2.2 PREDICTED ACID DEPOSITION CRITICAL-LOAD EXCEEDANCES ACROSS CANADA FROM A ONE-YEAR SIMULATION WITH A REGIONAL PARTICULATE-MATTER MODEL

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1. INTRODUCTION

Acid deposition has been an environmental concern in eastern North America since the 1970s. In response there have been major initiatives in both Canada and the U.S. to reduce acid deposition by means of SO_2 emission reductions (*1985 Eastern Canada Acid Rain Program*, Title IV of *1990 U.S. Clean Air Act Amendments*, *1991 Canada–United States Air Quality Agreement*, *1998 Canada-Wide Acid Rain Strategy for Post-2000, 2005 U.S. Clean Air Interstate Rule*).

Although these SO_2 emission control programs have been successful in reducing levels of sulphur deposition in eastern North America (e.g., Lynch et al., 1995, 2000; Sirois, 1997; Baumgardner et al., 1999, 2002; Holland et al., 1999; Butler et al., 2001; Malm et al., 2004; Vet et al., 2005), recent studies suggest that acid deposition is still occurring at damaging levels in parts of eastern North America (Jeffries and Ouimet, 2005; McNulty et al., 2007). Examples of harmful effects linked to high levels of acid deposition include elevated acidity in lakes and streams, loss of sensitive species, reduced biological diversity, reduced forest productivity, and forest mortality (e.g., Driscoll et al., 2001; Houle, 2005; Jeffries et al., 2005; Jeffries and Ouimet, 2005). It is thus important to be able to assess whether the acid deposition at any location is harmful or not.

One tool that has been developed for this purpose is the acid-deposition critical load (ADCL). An ADCL is a quantitative measure of the acid buffering capacity of an ecosystem. It provides an objective metric that can be used to determine both the spatial extent of a region being subjected to damaging levels of acid deposition and the magnitude of the acidification. An ADCL field will typically vary geographically, since some locations have a lower acid buffering capacity than others and hence are more sensitive to acid deposition. If the difference between the annual atmospheric total (=wet+dry) acid deposition to an ecosystem and the ecosystem's ADCL value is positive, then that difference is termed an ADCL *exceedance* since the acid deposition to that ecosystem is larger than the ecosystem's acid buffering capacity so that acidification is occurring.

An ADCL field for freshwater aquatic ecosystems has been available for eastern Canada since the 1990s (e.g., Jeffries and Lam, 1993), but more recently a new national ADCL field has been developed that (a) covers most of sub-Arctic Canada from coast to coast and (b) considers acidification of forest ecosystems as well as aquatic ecosystems (Jeffries and Ouimet, 2005; Aherne and Watmough, 2006). The availability of this new multi-ecosystem ADCL field makes it possible to evaluate the impact of acid deposition across Canada for past, present, and future conditions using some combination of acid-deposition measurements and modelling. (Work is now underway to develop comparable ADCL fields for the U.S. after some initial resistance [e.g., U.S. EPA, 1995; McNulty et al., 2007]).

The use of an acid-deposition model to estimate ADCL exceedances, however, offers two significant advantages compared to the use of acid-deposition measurements. First, the model can provide predictions of past or current acid deposition in regions where acid deposition was not or is not measured. And second, the model can provide predictions of acid deposition for future-year acidifying-emission scenarios for which measurements are again not available. Model-predicted ADCL exceedance fields can also be compared for members of a set of different acidifying-emission control scenarios in order to quantify and rank the expected environmental impacts of different management strategies (e.g., Moran, 2005).

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In this paper, total acid-deposition fields for North America for 2002 predicted by Environment Canada's AURAMS regional particulate-matter model are compared with the 2006 version of the new Canada-wide multi-ecosystem ADCL field to identify regions in Canada that either are being damaged or are at risk of being damaged by the atmospheric deposition of sulphur (S) and nitrogen (N) species. Model-predicted ADCL exceedance fields for Canada are also compared with recent independent estimates of ADCL exceedances based on measurement data.

The next section describes the 2006 versions of the new aquatic, forest, and combined national ADCL fields for Canada. The AURAMS modelling system and its application for a complete one-year simulation (for 2002) are then described in Section 3. Next, some AURAMS annual predictions for 2002 are shown in Section 4 along with results from an evaluation of these predictions with 2002 measurement data; the ADCL exceedance fields obtained using AURAMS-predicted 2002 acid-deposition fields are also presented in this section. Section 5 provides a discussion of the results, including a qualitative comparison of the AURAMS-predicted ADCL exceedance fields with ADCL exceedance fields obtained from 1994-98 measurement data. The final section, Section 6, closes the paper with a summary and conclusions.

2. CRITICAL LOAD FIELDS FOR CANADA

The critical-load concept applies more broadly than to just acid deposition. An often-referenced definition of "critical load" is that of Nilsson and Grennfelt (1988): "a quantitative estimate of exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge".

For acid deposition, an acid-deposition critical load quantifies the ability of an ecosystem to sustain continuing annual inputs of acidity without damage. This will depend on the natural acid neutralizing capacity of that ecosystem, which is primarily controlled by soil and bedrock geology. Canada-wide ADCL fields have been developed for two types of ecosystems: freshwater aquatic ecosystems and forest ecosystems.

2.1 ADCL Field for Aquatic Ecosystems

An ADCL field for freshwater aquatic ecosystems based on lake-chemistry surveys of selected lakes has been available for eastern Canada since 1990 (e.g., Jeffries and Lam, 1993; Jeffries et al., 1999). The aquatic-ecosystem ADCL is based on calculated acid neutralizing capacity (ANC) values, where ANC is defined to be the difference between base-cation and strong-acid-anion concentrations in lake water. ANC has the advantages that (a) it reflects both hydrogen ion toxicity and biotic toxity associated with other elements such as ionic aluminum and (b) addresses the case of extremely dilute lakes better than pH does (Jeffries and Ouimet (2005).

More recently aquatic-ecosystem ADCL values have also been calculated based on lake-chemistry data from western Canada. Jeffries and Ouimet (2005) presented and discussed a new Canada-wide gridded field of aquatic-ecosystem ADCL based on lake-chemistry data from all 10 Canadian provinces, where each grid cell is on the order of 127 km by 127 km in size. The new ADCL field (not shown) is expressed in terms of annual total (=wet+dry) acid deposition in units of eq/ha/yr. The ADCL value for a grid cell applies to the population of lakes within that grid cell and has been chosen to be protective of 95% of that population (i.e., 5% of lakes in that grid cell will still experience acidification for acid deposition at that ADCL level).

An important limitation of the 2005 version of this aquatic-ecosystem ADCL field is that it does not include all areas of Canada with lakes, but gaps in coverage will gradually be filled in as new lake-chemistry data become available. For example, new lake-chemistry surveys were carried out by Environment Canada (EC) in 2006 and 2007 in northwestern Manitoba and northeastern Saskatchewan, respectively.

2.2 ADCL Field for Upland Forest Ecosystems

Starting in the 1980s European researchers developed a simple mass balance approach for calculating forest-soil ADCLs (e.g., Sverdrup and de Vries, 1994; Hall et al., 2001; UBA, 2004). A single base-cation (BC) mass-balance equation is used to quantify the acid-neutralizing capacity of upland forest soils, where the total soil BC pool is represented by the sum of four elements (calcium, potassium, magnesium, and sodium). The three key terms in this BC mass-balance equation are BC sources due to weathering and atmospheric deposition and BC removal due to leaching.

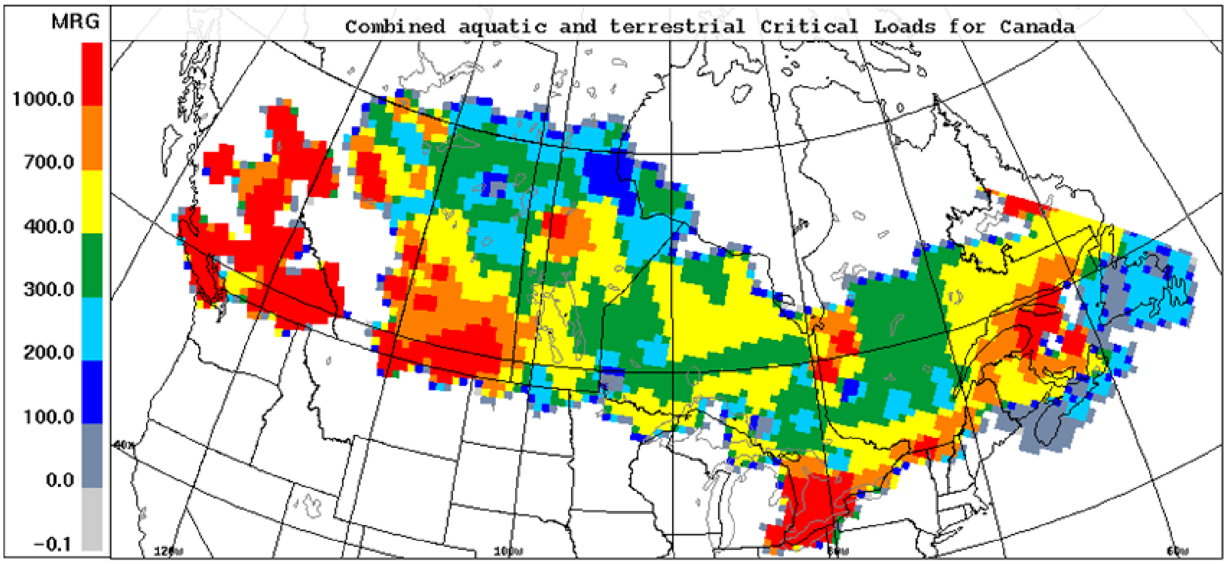


Figure 1. Combined aquatic and forest ADCL field for Canada (eq/ha/y). The white colour indicates areas for which no ADCL values are available.

