
5CONLY	2030	1,320	12,446	13,766	0.68	Regional Rollback	50% Cdn SOMA red'ns from caps, US same as 2010
25FCAP	2030	1,738	9,335	11,072	0.55	Uniform Rollback	25% Cdn red'ns from caps, 25% US from 2010
T5CUS2	2030	1,277	8,794	10,071	0.50	Regional Rollback	Same as "5CCUS2" except Saskatchewan, Manitoba, Newfoundland, and US SE, SW, and W left at 2010 levels
5CCUS2	2030	969	6,223	7,192	0.36	Uniform Rollback	50% cut from "CCUSA2" 2010 scenario
75FCAP	2030	579	3,112	3,691	0.18	Uniform Rollback	75% Cdn red'ns from caps, 75% US from 2010
PST2010A	2030	1,636	5,578	7,214	0.36	Regional Rollback	50% ON, 45% QU, 25% NB + NS from caps, 55% US from 2010
PST2010B	2030	1,636	4,967	6,603	0.33	Regional Rollback	50% ON, 45% QU, 25% NB + NS from caps, 60% US from 2010
PST2010C	2030	1,610	6,204	7,814	0.39	Sectoral/Reg. Rollbk	50% ON but major sources targetted, 50% QU but Noranda Rouyn targetted, 25% NB + NS from caps, 50% US from 2010
PST2010D	2030	1,601	8,116	9,717	0.48	Sectoral/Reg. Rollbk	Same as "PST2010C" except 30% NB and NBPC redistribution, 25% NS but NSPC redistribution, and 60% red'n in 20 NE US states but rest at 2010 levels
PST2010E	2030	1,601	4,984	6,585	0.33	Sectoral/Reg. Rollbk	Same as PST2010D except all U.S. states reduced by 60% from 2010
PST2010a	2030	1,690	5,578	7,268	0.36	Regional Rollback	50% ON, 34% QU, 25% NB + NS from caps, 55% US from 2010
PST2010b	2030	1,690	4,967	6,657	0.33	Regional Rollback	50% ON, 34% QU, 25% NB + NS from caps, 60% US from 2010
HLFO	2010	1,759	12,446	14,205	0.70	Sectoral	Reductions in Canadian heavy and light fuel oil sulphur content

3. Scenario Implementation

In order to implement the SO₂ emissions scenario described in the previous section, major-point-source emissions records and area-source gridded emissions fields used in the ADOM 2010 scenario “CCUSA2”, described in Environment Canada (1997) and in AETG (1997), were used as a starting point for the current modeling work.

ADOM separates emissions sources into two main categories: “major point sources” and “area sources” (see also Section 4). Major point sources include such sources as power stations and non-ferrous smelters and usually emit from large smokestacks. These sources are treated individually by ADOM, including a calculation of plume rise. Area sources, on the other hand, are associated with ADOM grid cells and in effect are the aggregate of all small sources located within a grid cell; they are assumed to emit at the surface.

Two electricity generating stations in New Brunswick (Courtenay Bay and Coleson Cove) burn bunker C heavy fuel oil and one (Dalhousie) burns Orimulsion, a natural bitumen emulsified in water using a surfactant (Robert Hughes, New Brunswick Department of the Environment, personal communication to Michael Moran, 11 October 2001). One electricity generating station in Nova Scotia (Tufts Cove) uses fuel switching and burns both HFO and coal (Michael Hingston, Nova Scotia Department of Environment and Labour, personal communication to Michael Moran, 11 October 2001).

All four of these generating stations are contained in ADOM’s emission files as major point sources, so it was possible to consider reductions to their SO₂ emissions individually. On the other hand, no significant Ontario or Quebec sources using HFO or LFO as fuel are contained in the ADOM major-point-source file.

Table 2.5 of Tushingham and Bellamy (2001) gives a value of 2.2% for the current average S content of HFO in the Maritimes and an expected average value of 0.8% for the future-year 1%-limit scenario. This 8:22 ratio was used to estimate the reduction in SO₂ emissions from the four Maritimes generating stations from fairly recent (1985) emission levels due to the substitution of low-S HFO. In all but one case (Dalhousie GS), the estimated reduction was larger than the level of SO₂ emissions assumed for these sources in the reference ADOM emission scenario (“CCUSA2”). As a consequence, emissions from these four sources were reduced to the extent possible by manipulating ADOM major-point-source emissions records upon input for selected sources as identified by record ID. Existing SO₂ emission values were replaced by scenario-specific values for the HLFO scenario. The list of point sources and scaling factors is given in Appendix B. The remaining emission reductions for each province were then obtained by scaling the area-source SO₂ emissions province by province.

The area-source scaling factors used for each province are given in Table A1. The desired reductions were 24.5 kT y⁻¹ in Newfoundland and Labrador, 24.5 kT y⁻¹ in Nova Scotia and PEI, 32.3 kT y⁻¹ in New Brunswick, 28.8 kT y⁻¹ in Quebec, and 33.6 kT y⁻¹ in Ontario, and 0 kT y⁻¹ in Manitoba and Saskatchewan. New Brunswick was a special case in that total SO₂ emissions from New Brunswick area sources for the ADOM “CCUSA2” scenario were only 22 kT y⁻¹. This mismatch was addressed by setting New Brunswick area-source SO₂ emissions to zero and scaling SO₂ emissions from the Dalhousie Generating Station point source back by 10.3 kT y⁻¹ more than might be expected for that point source due to switching to low-S HFO (see Table B1).

All of the scaling factors used are listed in Appendix A. The jurisdictional mask composed of ADOM 127 km by 127 km grid cells is shown below in Figure 1 superimposed on the ADOM modelling grid.

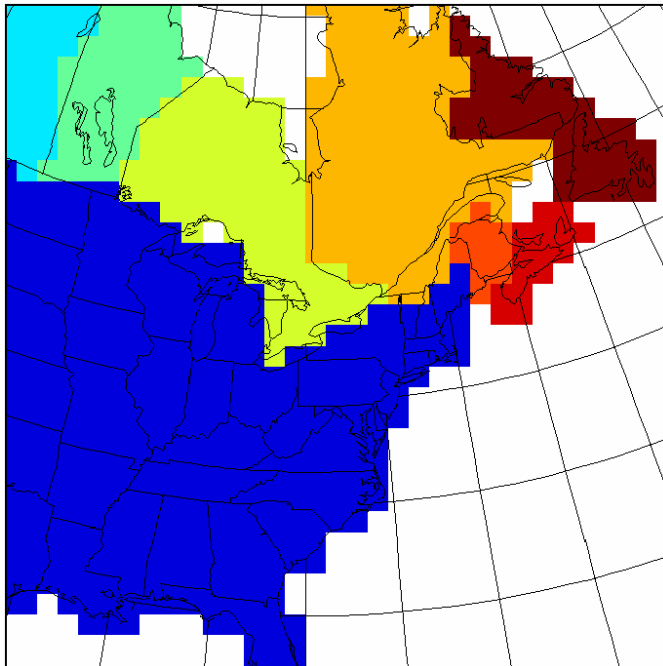


Figure 1. Plot of the jurisdictional mask used for emissions scaling.

The distribution of SO₂ emissions on the ADOM eastern North America grid for the HLFO scenario and for the CCUSA2 scenario are shown in Figure 2a and 2b, respectively.

The difference in SO₂ emissions between HLFO scenario and the ADOM 2010 scenario (“CCUSA2”) described in Environment Canada (1997) and AETG (1997) is shown in Figure 3. The latter scenario corresponds to the “base case” for the present study.

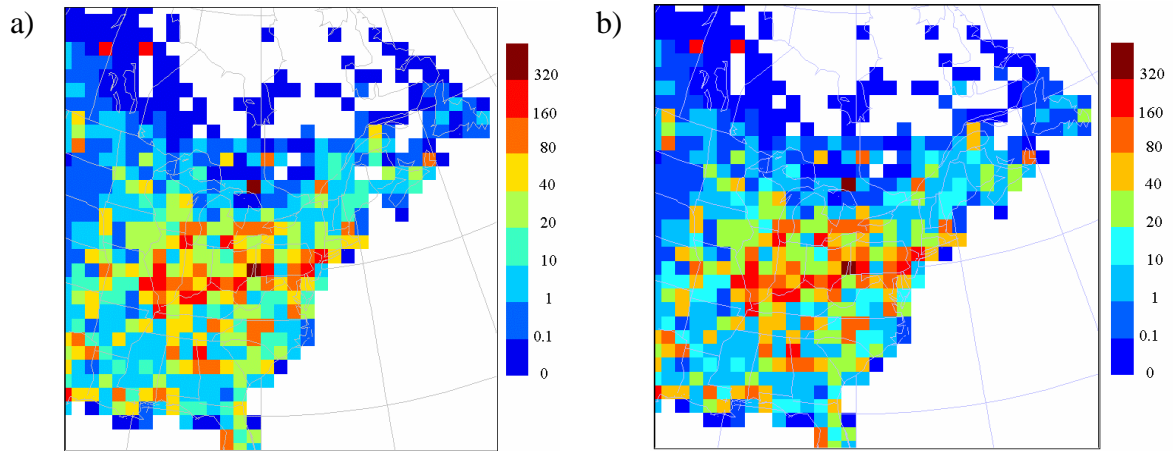


Figure 2. Plots of SO₂ emissions fields in units of Ktonnes yr⁻¹: (a) Scenario HLFO; (b) Scenario CCUSA2.