More recently, North American researchers have applied this BC mass-balance approach to estimate forest-soil ADCLs for Canada and the U.S. (e.g., Arp et al., 1996, 2001; NEG-ECP, 2001, 2007; Jeffries and Ouimet, 2005; McNulty et al., 2007). Jeffries and Ouimet (2005) presented and discussed a forest-soil ADCL map for Canadian upland forest ecosystems from the Manitoba-Ontario border eastward. Aherne and Watmough (2006) then applied the same methodology to extend the forest-soil ADCL map westward to include the neighbouring provinces of Manitoba and Saskatchewan. The 2006 version of the map (not shown), which covers Canada from Saskatchewan eastward, has been used here. The new ADCL field is expressed in terms of annual total acid deposition in units of eq/ha/yr.

2.3 Combined ADCL Field

In order to maximize the area of Canada for which at least one ecosystem-specific ADCL value is available, Jeffries and Ouimet (2005) used a simple algorithm to combine the available aquatic-ecosystem and forest-ecosystem ADCL fields for Canada to obtain a national combined ADCL field. For each grid cell of the two ecosystem-specific ADCL fields, they assigned an ADCL value to the same grid cell on the new combined ADCL field by taking the *smaller* of the aquatic and forest ADCL values if both were available or the one value that was available if only one of the aquatic and forest ADCL fields had a value for that grid cell.

This calculation has been redone to account for (a) the availability of the 2006 version of the Canadian forest ADCL field that includes coverage of Manitoba and Saskatchewan and (b) the higher-resolution AURAMS grid (42 km vs. 127 km). Figure 1 shows the resulting 2006 version of the combined ADCL field for Canada on the AURAMS continental grid with 42-km horizontal grid spacing (shown in Figure 2). Sensitive areas (i.e., areas with small ADCL values) can be seen in western and northern Newfoundland, in southern Nova Scotia and southern New Brunswick, in Ontario east of Lake Superior, and in northern Manitoba and Saskatchewan.

Note that there are still some gaps in the combined ADCL field for the two westernmost Canadian provinces, Alberta and British Columbia, but forest-ecosystem ADCL fields are under development for these provinces. Note also that the low values for cells on the periphery of the ADCL field are generally artifacts associated with the gridding procedure that occur when ADCL values are not available for a portion of a grid cell (i.e., "edge" effects).

2.4 The Role of N Deposition

S deposition is assumed always to be acidic since "S has little biological reactivity and SO_4^{2-} acts almost entirely as a mobile anion" (Jeffries and Ouimet, 2005, p. 344). The role of N deposition, however, is more complex since (a) nitrogen exists in many chemical forms and (b) it is very often the growth-limiting nutrient for temperate forest vegetation, so that atmospheric N deposited to this ecosystem is generally retained (e.g., Driscoll et al., 2001; Jeffries et al., 2005). If, however, sufficient N is available in forest soil so that it is no longer growth-limiting, a condition known as "nitrogen saturation" (e.g., Aber et al., 1998), then NO_3^- (including NH_4^+ converted to NO_3^- via a soil biological process known as nitrification) becomes a mobile anion and acts as an acidifying agent just like SO_4^{2-}.

Recent studies suggest that no Canadian forests are currently nitrogen saturated (Houle, 2005; Jeffries et al., 2005), but the capacity of forest ecosystems to accumulate N is finite so that regions experiencing elevated levels of N deposition will become saturated at some time in the future. Some forests in the eastern U.S. are already nitrogen saturated (e.g., Driscoll et al., 2001; McNulty et al., 2007). Note that Jeffries (1995) has suggested that annual N deposition greater than 10 kg N/ha/y may be considered to be "elevated".

As a consequence, two atmospheric acidity inputs are considered in this paper: (1) acidity due to S deposition alone; and (2) acidity due to both S and N deposition, where both oxidized and reduced N species are included. The former is appropriate for current Canadian conditions, where N is retained in forest ecosystems, while the latter corresponds to future N-saturated conditions following accumulation of N in soils from ongoing elevated atmospheric N deposition.

3. AURAMS 2002 ANNUAL SIMULATION

A one-year simulation has been performed for 2002 with a regional, size- and composition-resolved particulate-matter (PM) modelling system named AURAMS (A Unified Regional Air-quality Modelling System). Hourly wet and dry deposition fields were predicted by AURAMS on a North American domain for each hour of 2002 for a number of sulphur (S) and nitrogen (N) species, were output, and were then post-processed for the entire period to obtain the annual total-sulphur (t-S) total deposition and total-nitrogen (t-N) total deposition fields. These two fields were required in order to estimate ADCL exceedances across Canada for 2002 using the Canada-wide multi-ecosystem ADCL field shown in Figure 1. The AURAMS modelling system, the methodology followed for the 2002 annual simulation, and the additional post-processing steps are described below.

3.1 Modelling System Description

AURAMS consists of three main components: (a) a prognostic meteorological model, GEM; (b) an emissions processing system, SMOKE; and (c) an off-line regional chemical transport model, the AURAMS CTM.

The EC Global Environmental Multiscale (GEM) meteorological model is an integrated weather forecasting and data assimilation system designed to meet Canada's operational needs for both short- and medium-range weather forecasts (Côté et al., 1998a,b). Two versions of GEM are run operationally for different grid configurations. One is a uniform latitude/longitude resolution grid; the other is a variable-resolution, rotated latitude-longitude grid with finer uniform resolution in a core region positioned over a selected portion of the globe. The first grid configuration is used for GEM medium-range (i.e., two-week) forecasts; the second configuration is used for GEM short-term (i.e., two-day) forecasts, where the uniform core region is located over North America and its adjacent waters. The variable-resolution configuration of GEM is used for AURAMS (e.g., Figure 2).

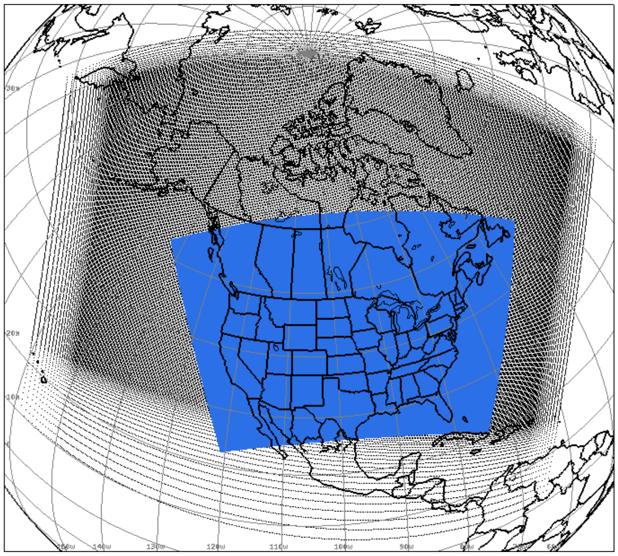


Figure 2. Locations of GEM (shown in black) and AURAMS CTM (shown in blue) grid domains.

The Sparse Matrix Operator Kernel Emissions (SMOKE) processing system (e.g., Houyoux et al., 2000; CEMPD, 2008) is used to process Canadian, U.S., and Mexican national emission inventories, which contain annual emissions of a small number of criteria air contaminants (CACs) reported on a jurisdictional basis (e.g., province, state, or county), in order to create hourly gridded emission fields of AURAMS model chemical species on the AURAMS CTM grid.

The multi-pollutant, regional AURAMS CTM was designed to simulate tropospheric ozone, PM, and acid deposition in a single "unified" modelling framework. The PM size distribution is represented using a sectional approach by 12 size bins ranging from 0.01 to 41 μm in geometric diameter. PM composition is represented with nine chemical components: sulphate (SO4); nitrate (NO3); ammonium (NH4); elemental carbon; primary organic matter (POM); secondary organic matter (SOM); crustal material (CM); sea salt; and particle-bound water. PM is assumed to be internally mixed within each size bin. CTM process representations include emissions from surface and elevated sources, grid-scale transport and vertical diffusion, gas-phase, aqueous-phase, and inorganic heterogeneous chemistry, SOM formation, particle nucleation, condensation, coagulation, and activation, and dry and wet deposition of gases and particles (e.g., Zhang et al., 2001, 2002; Gong et al., 2003; Makar et al., 2003; Gong et al., 2006).

The version of the AURAMS CTM that was used for the 2002 annual simulation described here was a prototype of version 1.3.2, the newest version of the model. One significant new feature of version 1.3.2 is the addition of time-invariant chemical lateral boundary conditions (CLBC). This new feature was used in this simulation instead of the zero-gradient CLBCs used in two previous 2002 annual simulations run with version 1.3.1 of AURAMS (e.g., Moran et al., 2007).

3.2 Modelling System Application

To simulate one full year (2002) of acid deposition, the hourly gridded anthropogenic emissions files needed by the AURAMS CTM for the simulation were prepared first from the 2002 Canadian, 2002 U.S., and 1999 Mexican national CAC emissions inventories using version 2.2 of the SMOKE emissions processing system. The SMOKE "target" grid was the AURAMS 42-km continental-scale grid shown in Figure 2. This 150x106 uniform horizontal grid with 42-km grid spacing is superimposed on a secant polar stereographic projection true at 60°N. Hourly biogenic emission fields, on the other hand, were predicted by the CTM during the simulation based on BEIS v3.09 algorithms and GEM meteorological fields.

Next, GEM and then the AURAMS CTM were run from 20 Dec. 2001 until 31 Dec. 2002; the reason for the early start was to reduce any model "spin-up" effects. GEM version 3.2.0 with physics version 4.2 was used with a 353x415 variable-resolution horizontal grid on a rotated latitude-longitude map projection (Figure 2). Grid spacing was approximately 24 km (0.22°) on the 270x353 uniform regional "core" grid centred on North America and larger elsewhere. In the vertical, 28 hybrid-coordinate levels stretched from the Earth's surface to 10 hPa with vertical spacing increasing monotonically with height. The lowest three GEM vertical sigma levels were located at 1.000, 0.995, and 0.985. An integration time step of 450 s was used. GEM was run in 30-h segments from analyzed meteorological fields every 24 hours, where the first six hours of each segment were treated as a "spin-up" period and were discarded. The remaining 24 hours of each successive simulation were then "stitched" together to create a complete set of hourly meteorological fields for 2002 for input to the AURAMS CTM.

A prototype of version 1.3.2 of the AURAMS CTM was then run on the AURAMS 42-km continental-scale horizontal grid shown in Figure 2. In the vertical, 28 terrain-following levels reached from the Earth's surface to 29 km. The lowest three vertical levels were located at 0, 15, and 55 meters. Climatological time-invariant CLBCs were used; most of the species profiles were "clean" background profiles interpolated from ADOM boundary profiles (see Fung et al., 1991), with the exceptions of CO and O_3, for which spatially-varying monthly values based on measurements were used, and p-SO_4, p-NO_3, p-NH_4, p-POM, and p-CM, for which mean measured values from a coastal mountain site in British Columbia (Whistler) were used. An integration time step of 900 s was used, and AURAMS-predicted fields were output once an hour. The CTM was run in 12-hour segments and predicted fields were output once per hour.

3.3 Model Output Post-processing

Hourly AURAMS CTM predictions from this 2002 simulation were post-processed to obtain (a) annual mean species concentration fields and accumulated wet deposition fields for evaluation against 2002 air-chemistry and precipitation-chemistry network measurements (see next section) and (b) annual total-sulphur (t-S) total deposition and annual total-nitrogen (t-N) total deposition fields for use in ADCL exceedance calculations.

A number of post-processing steps must be carried out in order to prepare the average or accumulated or aggregated fields needed for model evaluation or for ADCL exceedance calculations. The first step is to match time periods. AURAMS output fields are available for every hour of simulation whereas acid deposition is usually examined at the seasonal or annual scale. AURAMS hourly concentration fields must thus be averaged over all hours to obtain annual mean concentration fields and hourly wet and dry deposition flux fields must be summed or accumulated over all hours to obtain annual wet and dry deposition fluxes.

It is also useful to calculate a number of "aggregated" species concentration and deposition fields in order (a) to match measured quantities and (b) to obtain the wet plus dry deposition fluxes of multiple S and N species needed for ADCL exceedance calculations. For example, PM network measurements are made for selected particle size ranges in terms of aerodynamic diameter whereas AURAMS predicts PM concentrations in terms of 12 size bins or size sections specified by Stokes radius (e.g., Seigneur and Moran, 2003). For acid-deposition calculations, for which all particle sizes must be considered, AURAMS PM chemical components are summed over all 12 size bins to obtain, for example, total particle sulphate (p-SO_4), particle nitrate (p-NO_3), and particle ammonium (p-NH_4) concentration or deposition.

Similarly, to estimate the total acid-deposition impacts of all S and N species, the following six wet deposition fluxes (SO_2, p-SO_4, HNO_3, p-NO_3, NH_3, p-NH_4) and 10 dry deposition fluxes (SO_2, H_2SO_4, p-SO_4, NO_2, HNO_3, RNO_3, p-NO_3, PAN, NH_3, p-NH_4) predicted by AURAMS need to be considered. (RNO_3 is the AURAMS organic nitrate species.) Note that the list of dry deposition fluxes includes both the gas and particle phases of three species: H_2SO_4, HNO_3, and NH_3. Note too that it is assumed that five other AURAMS N species (NO, HONO, HNO4, N2O5, RO2N) neither dry deposit nor wet deposit.

The calculation of t-S and t-N total deposition fields also becomes more manageable and the contributions of different species easier to compare if the following intermediate aggregated fields are defined:

- t-SO_4 (sum of H_2SO_4 and p-SO_4 in SO_4 units);
- t-S (sum of SO_2 and t-SO_4 in S units);
- t-NO_3 (sum of HNO_3, RNO_3, and p-NO_3 in NO_3 units);
- t-NH_3 (sum of NH_3 and p-NH_4 in NH_3 units: for air chemistry);
- t-NH_4 (sum of NH_3 and p-NH_4 in NH_4 units: for precipitation chemistry);
- t-N_{OX} (sum of NO_2, PAN, t-NO_3 in N units);
- t-N_R (same as t-NH_3 but in N units);
- t-N (sum of t-N_{OX} and t-N_R in N units)

where "N_{ox}" denotes "oxidized N" and "N_R" denotes "reduced N". Note that Zhang et al. (2005) found that NO_2 and PAN both make non-negligible contributions to oxidized N deposition.

Finally, units must also be matched. AURAMS outputs abundance fields in units of µg kg^{-1} at ambient conditions. These units have been converted to units of ppbV for gas-phase species and µg m^{-3} STP (0°C) for particle-phase and mixed-phase species with the aid of GEM-predicted temperature and pressure fields. AURAMS outputs wet and dry deposition fluxes in units of µmoles m^{-2} h^{-1}, and these fluxes have been converted to units of kg ha^{-1} y^{-1}.

3.4 Measurement Data for Model Evaluation

Gas-phase air-chemistry measurements for 2002 from five networks, two in Canada (CAPMoN, NAPS) and three in the U.S. (AQS, CASTNet, STN), were obtained for AURAMS performance evaluation. PM air-chemistry measurements were obtained from eight networks and subnetworks, three in Canada (CAPMoN, NAPS-continuous, NAPS-filter) and five in the U.S. (AQS-continuous, AQS-filter, AQS-STN, CASTNet, and IMPROVE). Figures 3 and 4 show the locations of stations that measured air concentrations of a number of gas-phase and particle-phase species in 2002.

Note that the amalgamation of measurements from multiple air-chemistry networks is quite heterogeneous, since individual networks have different goals and objectives, choose different types of sampling locations, employ different sampling techniques and protocols, and measure different species (e.g., Eder and Yu, 2006). For example, some networks report hourly values, some report daily averages every third day, and one reports weekly averages. For this reason, AURAMS predictions for 2002 have been evaluated with individual network measurements as well as with multi-network measurements (see Moran et al., 2008), but only selected results from the multi-network comparisons are shown here.