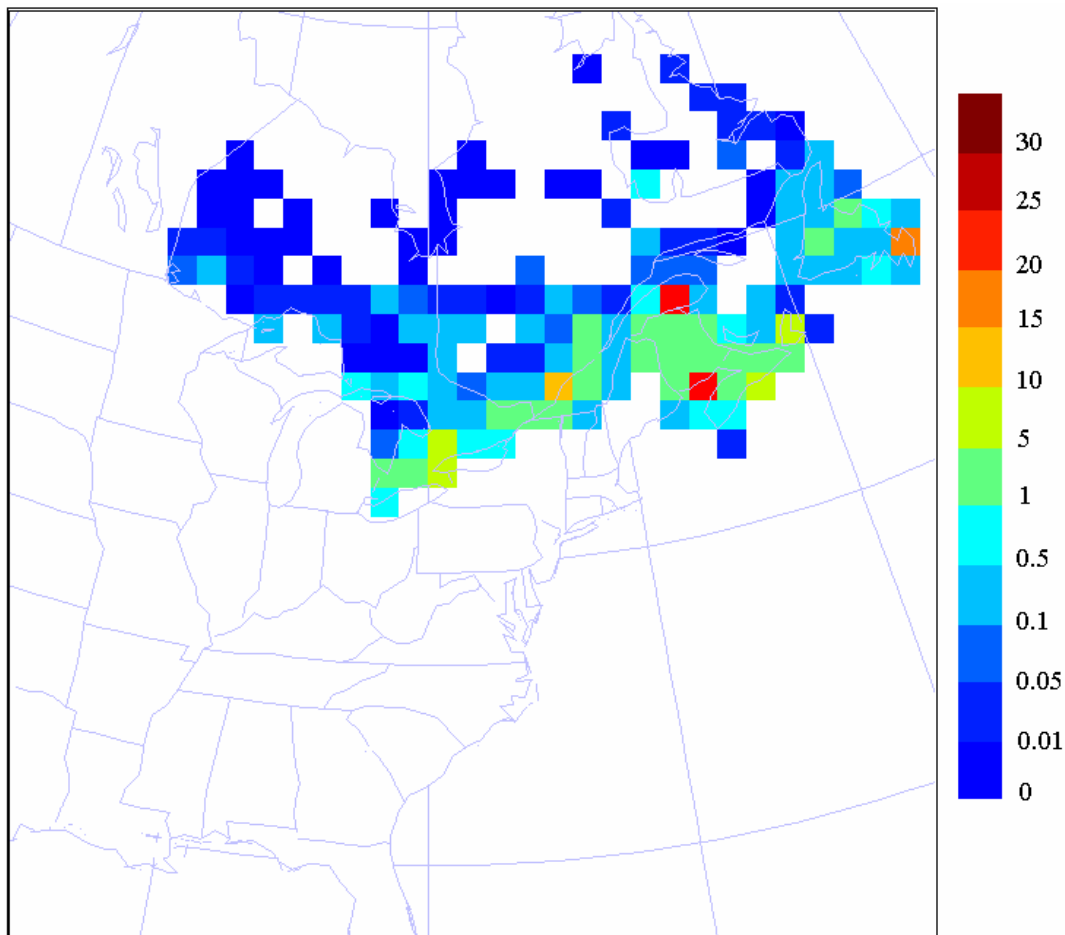


Figure 3. Plot of difference between 2010 scenario (“CCUSA2”) and Scenario HLFO SO₂ emissions fields (Ktonnes SO₂ yr⁻¹).

4. ADOM Simulation

Emission scenario simulation for this project was carried out with the ADOM episodic Eulerian chemical transport model (e.g., Venkatram et al., 1988; Misra et al., 1989; Fung et al., 1991; Padro et al., 1991; Environment Canada, 1997). Version two of the ADOM chemical mechanism has been used in this project. Significant differences between version two of the ADOM mechanism (ADOM-II) and the original Lurmann et al. (1986) mechanism include an explicit treatment of atomic oxygen, $O(^3P)$ and $O(^1D)$, and the treatment of isoprene as a separate species. The ADOM-II mechanism is comprised of 47 species, 98 chemical reactions, and 16 photolysis reactions.

Anthropogenic emissions for 18 ADOM chemical species, including SO_2 and SO_4 , have been taken from a set of model files of pre-processed emissions originally developed for the Canadian air-quality modelling community by ORTECH Inc., The MEP Co., and Sophos Inc. in the late 1980s and early 1990s. The emissions files are partitioned into major-point-source emissions files, which contain records for roughly 3000 large individual sources, and other-sources emissions files. The ADOM emissions files are based on the NAPAP emissions inventory for 1985 (U.S. EPA, 1989) with the exception of those major SO_2 point sources in Canada and the United States that are subject to SO_2 emissions control legislation. These sources have been set to their capped limits in Canada or to their EPA allowances in the case of U.S. power plants in the major-point-source emissions files (see Environment Canada, 1997). Note that the 32 NAPAP VOC lumped species have been “re-lumped” using reactivity weighting to obtain the 11 ADOM VOC lumped species.

ADOM is an episodic regional air-quality model that predicts hourly concentration and deposition fields. Since ADOM was not designed or intended to be used for long-term simulations, the episode aggregation technique as described in Environment Canada (1997) has been used to obtain estimates of annual fields from a set of ADOM simulations spanning a period of less than a year. Accordingly, ADOM integrations were carried out for each of the emissions scenarios for a set of 33 three-day aggregation episodes. These episodes were drawn from the summer, autumn, and early winter of 1988 and the late winter and spring of 1990. Predicted flux, surface concentration, and domain-mass-budget files were extracted for each episode day and archived at the Canadian Meteorological Centre in Montréal.

In total, the model was executed for 116 model days, where only 82 model days were considered to be “episode” days. The other days were “spin-up days” at the beginning of an episode, during which time the model is reaching a balance between emissions and removal processes: that is, each aggregation episode consists of 2 spin-up days followed by three episode days. Although the use of 33 episodes might suggest that 165 days need to be simulated in total, there is some overlap between some episodes so that fewer days (116) actually must be simulated.

Of these 116 simulation days, 91 days were run in two 12-hour steps and 25 days were run in four 6-hour steps, for a total of $91*2+25*4 = 282$ separate model runs. This run configuration was devised for an older NEC SX-3 multi-processor vector supercomputer but was retained for this project, where ADOM was executed on a NEC SX-4 multi-processor vector supercomputer. Each model day required approximately 1400 seconds of CPU time, for a total execution time on the NEC of approximately 50 CPU hours. On a time-sharing computer like the NEC, this translates to 7-14 days of “wall-clock time”, depending on system load and restart occurrences.

The main model output files were archived for each scenario. The total size of the archive is 400MB per scenario, and it consists of 82 flux files, 82 concentration files, and 202 mass diagnostic files (a total of 366 files). In addition, there are about 60MB of intermediate files produced by the aggregation process (see next section).

5. ADOM Episode Aggregation and Post-Processing

Concept and Purpose of Episode Aggregation

Episode aggregation is a semi-empirical approach that utilizes a set of short-term ADOM simulations to estimate concentration and deposition fields for annual and longer time periods. The aggregation approach was originally developed to provide a means for using the U.S. EPA’s Regional Acid Deposition Model to address policy questions and to support effects research (Brook et al., 1995a,b). It is based upon two main concepts: (a) that at any given location in eastern North America, wet deposition is determined by a number of deposition events belonging to a set of recurring weather patterns; and (b) that, if a series of deposition events representative of these different patterns can be identified, they can be combined or aggregated to produce a realistic estimate of the annual deposition total. Furthermore, observations of the frequency of occurrence of these recurring weather patterns and the mean precipitation amount and pollutant concentrations associated with them can be used to devise a weighting scheme to improve the long-term aggregate model estimates.

In practice, the two main components of this method are the selection of a representative set of three-day weather “events” for model simulation and the weighted aggregation of the model-predicted deposition totals associated with these events to estimate annual totals. See Environment Canada (1997) for details on the selection of the 33 ADOM aggregation episodes and the estimation of the ADOM aggregation-episode weighting factor fields.

Limitation In Areal Extent of Aggregated Fields

Episode aggregation is semi-empirical (i.e. partly dependent on data) because the weighting factor fields needed to combine ADOM episode predictions are determined from meteorological and air-quality measurements at stations. In order to create gridded weighting factor fields from scattered station measurements, a horizontal interpolation technique called kriging was used (e.g., Finkelstein, 1984; Federov, 1989; Schaug et al., 1993). The only limitation of this approach is that kriging can only estimate values within the confines of the convex hull (the area that is defined by drawing connecting lines between every pair of stations and then deleting all interior lines) determined by the geographic distribution of measurement stations on the periphery of the measurement network. Hence the spatial coverage of the ADOM-aggregated annual fields is constrained by the spatial coverage of the historical factor fields determined from kriged station measurements, even though the “raw” ADOM-predicted fields cover the entire ADOM domain (e.g., Figure 2a vs. Figure 5a).

Hour-To-Day-To-Episode-To-Annual Processing

ADOM outputs hourly fields of concentrations and depositions. A set of scripts is then used to produce first a set of daily fields from ADOM hourly output fields, then a set of 3-day episode fields from three sets of daily fields, and finally, using the gridded weighting factor fields described above, a set of annual concentration and deposition fields is produced by weighted averaging. The aggregation equations used for this last step are described in Environment Canada (1997).

Model Performance and Wet Deposition Calibration

ADOM performance has undergone extensive evaluation, both operational and diagnostic, and has been discussed in a number of publications, including Macdonald et al. (1993), Li et al. (1994), Moran (1998), and Environment Canada (1997). As reported in the last document (p. 2-180), for a comparison of ADOM predictions of annual wet SO₄ deposition in eastern North America against station measurements for the period 1986-90, “... the linear regression line has a slope of 1.00, a y-intercept of 0.3 kg ha⁻¹ (i.e., a slight positive model bias), and an R² correlation coefficient of 0.73. Only two points lie outside the factor-of-2 lines.” This is a very good result relative to the performance of other regional acid deposition models.

In addition, following Environment Canada (1997), an adjustment was performed in order to minimize the impact of statistical fluctuations resulting from the relatively small number of episodes sampled to represent the full range of meteorological conditions contributing to the long-term transport and chemical climatology of eastern North America. The ADOM annual

wet SO_4^- deposition field estimated by aggregation for each future-year scenario was multiplied by the ratio of the observed 1986-1990 mean annual wet SO_4^- deposition field to the predicted annual wet SO_4^- deposition field for the ADOM 1989 base-case simulation (a procedure equivalent to multiplying the observed field by the ratio of the future-year to base-case predicted fields). This procedure has the advantage of making use of available observations to augment model predictions. It also emphasizes ADOM's prediction of relative changes as opposed to absolute changes for different scenarios. It is worth noting that U.S. EPA guidance for ozone and PM attainment modelling recommends this same approach, and refers to the ratio of model-predicted scenario to model-predicted base case as relative-reduction-factor fields.

Critical-Load Exceedance Calculation

As noted in Section 1, the environmental measure used to evaluate ADOM-predicted future-year deposition fields for sustainability was the sulphate-deposition critical-load field for eastern Canadian aquatic ecosystems developed during the 1990 acid-deposition science assessment (RMCC, 1990). The sulphate-deposition critical-load field for eastern Canada is shown in Figure 4 plotted on a 42.3-km-by-42.3-km, grid (cf. Fig. 3.1 of Volume 2 of the 1997 Canadian Acid Rain Assessment report [Environment Canada, 1997]).

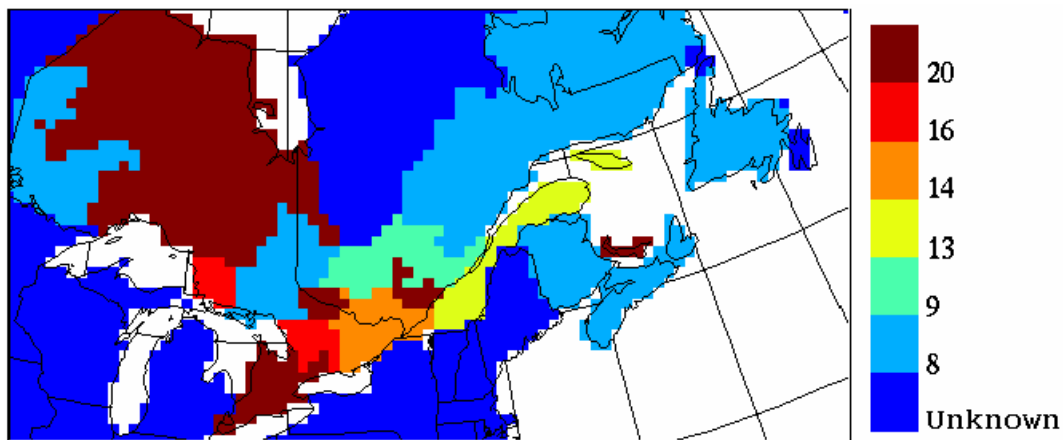


Figure 4. Plot of wet-sulphate-deposition critical-load field for Canada at 5% lake-sacrifice level in units of $\text{kg SO}_4 \text{ ha}^{-1} \text{ yr}^{-1}$ (from Environment Canada [1997] and based on RMCC [1990]).

To identify areas with wet SO_4 deposition critical-load exceedances, that is, deposition values larger than critical load, the predicted scenario annual wet SO_4 deposition field (in units of $\text{kg SO}_4 \text{ ha}^{-1} \text{ yr}^{-1}$) on the ADOM 127-km-by-127-km grid was first horizontally interpolated to the higher-resolution, 42.3-km-by-42.3-km critical-load grid by bilinear interpolation. The critical-

load field was then subtracted from the scenario wet SO₄ deposition field for each scenario. Any positive values correspond to areas where the wet SO₄ deposition is predicted to remain higher than the critical load for the same area.

Note that the impact of acid deposition depends on total deposition, that is, on the sum of both wet and dry deposition. Dry deposition is still a much more difficult quantity than wet deposition to measure. In developing the critical-load field a decade ago, dry deposition was accounted for in a crude way by dividing the critical-load SO₄ total-deposition value by 1.15, based on the assumption that dry SO₄ deposition is equal to 15% of wet SO₄ deposition (e.g., Jeffries et al., 2000). Note, too, that a lake sacrifice level of 5% was assumed in calculating these aquatic-ecosystem-based critical-load values; that is, the critical-load value is the total SO₄ deposition that can be sustained or tolerated by 95% of area lakes.

6. Results

A plot of a calibrated wet SO₄ annual deposition patterns is shown in Figure 5. The first panel shows the predicted patterns corresponding to the current HLFO scenario, whereas the second panel shows the wet SO₄ annual deposition pattern for the ADOM 2010 scenario (“CCUSA2”: see Table 1). The CCUSA2 scenario serves here as a nominal “base” scenario, since the new scenario considers further emission reductions relative to 2010 emission levels

Figure 6 shows the difference between the calibrated wet SO₄ annual deposition pattern for the ADOM 2010 (or “CCUSA2”) scenario and the current HLFO ADOM scenario.

Figure 7 shows the percentage difference between the calibrated wet SO₄ annual deposition pattern for the ADOM 2010 (“CCUSA2”) scenario and the current HLFO ADOM scenario.

Figure 8 shows the wet SO₄ deposition critical-load exceedance fields or critical-load “gaps” for eastern Canada in units of kg SO₄ ha⁻¹ yr⁻¹. Those regions of eastern Canada predicted to be in exceedance for the new emissions scenario have total SO₄ depositions greater than can be neutralized (as noted in Section 5, dry deposition has also been taken into account in a rough way in developing Figure 4). Negative numbers indicate deposition values less than the critical-load plotted in Figure 4, that is, sustainable acid deposition level.

The difference between wet SO₄ deposition critical-load exceedance fields (units of kg SO₄ ha⁻¹ yr⁻¹) for CCUSA2 and HLFO SO₂ emission scenarios is shown in Figure 9.

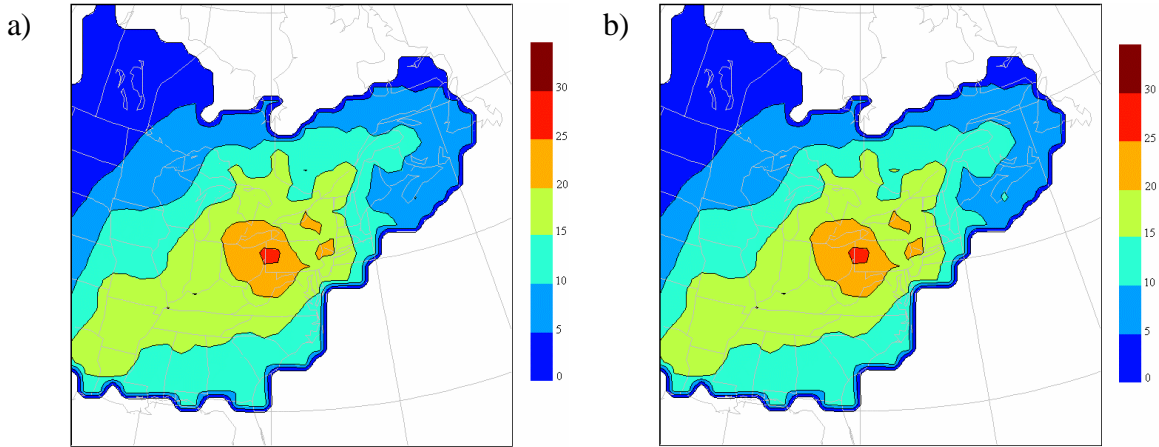


Figure 5. Plots of calibrated wet SO₄ annual deposition patterns in units of kg SO₄ ha⁻¹ yr⁻¹ for (a) HLFO and (b) CCUSA2 ADOM SO₂ emissions scenarios.