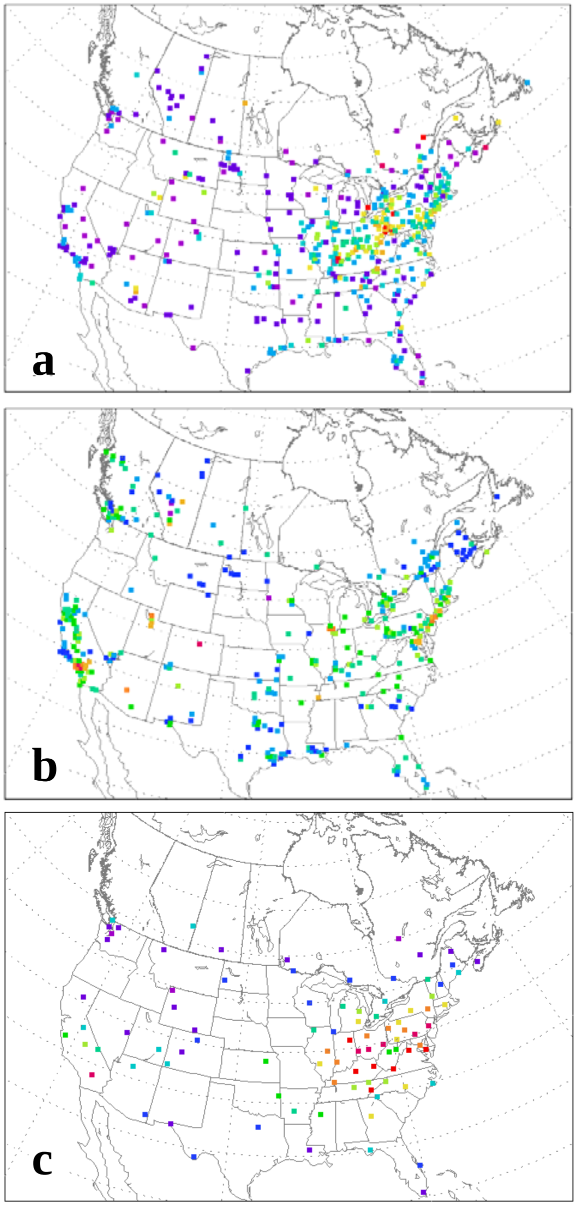


Figure 3. Locations of air-chemistry stations in 2002 measuring (a) SO_2, (b) NO_2, and (c) HNO_3 air concentrations. The colour of the symbol marking each station location indicates annual concentration value (cf. Moran et al., 2008).

Precipitation-chemistry measurements for 2002 were obtained from eight networks and subnetworks, six in Canada (BCPCSN, CAPMoN, NBPMN, NSPSN, PQMPA, REPQ) and two in the U.S. (NADP-AIRMoN, NADP-NTN). There is greater uniformity in sampling protocols and operating procedures across the precipitation-chemistry networks, but some precipitation-chemistry networks perform daily sampling, some perform weekly sampling, and some perform both. Figure 5 shows the locations of stations operating in 2002 for these eight networks and subnetworks. Completeness criteria were imposed on station measurement data of each species to ensure that annual means calculated with the valid measurements available for each station would be representative of an annual value. For air-chemistry station measurements, at least 75% of 2002 samples at a station had to be valid to be used for model evaluation, and for precipitation-chemistry station measurements, a station must have operated for at least 90% of 2002 and the percent total precipitation of valid species samples had to be at least 70% of measured annual precipitation. Unit conversions were also

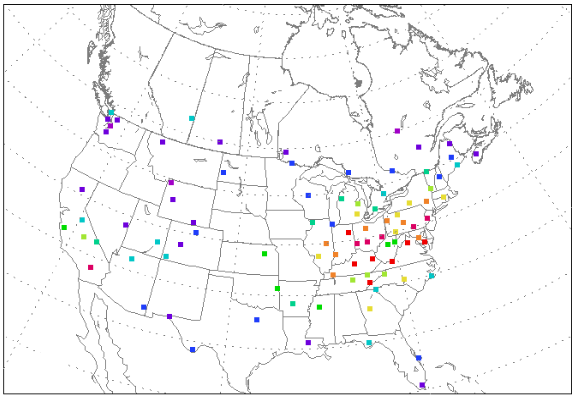


Figure 4. Same as Figure 3 but for locations of air-chemistry stations in 2002 measuring p-SO_4, p-NO_3, and p-NH_4 air concentrations.

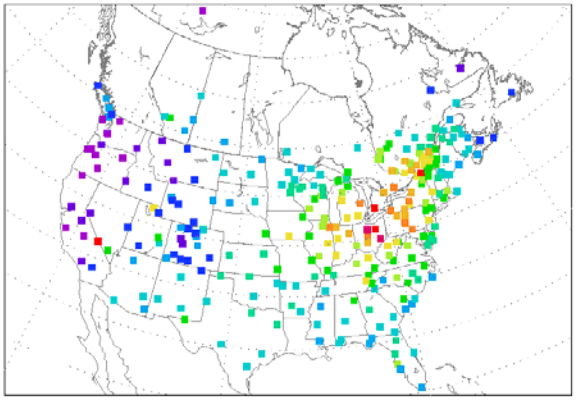


Figure 5. Same as Figure 3 but for locations of precipitation-chemistry stations in 2002 measuring precipitation amount and $SO_4^=$, NO_3^-, and NH_4^+ wet deposition.

performed on the calculated annual measured values so that all AURAMS air-concentration predictions and network measurements could be compared in ppbV for gas-phase species and in µg m^{-3} STP (0°C) for PM species and mixed-phase species.

4. RESULTS

This section first shows results from an evaluation of AURAMS skill in predicting a number of acid-deposition-related annual air-chemistry and precipitation-chemistry fields. Note that skill in predicting surface air concentration fields is used as a rough proxy for model skill in predicting dry deposition fluxes because the latter are not measured routinely by any North American networks. AURAMS-predicted fields of 2002 annual t-S and t-N total deposition are then used to calculate Canada-wide ADCL exceedances.

4.1 AURAMS Annual Performance Evaluation

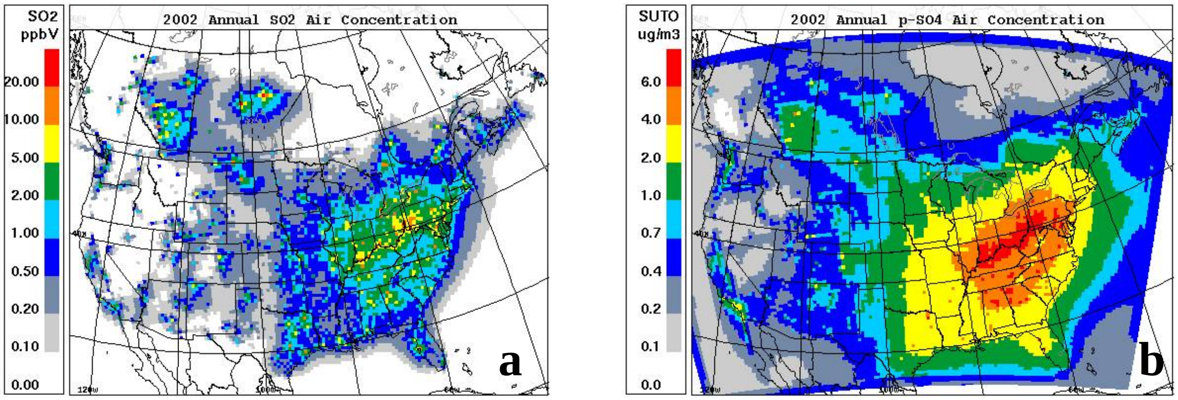
4.1.1 Air-chemistry comparison

Figure 6 shows the spatial distribution of the annual mean surface air concentration fields predicted by AURAMS for 2002 for 10 S and N species associated with acid deposition. Four are gas-phase species (SO_2, NO_2, HNO_3, and NH_3), three are particle-phase species (p-SO_4, p-NO_3, and p-NH_4), and three are aggregated mixed-phase species (t-SO_4, t-NO_3, and t-NH_3).

The spatial distributions of each of these species are broadly similar at the regional scale: that is, higher concentrations tend to be confined to the North American land mass and concentrations tend to be higher in eastern North America than in western North America, consistent with the spatial distribution of SO_2, NO_2, and NH_3 emissions (e.g., NARSTO, 2006; Moran et al., 2008). The spatial patterns of the annual concentrations of these three directly-emitted (i.e., primary) species, however, have sharper gradients and more "structure" or granularity (Figures 6a, 6d, and 6h) than the patterns of the secondary and mixed species in Figure 6. Note also that the elevated levels of the particle-phase species at some of the boundaries, especially the northern boundary, are artifacts due to the values specified for these species for the time-invariant CLBCs. (This simulation was the first test of these new CLBCs for an extended period.)

Air-concentration measurements are available for six of the species in Figure 6, namely SO_2, p-SO_4, NO_2, HNO_3, p-NO_3, and p-NH_4. Figure 7 presents scatterplots for these six species that compare annual predicted vs. measured values for all measurement stations with valid annual values and all networks measuring those species regardless of differences in network operating practices. (Note that measurements of $PM_{2.5}$-SO_4, $PM_{2.5}$-NO_3, and $PM_{2.5}$-NH_4 are also available from other networks but are not shown here.) Measurements from different networks are denoted on these scatterplots by different symbols, and the calculated statistics (correlation coefficient R, root mean square error, normalized mean bias [NMB], and normalized mean error) that are provided follow the definitions given by Eder and Yu (2006). The number of data points varies by species due both to the number of stations measuring the species and to the number of complete annual measurements. For 2002, for example, there were 463 valid SO_2 annual values but only 87 valid HNO_3 annual values.

As expected *a priori*, agreement is less good for the two primary pollutants (SO_2 and NO_2) than for the four secondary pollutants. For SO_2, the values of R and NMB are 0.56 and 7.6%, respectively, and for NO_2 they are 0.19 and -37%. For p-SO_4, on the other hand, the values of R and NMB are



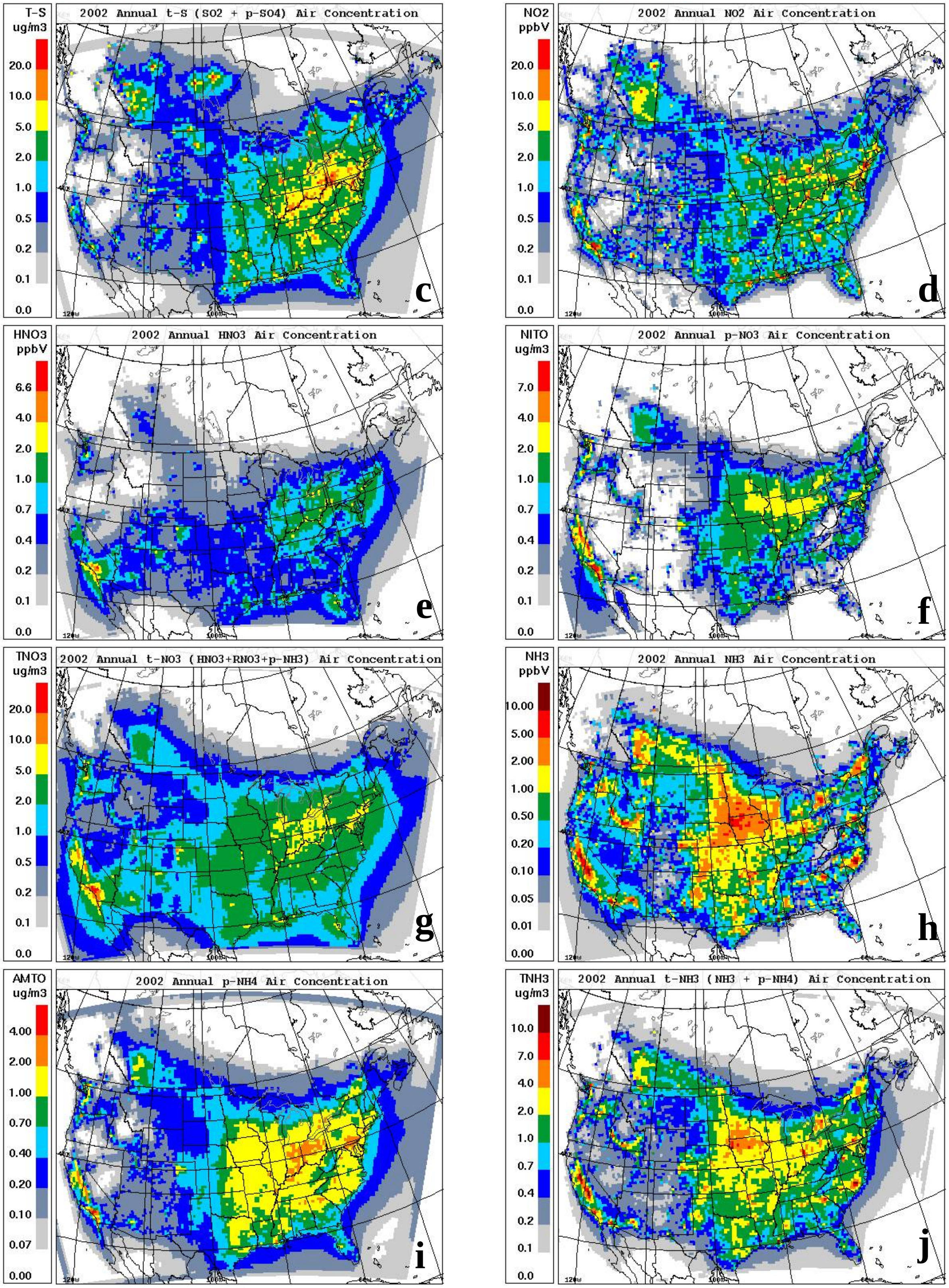


Figure 6. AURAMS-predicted 2002 annual mean surface (a) SO2, (b) p-SO4, (c) t-S, (d) NO2, (e) HNO3, (f) p NO3 (g) t-NO3, (h) NH3, (i) p-NH4 and (j) t-NH3 air concentration fields [gases in units of ppbV, others in units of µg m 3 STP (0°C)].

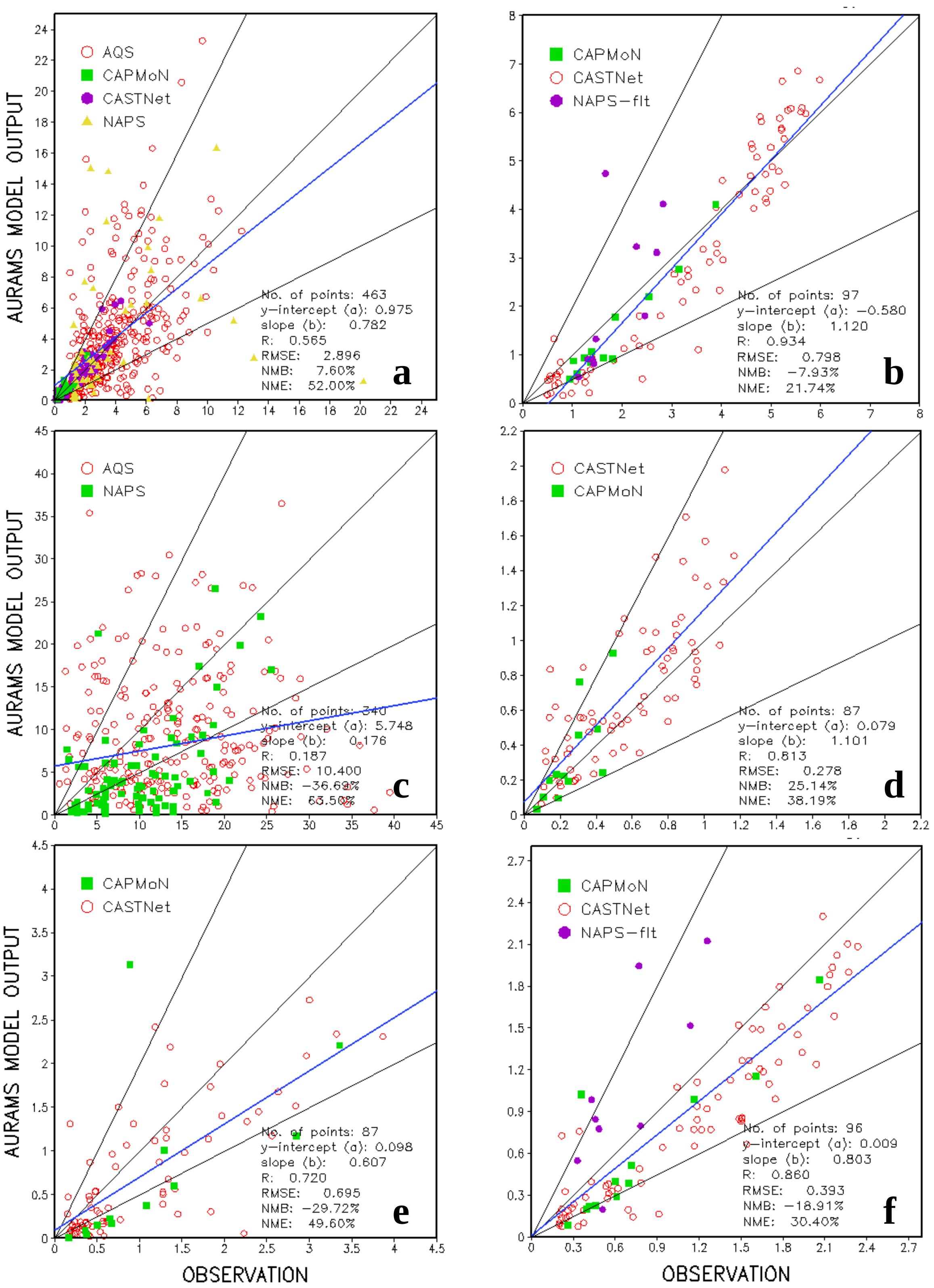


Figure 7. Scatterplots of measured vs. predicted 2002 annual surface (a) SO_2, (b) p-SO_4, (c) NO_2, (d) HNO_3, (e) p-NO_3, and (f) p-NH_4 air concentration fields [gases in units of ppbV, particles in units of µg m^{-3} STP (0°C)]. The 1:1, 1:2, and 2:1 lines are also plotted.