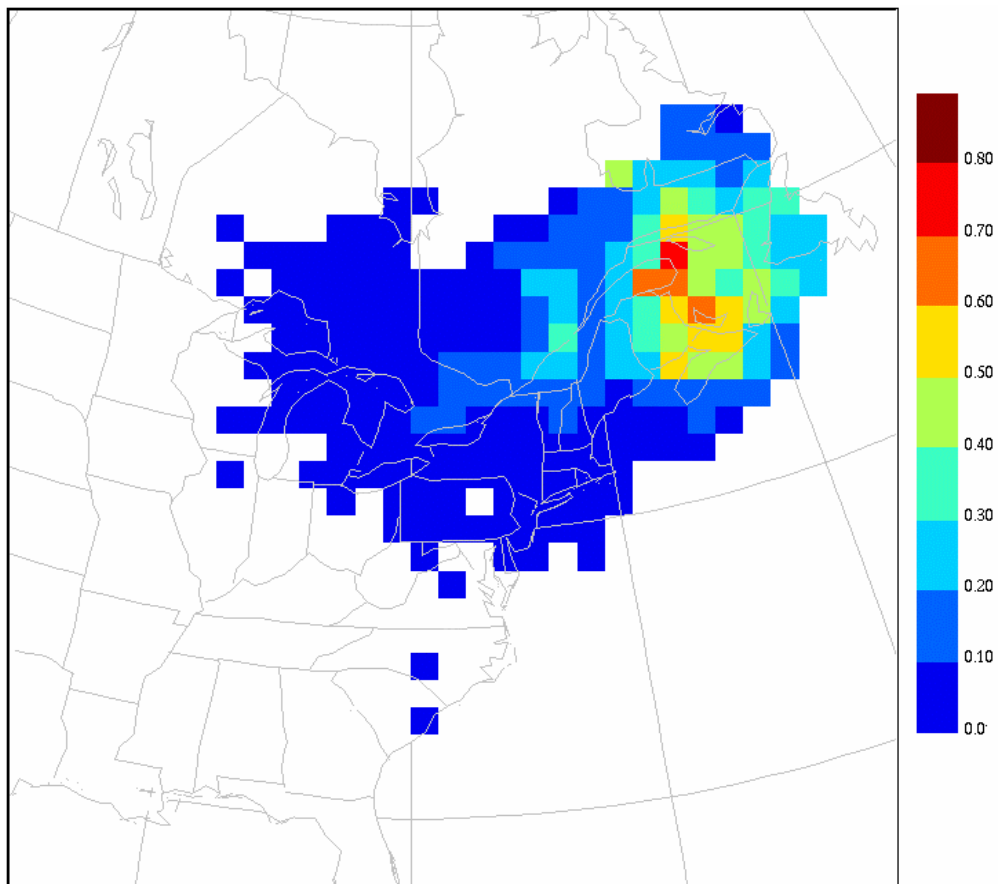


Figure 6. Plot of difference between wet SO₄ annual deposition fields (units of kg SO₄ ha⁻¹ yr⁻¹) for CCUSA2 and HLFO SO₂ emission scenarios.

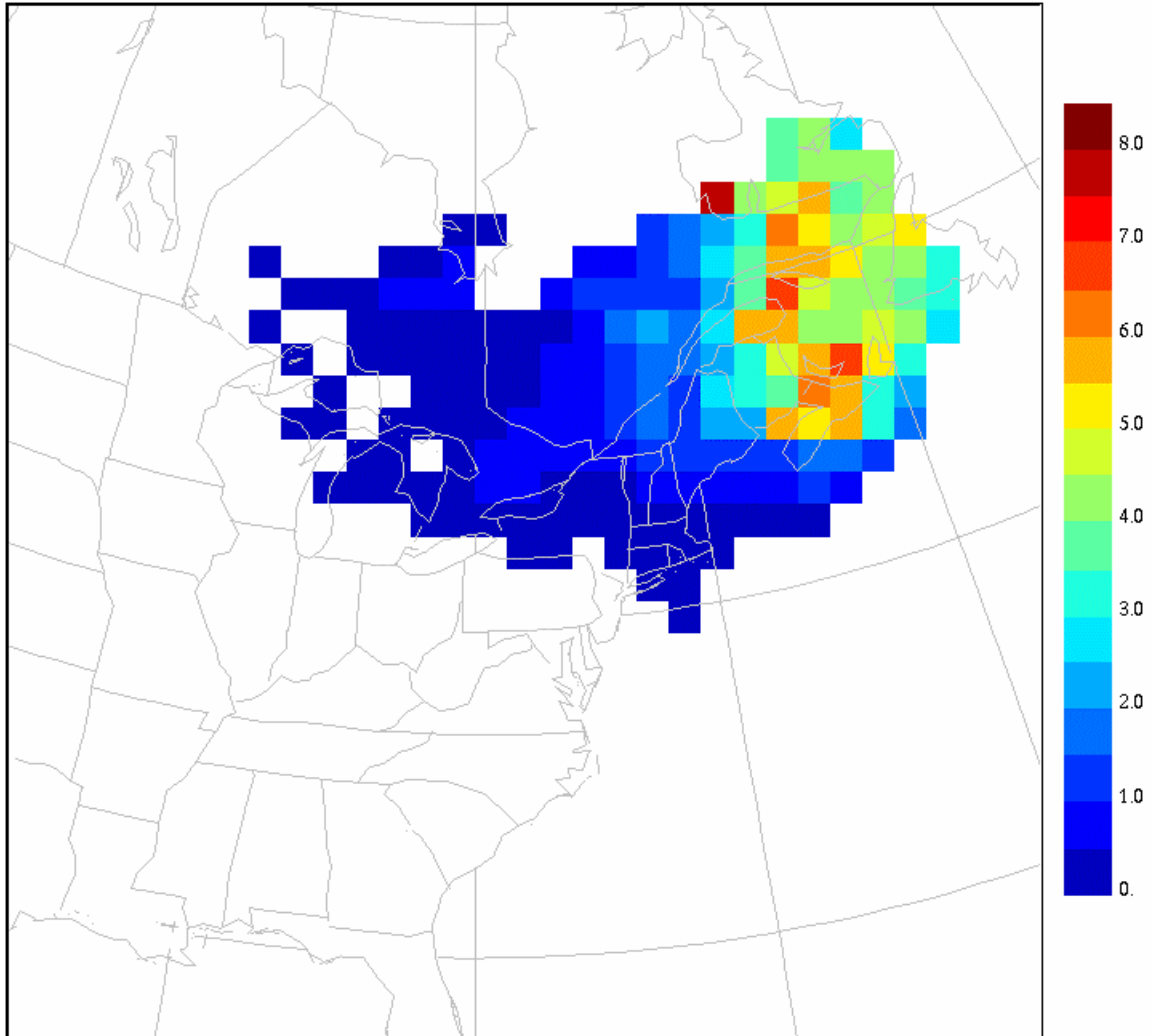


Figure 7. Same as Figure 6, but for percentage difference (%).

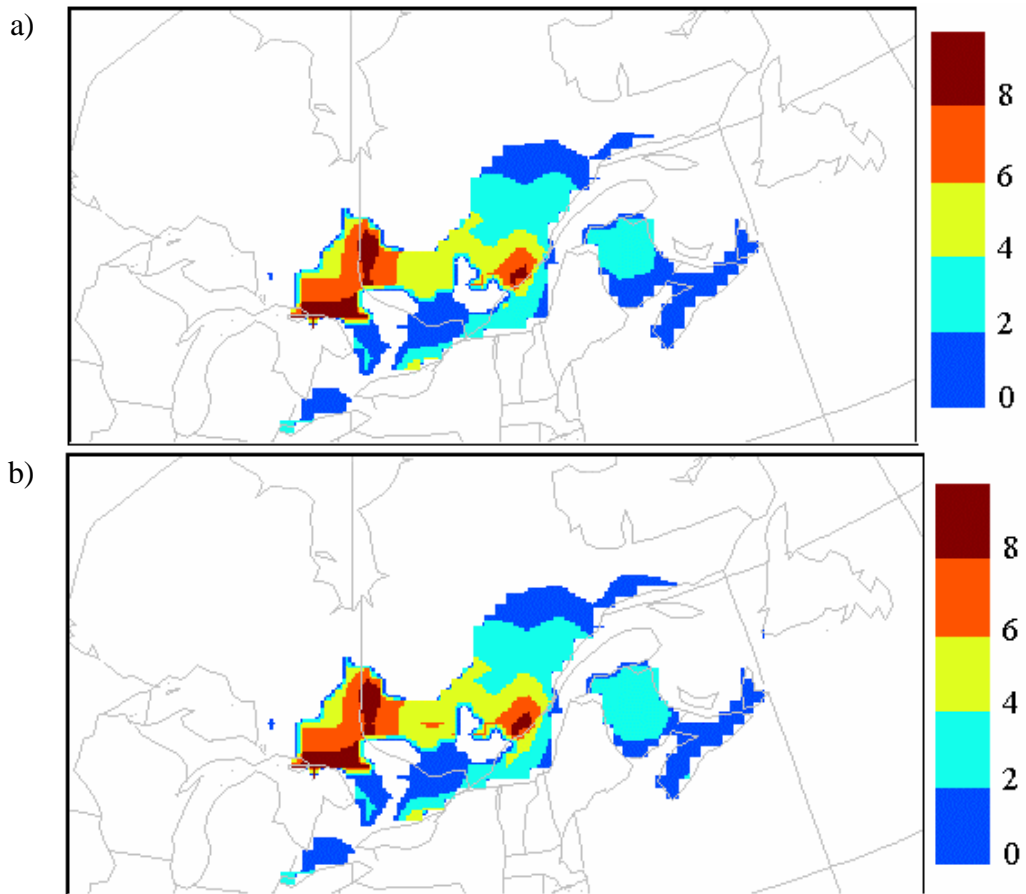


Figure 8. Plots of wet SO₄ deposition critical-load exceedance fields (units of kg SO₄ ha⁻¹ yr⁻¹) (a) HLFO and (b) CCUSA2 ADOM SO₂ emissions scenarios.