0.93 and -7.9%, for HNO_3 they are 0,.81 and 25%, for p-NO_3 0.72 and -30%, and for p-NH_4 0.86 and -19%. Note that the biases for HNO_3 and p-NO_3 are of opposite sign, suggesting that the predicted partitioning between these two phases may be a contributing factor to the observed errors.

Since routine direct measurements of dry deposition are not available from any North American networks, model performance in predicting surface air concentrations can be viewed as a rough surrogate for model skill in predicting dry deposition. Model skill in predicting wet deposition, on the other hand, can be evaluated directly.

4.1.2 Precipitation-chemistry comparison

Figure 8 shows the spatial distribution of the annual total precipitation field and the annual wet deposition fields for $SO_4^=$, NO_3^-, and NH_4^+ predicted by AURAMS for 2002. The pronounced extrema for precipitation are striking, with local maxima located along the Pacific Northwest coast and over the western Atlantic Ocean and local minima located over the desert regions of the U.S. southwest and northwestern Mexico. This pattern is significant since the wet deposition of pollutants is modulated by the frequency and amount of precipitation. AURAMS predicted the areas of maximum annual wet deposition of $SO_4^=$ and NO_3^- in 2002 to occur over the southern Great Lakes, south-central Canada, and the northeastern U.S. Annual NH_4^+ wet deposition is predicted to be elevated in these same areas but to be even higher in the midwestern U.S. states of Iowa and Minnesota (where NH_3 emissions and air concentrations are high – see Figures 6h and 6j).

Figure 9 presents scatterplots evaluating AURAMS skill in predicting annual precipitation and annual wet deposition of $SO_4^=$, NO_3^-, and NH_4^+. Prediction of annual precipitation by the GEM model is very good, with an R value of 0.85, a small NMB value of 5.8%, and a slope of 0.85 for the best-fit line. The background to this judgement is that precipitation is a notoriously difficult quantity to predict: even annual precipitation exhibits considerable small-scale spatial variation due to the convective nature of

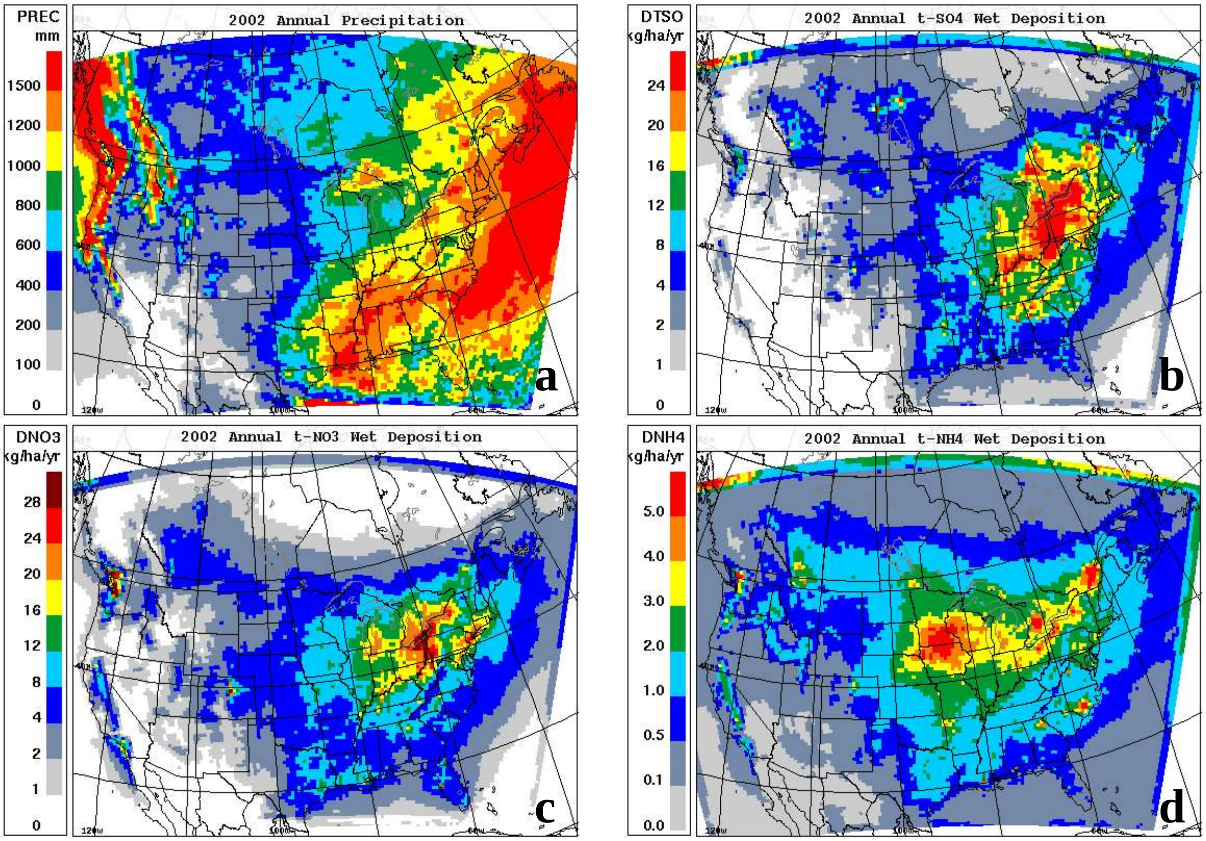


Figure 8. AURAMS-predicted 2002 annual (a) precipitation [mm] and (b) $SO_4^=$, (c) NO_3^-, and (d) NH_4^+ wet deposition fields [kg ha^{-1} y^{-1}].

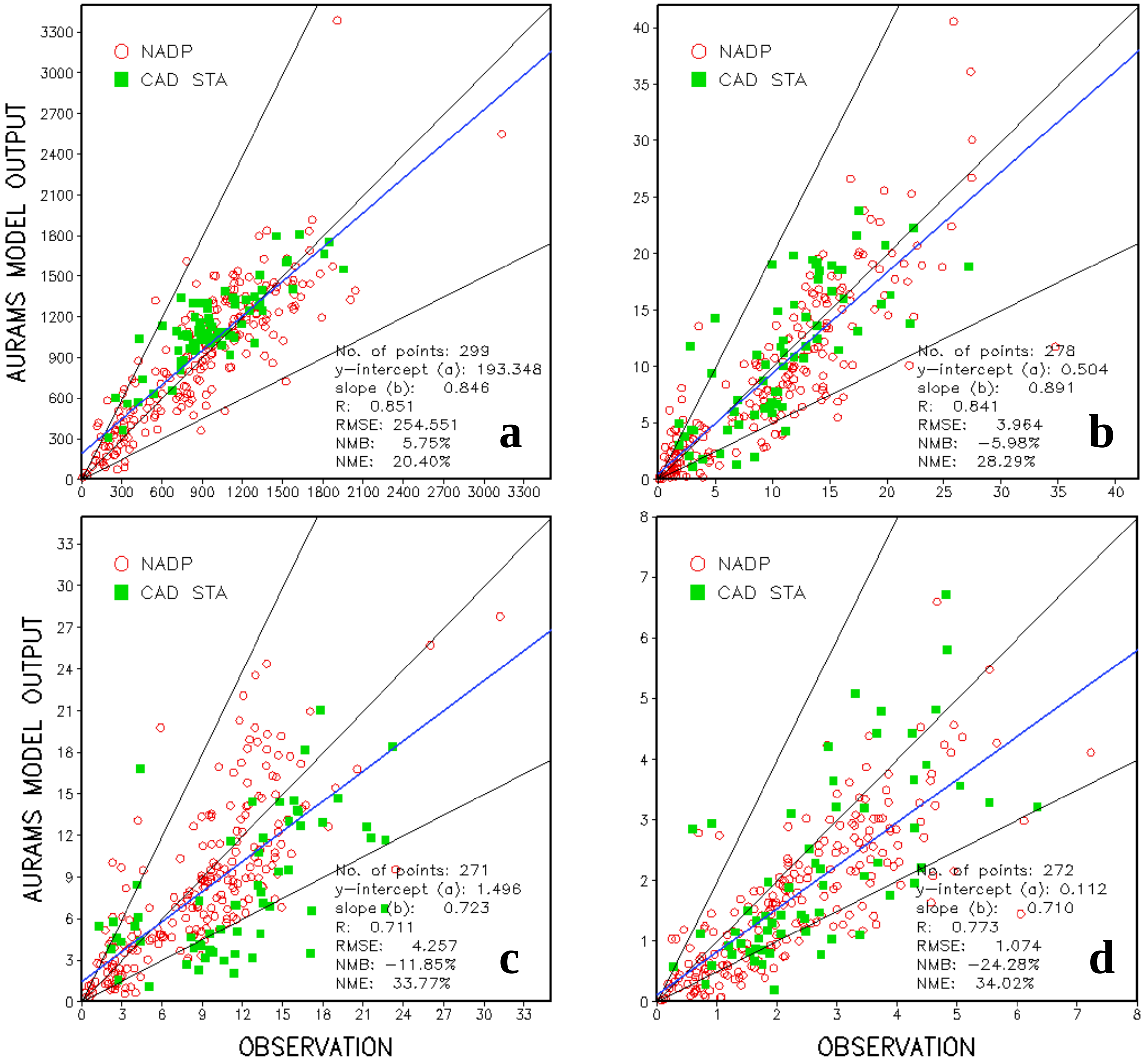


Figure 9. Scatterplots of measured vs. predicted 2002 annual (a) precipitation [mm] and (b) $SO_4^{=}$, (c) NO_3^-, and (d) NH_4^+ wet deposition [kg ha^{-1} y^{-1}]. The 1:1, 1:2, and 2:1 lines are also plotted.

much precipitation and to subgrid-scale variations in orography. Moreover, this comparison is made more difficult due to the incommensurability between point (station) measurements and 42-km grid volume predictions.

Model predictions of $SO_4^{=}$, NO_3^-, and NH_4^+ annual wet deposition are also good, with R values of 0.83, 0.72, and 0.80, respectively, NMB values of -6.0%, -12%, and -24%, and best-fit-line slope values of 0.89, 0.72, and 0.71.

4.2 2002 Critical-Load Exceedances

Following the output post-processing described in Section 3.3, Figures 10a-c show the spatial distributions of the annual t-S, t-N_{OX}, and t-N_R total deposition fields predicted by AURAMS for 2002. These three fields can then be summed to obtain the field of t-S+N total deposition for 2002 (Figure 10d). The four panels of this figure are plotted in identical units (eq ha^{-1} y^{-1}) using the same set of (unevenly spaced) contour intervals to allow the relative contributions of t-S, t-N_{OX}, and t-N_R total acid deposition to be assessed visually. Interestingly, in these units the t-S and t-N_{OX} total deposition fields are predicted to cover roughly the same geographic area (with the exception of the west coast of the U.S.) and to have roughly the same magnitude. The t-N_R total deposition field, on the other hand, is smaller both in geographic extent and in magnitude and is somewhat confined to the major NH_3 emission region in mid-continent.

If it is assumed that only t-S deposition is acidifying, as is approximately the case in Canada at the present time (Houle, 2005; Jeffries et al., 2005), then an ADCL exceedance field for 2002 can be calculated by subtracting the ADCL field shown in Figure 1 from the predicted 2002 t-S total deposition field (Figure 10a). The resulting ADCL exceedance field is shown in Figure 11.

The areas predicted to be in exceedance in 2002 in Figure 11 include much of northeastern and western Newfoundland, Nova Scotia, and southern New Brunswick, parts of southern Quebec and central Ontario, the Lake of the Woods region of northwestern Ontario, and pockets of central Manitoba and Saskatchewan and northeastern Alberta. Interestingly, depending on the region, there are different explanations for the areas predicted to be in ADCL exceedance. In the Maritimes and the Lake of the Woods region of Ontario, the predicted exceedances are due to the combination of very sensitive ecosystems (see Figure 1) and long-range transport. In southern Ontario and central Quebec, exceedances are due to a combination of moderately sensitive ecosystems and elevated levels of t-S total deposition (Figure 10a) due to both local sources and long-range transport. And in the western provinces, exceedances are due to the combination of moderately sensitive ecosystems and a handful of large, isolated local SO_2 emission sources. It should be noted that the exceedances in northeastern Newfoundland may be at least in part a CLBC artifact (Figure 10a), though Jeffries and Ouimet (2005) also showed exceedances in this area for an independent calculation based on measured deposition (see Figure 13).

If both t-S and t-N are assumed to be acidifying (i.e., N saturation case), then a second ADCL exceedance field for 2002 can be calculated by subtracting the ADCL field shown in Figure 1 from the 2002 t-S+N total deposition field (Figure 10d). The resulting ADCL exceedance field is shown in Figure 12.

As would be expected from a comparison of Figure 10d and Figure 10a, the extent of the areas of Canada predicted to be in exceedance of ADCL due to t-S+N total deposition is considerably larger

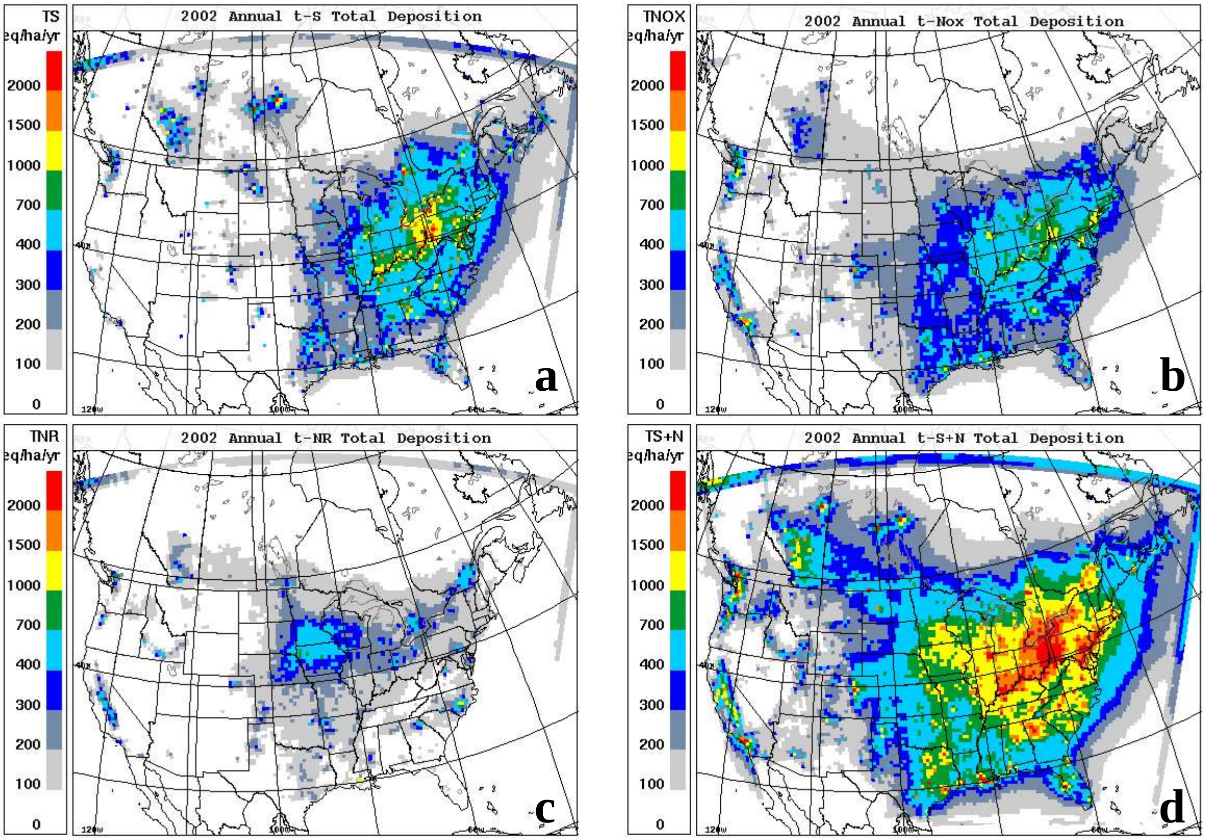


Figure 10. AURAMS-predicted 2002 annual (a) t-S, (b) t-N_{OX}, (c) t-N_R, and (d) t-S+N total deposition fields [eq ha^{-1} y^{-1}].