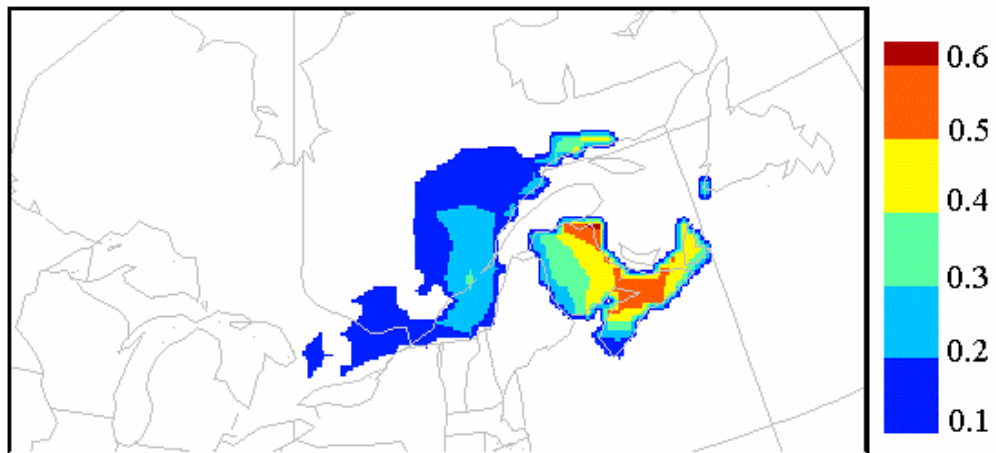


Figure 9. Plot of difference of wet SO₄ deposition critical-load exceedance fields (units of kg SO₄ ha⁻¹ yr⁻¹) for CCUSA2 and HLFO SO₂ emission scenarios.

A plot of ambient near-surface SO_4 air concentration patterns is shown in Figure 10. The first panel shows the predicted patterns corresponding to the current HLFO scenario, whereas the second panel shows ambient SO_4 concentration pattern for the ADOM 2010 scenario (“CCUSA2”: see Table 1). Again, the CCUSA2 scenario serves here as a nominal “base” scenario, since the new scenario considers further emission reductions relative to 2010 emission levels.

Figure 11 shows the difference between ambient near-surface SO_4 air concentration patterns for the ADOM 2010 (or “CCUSA2”) scenario and the current HLFO ADOM scenario.

Figure 12 shows the percentage difference between the ambient SO_4 patterns for the ADOM 2010 (“CCUSA2”) scenario and the current HLFO ADOM scenario.

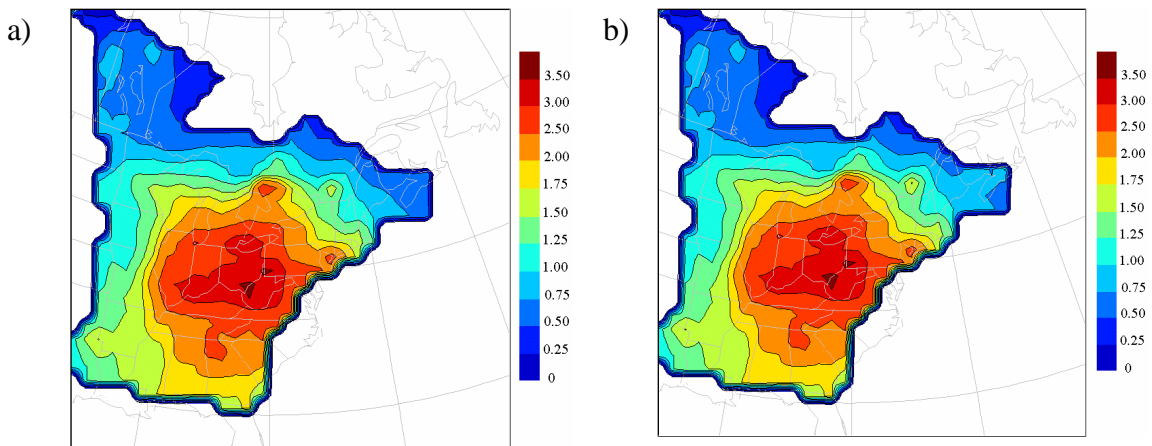


Figure 10. Plots of ambient near-surface SO_4 annual air-concentration patterns in units of $\mu\text{g SO}_4 \text{ m}^{-3}$ for (a) HLFO and (b) CCUSA2 ADOM SO_2 emissions scenarios.

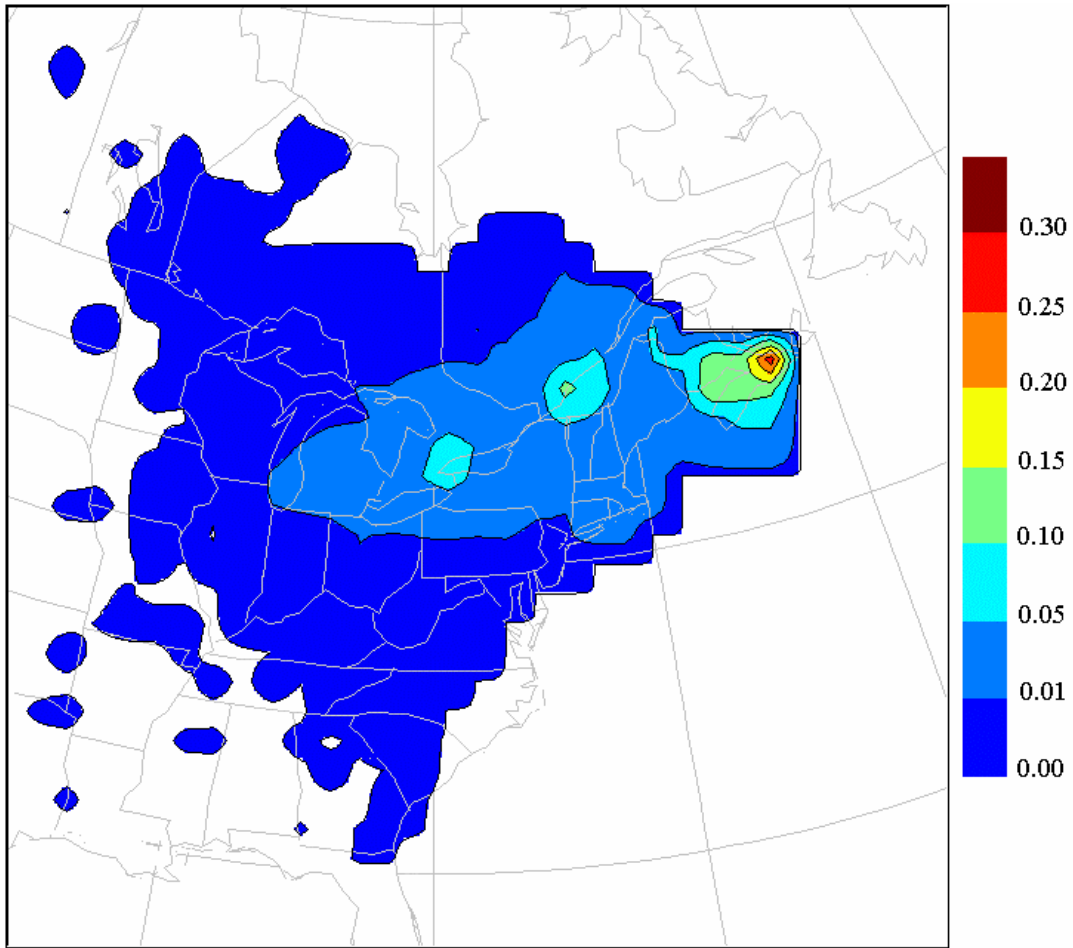


Figure 11. Plot of difference between ambient SO₄ annual fields (units of µg m⁻³) for the CCUSA2 and HLFO SO₂ emission scenarios.

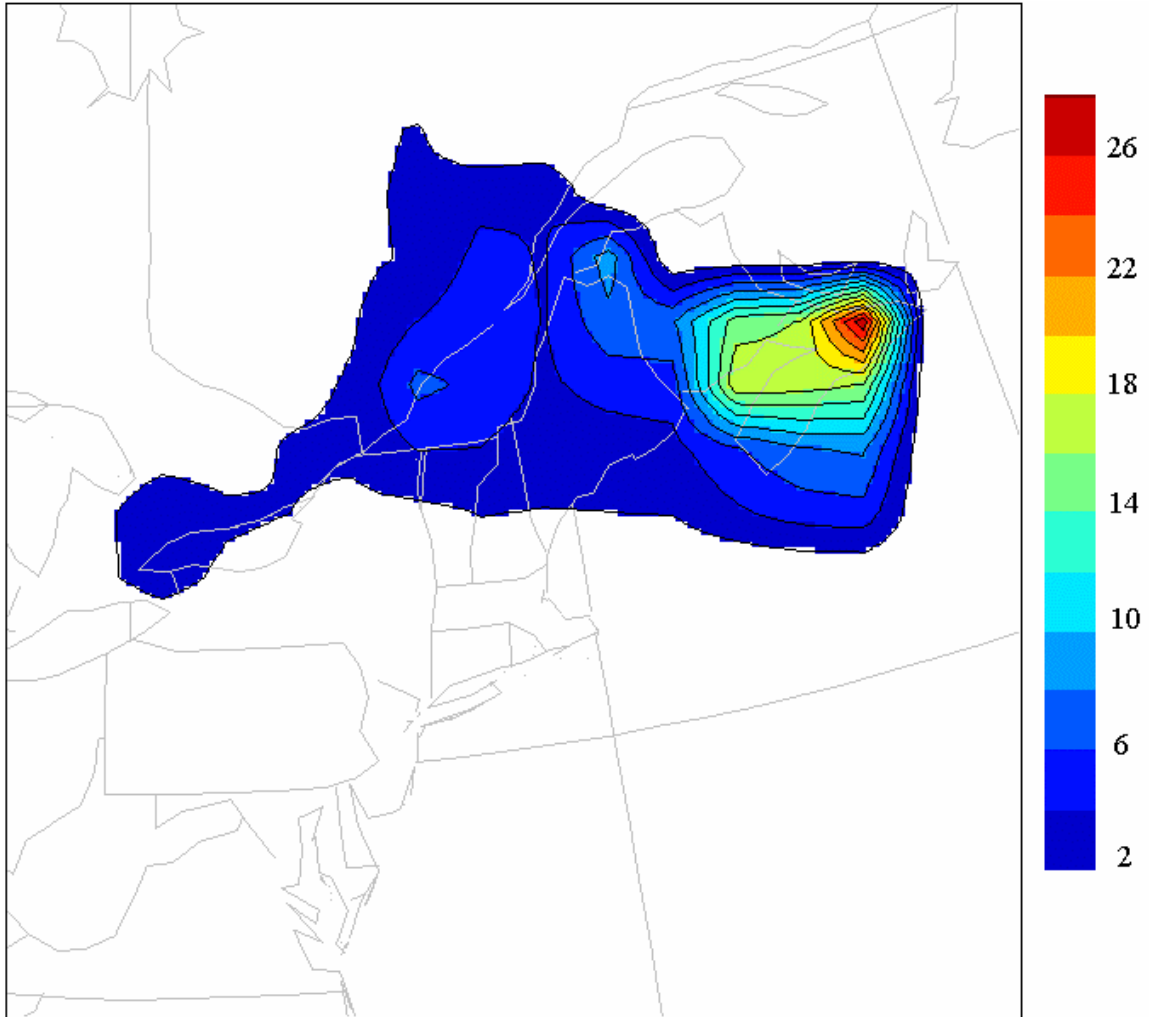


Figure 12. Plot of percentage difference (%) between ambient SO₄ annual fields for the CCUSA2 and HLFO SO₂ emission scenarios. The map was enlarged over the impacted area.

7. Discussion and Analysis

The implemented emission reductions constitute about 9% of the total SO₂ emitted over Canada (i.e., 180 vs 1,939 Ktonnes yr⁻¹). The emission reductions are relatively small on domain-scale (i.e., 180 vs. 14,385 Ktonnes yr⁻¹), on the other hand, and constitute about 1% of the total SO₂ emitted over the domain. However, the impacts of the emission reductions are localized and large because the reductions are concentrated in Maritimes. The decrease of 4-8% in wet SO₄ deposition (Figure 7), corresponding to 0.25-0.75 kg ha⁻¹ (Figure 6), is sufficient to impact on critical-load gaps in the Maritimes. The size of the area of eastern Canada in exceedance of critical load is summarized in Table 2 for the reference (CCUSA2) and the current (HLFO) ADOM SO₂ emission scenarios. As discussed in Environment Canada (1997), this quantity is one possible metric for comparing different scenarios quantitatively. The contribution that emission reductions make to wet SO₄ deposition in Ontario, Quebec and in the Maritimes is suggested by the differences between Figures 8a and 8b. There is a small decrease (~1.4%) of the land area in exceedance in Quebec and in Newfoundland and Labrador.

Table 2. Eastern Canadian land area in exceedance of wet SO₄ critical load for ADOM CCUSA2 and HLFO SO₂ emission scenarios.

Scenario Name	Nominal Year	Area (x 1,000 km ²)					All Eastern Canada
		Ontario	Quebec	New Brunswick	Nova Scotia	Nfld & Labrador	
CCUSA2	2010	204	406	95	82	4	791
HLFO	2010	204	399	95	82	0	780

The size of the area of eastern Canada in exceedance for different threshold levels (same as in Figures 8 and 9) of critical load is summarized in Table 3 for the reference (CCUSA2) and the current (HLFO) ADOM SO₂ emission scenarios. Also, the total eastern Canadian land area where exceedance of wet SO₄ critical load was reduced from above 1 kg SO₄ ha⁻¹ yr⁻¹ to below 1 kg SO₄ ha⁻¹ yr⁻¹ for the ADOM HLFO SO₂ emission scenario as compared to the ADOM CCUSA2 scenario was 37,634 km².

Table 3. Eastern Canadian land area (in 1,000 km²) in exceedance of wet SO₄ critical load for ADOM CCUSA2 and HLFO SO₂ emission scenarios for different thresholds.

Scenario Name	Nominal Year	Exceedance threshold (kg SO ₄ ha ⁻¹ yr ⁻¹)				
		0-2	2-4	4-6	6-8	≥8

CCUSA2	2010	290	246	125	81	50
HLFO	2010	308	229	122	73	48

The simulated emission reductions resulted in an 8-28% reduction in ambient SO₄ concentration in the Maritimes and a 2-6% reduction from Toronto to Montreal as shown in Figure 12. The decrease in ambient SO₄ concentrations would also result in a decrease in fine particulate matter (PM_{2.5}), of which sulphate particles are a significant fraction in eastern Canada (up to 40%: see Environment Canada, 2001), and thus result in improved health conditions (e.g., Environment Canada, 2001).

8. Conclusions

The impact of the proposed reduction to sulphur content in HFO and LFO resulted in SO₂ emissions reduction of 180 Ktonnes yr⁻¹ in eastern Canada, but the reductions were concentrated in the Maritimes.

The resulting changes in wet SO₄ deposition and ambient SO₄ concentration were largest in the Maritimes. The reduction in ambient sulphate concentration was on the order of 2% in southern Ontario, 6% around Montreal, and ~28% in the Maritimes, where up to 40% fine particulate matter is made up of sulphate. These changes are dominated by the local emission reductions and a small decrease in long-range transport from Ontario and Quebec.

9. Acknowledgments

The project team would like to acknowledge the Scientific Authorities for guidance and for useful comments and much suggested text for earlier versions of this report. We would also like to thank the members of the Oil, Gas and Energy Branch of Environment Canada for providing emission-scenario descriptions and for their helpful comments on the review version of this report. We acknowledge too, the access to MSC computer facilities, especially the NEC SX-4 supercomputers, provided by the Meteorological Service of Canada. The CCUSA2 ADOM scenario referred to in this report was produced by the Meteorological Service of Canada.

10. References

AETG, *Towards A National Acid Rain Strategy*. Report submitted to the National Air Issues Coordinating Committee by the Acidifying Emissions Task Group, Environment Canada, 98 pp., Oct. 1997.

ARM Consultants, Emissions-scenario simulations using the Acid Deposition and Oxidant Model, Report prepared for the Canadian Council of Ministers of the Environment Inc., 53 pp., October 2000.

Brook, J.R., P.J. Samson, and S. Sillman, Aggregation of selected three-day periods to estimate annual and seasonal wet deposition totals for sulfate, nitrate, and acidity. Part I. A synoptic and chemical climatology for eastern North America. *J. Appl. Meteor.*, 34, 297-325, 1995a.

Brook, J.R., P.J. Samson, and S. Sillman, Aggregation of selected three-day periods to estimate annual and seasonal wet deposition totals for sulfate, nitrate, and acidity. Part II. Selection of events, deposition totals, and source-receptor relationships. *J. Appl. Meteor.*, 34, 326-339, 1995b.

Deslauriers, M. Canadian emission inventory of criteria air contaminants (1990). Environmental Protection Service Report EPS 5/AP/7E, Environmental Protection Service, Environment Canada. Ottawa, 75 pp., 1996.

Environment Canada, *1994 Annual Report on the Federal-Provincial Agreements for the Eastern Canada Acid Rain Program*, Cat. EN40-11/29-1994E, ISBN: 0-662-23665-3, Environment Canada, 14 pp., 1994.

[See also website http://www.ec.gc.ca/pdb/pdb_er.html]

Environment Canada, *1997 Canadian Acid Rain Assessment, Volume 2, Atmospheric Science Assessment Report*, Supply and Services Canada, 302 pp., 1997.

Environment Canada, *1997 Annual Report on the Federal-Provincial Agreements for the Eastern Canada Acid Rain Program*, Cat. EN40-11/29-1997, ISBN: 0-662-63700-3, ISSN: 0846-3964, Environment Canada, 5 pp., 1998.

[See also website http://www.ec.gc.ca/pdb/pdb_er.html]

Environment Canada, Precursor contributions to ambient fine particulate matter in Canada, Cat. EN56-167/2001E, ISBN:0-662-30650-3, 237 pp., 2001.

[See also website <http://www.msc-smc.ec.gc.ca/saib>]

Federov, V.V., Kriging and other estimators of spatial field characteristics (with special reference to environmental studies). *Atmos. Environ.*, 23, 175-184, 1989.

Finkelstein, P.L., The spatial analysis of acid precipitation data. *J. Clim. Appl. Meteor.*, 23, 52-62, 1984.

Fung, C. S., P.K. Misra, R. Bloxam, and S. Wong, A numerical experiment on the relative importance of H₂O₂ and O₃ in aqueous conversion of SO₂ to SO₄²⁻. *Atmos. Environ.*, 25A, 411-423, 1991.

Jeffries, D.S., D.C.L. Lam, I. Wong and M.D. Moran, Assessment of changes in lake pH in southeastern Canada arising from present levels and expected reductions in acidic deposition. *Can. J. Fish. Aquat. Sci.*, 2000 (In press).

Li, S-M., K.G. Anlauf, H.A. Wiebe, J.W. Bottenheim, and K.J. Puckett, Evaluation of a comprehensive Eulerian air quality model with multiple chemical species measurements using principal component analysis. *Atmos. Environ.*, 28, 3449-3461, 1994.

Lurmann, F. W., A. C. Lloyd and R. Atkinson, A chemical mechanism for use in long-range transport/acid deposition computer modeling, *J. Geophys. Res.*, 91, 10905-10936, 1986.

Macdonald, A.M., C.M. Banic, W.R. Leitch, and K.J. Puckett, Evaluation of the Eulerian Acid Deposition and Oxidant Model (ADOM) with summer 1988 aircraft data. *Atmos. Environ.*, 27A, 1019-1034, 1993.

Misra, P.K., R. Bloxam, C. Fung, and S. Wong, Non-linear response of wet deposition to emissions reduction: a model study, *Atmos. Environ.*, 23, 671-687, 1989.

Moran, M.D., Operational evaluation of ADOM seasonal performance with surface data from the Eulerian Model Evaluation Field Study. *Proc. 10th AMS/AWMA Joint Conf. on Applications of Air Pollution Meteorology*, Jan. 11-16, Phoenix, Arizona, American Meteorological Society, Boston, pp. 404-408, 1998.

Padro, J., G. Den Hartog and H. H. Neumann, An investigation of the ADOM dry deposition module using summertime O₃ measurements above a deciduous forest, *Atmos. Environ.*, 25, 1689-1704, 1991.

RMCC (Federal/Provincial Research and Monitoring Coordinating Committee), *The 1990 Canadian Long-Range Transport of Air Pollutants and Acid Deposition Assessment Report: Part 4 – Aquatic Effects*, 151 pp., Federal/Provincial Research and Monitoring Coordinating Committee, Environment Canada, Ottawa, 1990.

Schaug, J., T. Iversen, and U. Pedersen, Comparison of measurements and model results for airborne sulphur and nitrogen compounds with kriging. *Atmos. Environ.*, 27A, 831-844, 1993.

Tushingham, M., and J. Bellamy, Potential to reduce emissions of sulphur dioxide through reducing sulphur levels in heavy and light fuel oils. Discussion paper, 30 pp., Oil, Gas and Energy Branch, Environment Canada, 2001.

U.S. EPA, *The 1985 NAPAP emission inventory (version 2). Development of the annual data and modelers' tapes*, Rep. EPA-600/7-89-012a, United States Environmental Protection Agency, 692 pp., Tech. Info. Serv., Springfield, Va., 1989.

U.S. EPA (United States Environmental Protection Agency), *Aerometric Information Retrieval System (AIRS) User's Guide*, Office of Air Quality Planning and Standards, Research Triangle Park, N.C., Dec. 1989b.

Venkatram, A., P. K. Karamchandani and P. K. Misra, Testing a comprehensive acid deposition model, *Atmos. Environ.*, 22, 737-747, 1988.

Appendix A – Scenario Scaling Factors

Table A1. Scenario scaling factors used in ADOM scenario implementation for Scenario HLFO.

Province	ID	Scenario Scale Factor
Saskatchewan	1	1.0000
Manitoba	2	1.0000
Ontario	3	0.7832
Quebec (*)	4	0.8041
New Brunswick (*)	5	0.0000
Nova Scotia/PEI	6	0.4896
Newfoundland	7	0.3875
US	-1	1.0000

(*) The jurisdiction of ADOM grid point (24,23) was re-assigned from Quebec to New Brunswick so that the New Brunswick Power Commission Generating Station at Dalhousie, located near the southern edge of this grid cell, would be scaled properly.

Note that the scenario scaling factors are complements of the reduction factors: for example, for a 21.68% reduction, Ontario SO₂ emissions are scaled by 78.32%.

Appendix B – Point-Source-Specific Emissions Values

The SO₂ emissions used in the reference scenario (“CCUSA2”) for the three New Brunswick generating stations that use HFO, Courtenay Bay, Coleson Cove, and Dalhousie, were 3.6, 21.3, and 43.4 kT y⁻¹, respectively. As shown in Table B1, emissions from these three sources were reduced by 3.6, 21.3, and 28.0 kT y⁻¹, respectively, for the new HFO/LFO ADOM scenario. SO₂ emissions for Tufts Cove Generating Station in Nova Scotia had already been set to zero in the reference scenario, so these were not changed for the new scenario. Note that two smokestacks are considered for Dalhousie Generating Station.

Table B1. Changes made to individual Canadian major point sources.

ADOM Record ID	Source Name	Emissions Scaling
3425	Courtenay Bay GS, N.B.	Emissions set to zero
3428	Coleson Cove GS, N.B.	Emissions set to zero
3426	Dalhousie GS, N.B.	$E_{\text{new}}=E_{\text{old}} * 0.6452$

3427	Dalhousie GS, N.B.	$E_{\text{new}}=E_{\text{old}} * 0.6452$
------	--------------------	--

Appendix C – SO₂ Emission Levels From HFO and LFO

Table C1 was compiled from Tables 2.5 and 3.6 of the discussion paper by Tushingham and Bellamy (2001). It includes the net contributions to each region of international exports but not the contribution of inter-regional transfers within Canada.

Table C1. SO₂ emission levels based on production of HFO and LFO per region.

REGION	Current SO ₂ Emissions (tonnes yr ⁻¹)			1% HFO/1000ppm LFO Scenario (tonnes yr ⁻¹)			Difference (tonnes yr ⁻¹)		
	HFO	LFO	HFO+LFO	HFO	LFO	HFO+LFO	HFO	LFO	HFO+LFO
Atlantic	146,346	3,140	149,486	53,227	1,784	55,011	93,119	1,356	94,475
Quebec	54,174	6,944	61,118	34,696	2,067	36,763	19,478	4,877	24,355
Ontario	60,180	4,864	65,044	25,082	1,636	26,718	35,098	3,228	38,326
Prairies	13,926	0	13,926	8,415	0	8,415	5,511	0	5,511
B.C.	2,686	124	2,810	1,245	124	1,369	1,441	0	1,441
CANADA	277,312	15,072	292,384	122,665	5,611	128,276	154,647	9,461	164,108

Table C2 was constructed from Tables 2.1 and 3.1 of Tushingham and Bellamy (2001) using the sum of sales and refinery consumption in the course of the production process as a consumption surrogate. These figures account for production and net imports within each region, including both inter-regional and international transport, and hence better reflect regional SO₂ emissions from HFO and LFO combustion within each region.

Table C2. SO₂ emission levels based on consumption of HFO and LFO per region.

REGION	Current SO ₂ Emissions (tonnes yr ⁻¹)			1% HFO/1000ppm LFO Scenario (tonnes yr ⁻¹)			Difference (tonnes yr ⁻¹)		
	HFO	LFO	HFO+LFO	HFO	LFO	HFO+LFO	HFO	LFO	HFO+LFO
Atlantic	170,309	4,097	174,406	61,931	1,164	63,094	108,379	2,933	111,311
Quebec	59,435	8,689	68,124	38,069	1,293	39,362	21,366	7,396	28,762
Ontario	49,455	5,696	55,152	20,617	958	21,575	28,838	4,739	33,577
Prairies	1,446	123	1,569	874	26	900	572	97	669
B.C.	28,100	2	28,102	13,024	0	13,025	15,076	1	15,077

CANADA	308,746	18,606	327,352	134,515	3,441	137,956	174,231	15,165	189,396
--------	---------	--------	---------	---------	-------	---------	---------	--------	---------

Appendix D – Literature Review and Guide to ADOM Legacy

ADOM – Development

The Acid Deposition and Oxidant Model (ADOM) was developed by a US consulting company ERT (Environmental Research and Technology), under a contract to several governmental agencies: the Ontario Ministry of the Environment (OME), the UmweltBundesamt of West Germany, and Environment Canada (EC).

The first version of the model was released in February 1986. A User's Guide was provided (**Scire et al., 1986**).

Scire, J.S., F.W. Lurmann, P. Karamchandani, A. Venkatram, R. Yamartino, J. Young, and J. Pleim, ADOM/TADAP user's guide, ADOM/TADAP Model development program, Volume 9, ERT, 1986.

The chemical mechanism in the original model was based on work published by **Lurmann et al. (1986)**.

Lurmann, F. W., A. C. Lloyd and R. Atkinson, A chemical mechanism for use in long-range transport/acid deposition computer modeling, *J. Geophys. Res.*, 91, 10905-10936, 1986.

Initial model testing and evaluation was carried out by researchers from the original model development team and summarized by **Venkatram et al. (1988)**.

Venkatram, A., P. K. Karamchandani and P. K. Misra, Testing a comprehensive acid deposition model, *Atmos. Environ.*, 22, 737-747, 1988.

Subsequent model improvements were carried out by several groups of researchers at Environment Canada and the Ontario Ministry of the Environment.

Research on parameterization of dry deposition was carried out by **Padro et al. (1991, 1993)** at EC and resulted in several modifications to the dry deposition module in ADOM.

Padro, J., G. Den Hartog and H. H. Neumann, An investigation of the ADOM dry deposition module using summertime O₃ measurements above a deciduous forest, *Atmos. Environ.*, 25, 1689-1704, 1991.

Padro, J., K.J. Puckett, and D.N. Woolridge, The sensitivity of regionally averaged O₃ and SO₂ concentrations to ADOM dry deposition velocity parameterizations. *Atmos. Environ.*, 27A, 2239—2242, 1993.

Research on sulphate chemistry and wet deposition was carried out at OME (**Misra et al., 1989; Fung et al., 1991, 1992**).

Misra, P.K., R. Bloxam, C. Fung, and S. Wong, Non-linear response of wet deposition to emissions reduction: a model study. *Atmos. Environ.*, 23, 671-687, 1989.

Fung, C. S., P.K. Misra, R. Bloxam, and S. Wong, A numerical experiment on the relative importance of H₂O₂ and O₃ in aqueous conversion of SO₂ to SO²⁻₄. *Atmos. Environ.*, 25A, 411-423, 1991.

Fung, C., R. Bloxam, P.K. Misra, S. Wong, and D. Yap, Evaluating the comprehensive model ADOM with data from three seasons. OME Report PIBS-2064, Ontario Ministry of the Environment, Toronto, Ontario, 64 pp., 1992.

Further research on sulphate production and in-cloud chemistry was carried out by **Karamchandani and Venkatram (1992)**.

Karamchandani, P.K. and A. Venkatram, The role of non-precipitating clouds in producing ambient sulfate during summer: results from simulations with the Acid Deposition and Oxidant Model (ADOM). *Atmos. Environ.*, 26A, 1041-1052, 1992.

An extensive model overview is included in an **Environment Canada (1997)** report.

Environment Canada, 1997 *Canadian Acid Rain Assessment, Volume 2, Atmospheric Science Assessment Report*, Supply and Services Canada, 302 pp., 1997.

ADOM – Performance Evaluation

Model performance evaluation and intercomparison with another comprehensive acid deposition model, RADM (Regional Acid Deposition Model: **Chang et al., 1987**) was carried out by several groups at OME and EC.

Chang J. S., R. A. Brost, I. S. A. Isaksen, S. Madronich, P. Middleton, W. R. Stockwell and C. J. Walcek, A three-dimensional Eulerian acid deposition model: Physical concepts and formulation, *J. Geophys. Res.*, 92, 14681-14700, 1987.

ERP, Eulerian Model Evaluation Field Study (EMEFS): Report of the Fourth Meeting of the External Review Panel. Draft report, May 25-27, Niagara-on-the-Lake, Ontario, Canada, July, 18 pp. [Available from ARQP, Atmospheric Environment Service, Downsview, Ontario], 1994.

The Eulerian Model Evaluation Field Study (EMEFS) also provided an opportunity for evaluation of different components of ADOM using surface data by **Sirois et al. (1995)** and **Moran (1998)**.

Sirois, A., M.P. Olson, and B. Pabla. The use of spectral analysis to examine model and observed O₃ data. *Atmos. Environ.*, 29, 411-422, 1995.

Moran, M.D., Operational evaluation of ADOM seasonal performance with surface data from the Eulerian Model Evaluation Field Study. *Proc. 10th AMS/AWMA Joint Conf. on Applications of Air Pollution Meteorology*, Jan. 11-16, Phoenix, Arizona, American Meteorological Society, Boston, pp. 404-408, 1998.

Model evaluation with vertical measurements was carried out by Macdonald et al. (1993) and by Hoff et al. (1995).

Macdonald, A.M., C.M. Banic, W.R. Leitch, and K.J. Puckett, Evaluation of the Eulerian Acid Deposition and Oxidant Model (ADOM) with summer 1988 aircraft data. *Atmos. Environ.*, 27A, 1019-1034, 1993.

Hoff, R.M., R.E. Mickle, and C. Fung, Vertical profiles of ozone during the EMEFS-I experiment in southern Ontario. *Atmos. Environ.*, 29, 1735-1747, 1995.

Model evaluation for several species using principal component analysis was carried out by **Li et al. (1994)**.

Li, S.-M., K.G. Anlauf, H.A. Wiebe, J.W. Bottenheim, and K.J. Puckett, Evaluation of a comprehensive Eulerian air quality model with multiple chemical species measurements using principal component analysis. *Atmos. Environ.*, 28, 3449-3461, 1994.

ADOM - Episode Aggregation

Work on episode aggregation was carried out by **Brook et al. (1995a,b)**. Episode aggregation is a semi-empirical approach that utilizes a set of short-term ADOM simulations to estimate concentration and deposition fields for annual and longer time periods. Episode aggregation approach is used in generating ADOM emissions reduction scenarios. The application of episode aggregation in ADOM is described in Environment Canada (1997).

Brook, J.R., P.J. Samson, and S. Sillman, Aggregation of selected three-day periods to estimate annual and seasonal wet deposition totals for sulfate, nitrate, and acidity. Part I. A synoptic and chemical climatology for eastern North America. *J. Appl. Meteor.*, 34, 297-325, 1995a.

Brook, J.R., P.J. Samson, and S. Sillman, Aggregation of selected three-day periods to estimate annual and seasonal wet deposition totals for sulfate, nitrate, and acidity. Part II. Selection of events, deposition totals, and source-receptor relationships. *J. Appl. Meteor.*, 34, 326-339, 1995b.

Environment Canada, *1997 Canadian Acid Rain Assessment, Volume 2, Atmospheric Science Assessment Report*, Supply and Services Canada, 302 pp., 1997.

ADOM – Applications and Assessments

Application of ADOM to different emission reduction scenarios was carried out over the years. A list of seventeen ADOM scenarios is given in Table 1. Results of ADOM scenario simulations are described in several of the reports. Also, results from ADOM simulations were used in several of the assessment reports listed below.

AETG, *Towards A National Acid Rain Strategy*. Report submitted to the National Air Issues Coordinating Committee by the Acidifying Emissions Task Group, Environment Canada, 98 pp., Oct. 1997.

ARM Consultants, Emissions-scenario simulations using the Acid Deposition and Oxidant Model, Report prepared for the Canadian Council of Ministers of the Environment Inc., 53 pp., October 2000.

Environment Canada, *1994 Annual Report on the Federal-Provincial Agreements for the Eastern Canada Acid Rain Program*, Cat. EN40-11/29-1994E, ISBN: 0-662-23665-3, Environment Canada, 14 pp., 1994.

[See also website http://www.ec.gc.ca/pdb/pdb_er.html]

Environment Canada, *1997 Canadian Acid Rain Assessment, Volume 2, Atmospheric Science Assessment Report*, Supply and Services Canada, 302 pp., 1997.

Environment Canada, *1997 Annual Report on the Federal-Provincial Agreements for the Eastern Canada Acid Rain Program*, Cat. EN40-11/29-1997, ISBN: 0-662-63700-3, ISSN: 0846-3964, Environment Canada, 5 pp., 1998.

[See also website http://www.ec.gc.ca/pdb/pdb_er.html]

Environment Canada, Precursor contributions to ambient fine particulate matter in Canada, Cat. EN56-167/2001E, ISBN:0-662-30650-3, 237pp., 2001.

[See also website <http://www.msc-smc.ec.gc.ca/saib>]

RMCC (Federal/Provincial Research and Monitoring Coordinating Committee), *The 1990 Canadian Long-Range Transport of Air Pollutants and Acid Deposition Assessment Report: Part 4 – Aquatic Effects*, 151 pp., Federal/Provincial Research and Monitoring Coordinating Committee, Environment Canada, Ottawa, 1990.

ADOM - Emissions

Generation of model emissions files from US and Canadian inventories is described in **ORTECH (2000)**. In addition, Canadian emission inventory is described in **Deslauriers (1996)**. However, the model emissions files for 1985 and 1988 generated for ADOM were documented in some internal MEP and ORTECH reports and the corresponding inventories were described in two **EPA reports (1989a,b)**.

Deslauriers, M. Canadian emission inventory of criteria air contaminants (1990). Environmental Protection Service Report EPS 5/AP/7E, Environmental Protection Service, Environment Canada, Ottawa, 75 pp., 1996.

ORTECH, Preparation of 1990 North American emissions inventory modeling files for AES regional air quality models, Report 10193 (final) by Canadian ORTECH Environmental, 298 pp + 6 appendices, 2000.

U.S. EPA, *The 1985 NAPAP emission inventory (version 2). Development of the annual data and modelers' tapes*, Rep. EPA-600/7-89-012a, United States Environmental Protection Agency, 692 pp., Tech. Info. Serv., Springfield, Va., 1989a.

U.S. EPA (United States Environmental Protection Agency), *Aerometric Information Retrieval System (AIRS) User's Guide*, Office of Air Quality Planning and Standards, Research Triangle Park, N.C., Dec. 1989b.