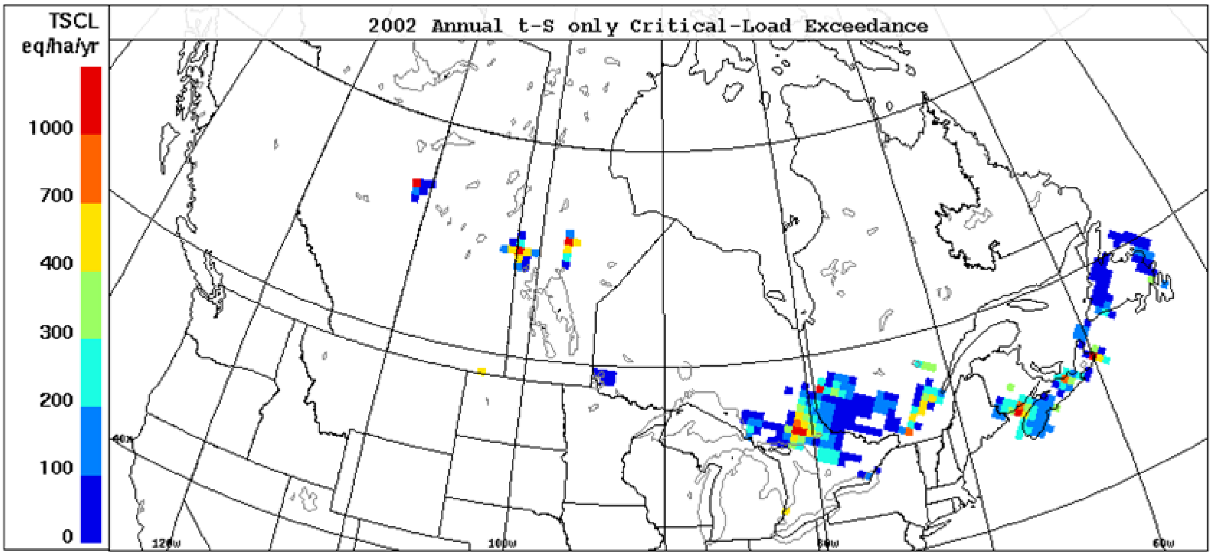


Figure 11. Predicted 2002 t-S-only ADCL exceedance field for Canada [eq ha^{-1} y^{-1}] (based on Figure 8.8a of Jeffries and Ouimet [2005]).

(Figure 12) than was the case for t-S total deposition alone (Figure 11). All of the areas that were predicted to be in exceedance in Figure 11 have expanded in size. Note that the new exceedance area predicted in the Northwest Territories is very likely a CLBC artifact (cf. Figure 10d), but the new exceedance area predicted in the Lower Fraser Valley of southwestern British Columbia is associated with local acidic emissions and is not an artifact.

5. DISCUSSION

The performance evaluation of AURAMS predictions of 2002 annual acid-deposition-related fields reported in Section 4.1 suggests that on an annual basis the model displayed considerable skill in predicting fields contributing to acid deposition. These results provide support for the use of AURAMS-predicted annual t-S and t-S+N total deposition fields in Section 4.2 to calculate ADCL exceedance fields for Canada.

There are several implications from those two ADCL exceedance fields (Figures 11 and 12). First and foremost is that the SO_2 emission reductions that have occurred in both Canada and the U.S. over the past 15 to 20 years have

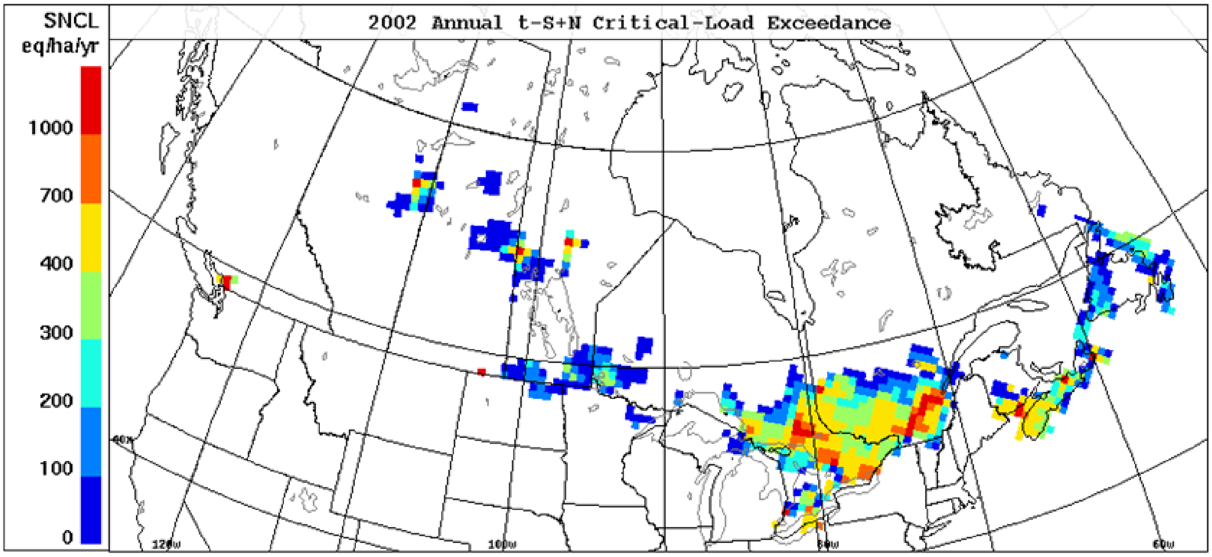


Figure 12. Predicted 2002 t-S+N ADCL exceedance field for Canada [eq ha^{-1} y^{-1}].

reduced acid deposition but they have not “solved” the acid- deposition problem in eastern Canada.

Second, it has been assumed that damaging levels of acid deposition were restricted to eastern Canada, but there were insufficient data available for western Canada to confirm that expectation. There are still few measurements of acid deposition available for western Canada (e.g., Vet et al., 2005), but the extension of the forest-soil ADCL field into western Canada (Aherne and Watmough, 2006), which provides a systematic identification in much of the region of sensitive forest ecosystems, and the availability of AURAMS model predictions for the region, permit an assessment of current acid-deposition impacts in western Canada.

Figure 11 suggests that some areas of western Canada in the vicinity of large SO_2 sources may in fact be in exceedance of their ADCL values. This is a concern if borne out because, unlike eastern North America, emissions of acidifying emissions are projected to increase in the next decade in western Canada (e.g. Environment Canada, 2008). At this point, however, the model results can only be considered suggestive due to the approximations involved in modelling pollutant dispersion from single point sources with a regional-scale acid-deposition model with 42-km grid spacing.

It is also of interest to compare the ADCL exceedance fields predicted by AURAMS for 2002 with an earlier pair of ADCL exceedance fields that were calculated based on an earlier version of the Canadian national ADCL field and measured mean annual $SO_4^=$, NO_3^-, and NH_4^+ wet deposition station data and estimated dry deposition of these species and SO_2 and HNO_3 for the five-year period 1994-98. Figures 13 and 14 are the resulting plots of ADCL exceedance fields that appeared in the *2004 Canadian Acid Deposition Science Assessment* (Jeffries and Ouimet, 2005). These earlier ADCL exceedance fields were limited by a lack of both acid-deposition measurements and ADCL values in western Canada, but in eastern Canada there is good qualitative agreement between Figures 11 and 13 and between Figures 12 and 14.

It should be noted that there are some methodological differences underlying these two sets of ADCL exceedance fields that can explain some of the observed differences between them. For one thing, SO_2 emissions were higher in eastern North America during the 1994-1998 period than in 2002 (e.g., Vet et al., 2005). The ADCL exceedance field shown in Figure 13 also included a contribution to effective acid deposition from N leaching that was excluded from the calculation for Figure 11. The ADCL exceedance calculation for Figure 12, on the other hand, included contributions to t-N total deposition from some N species (PAN, NH_3, and NO_2 dry deposition) that were predicted by AURAMS but

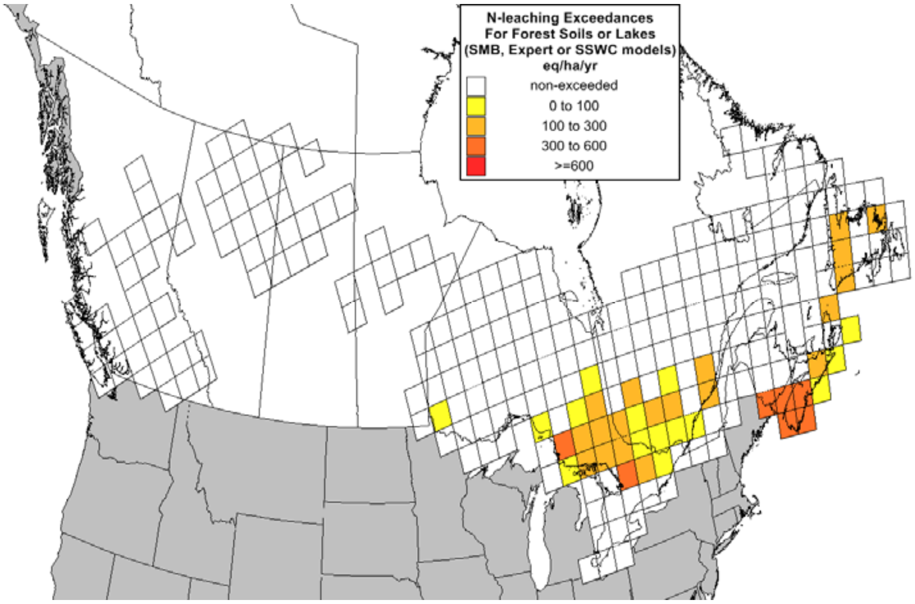


Figure 13. Measured 1994-98 N-leaching ADCL exceedance field for Canada [eq ha^{-1} y^{-1}].

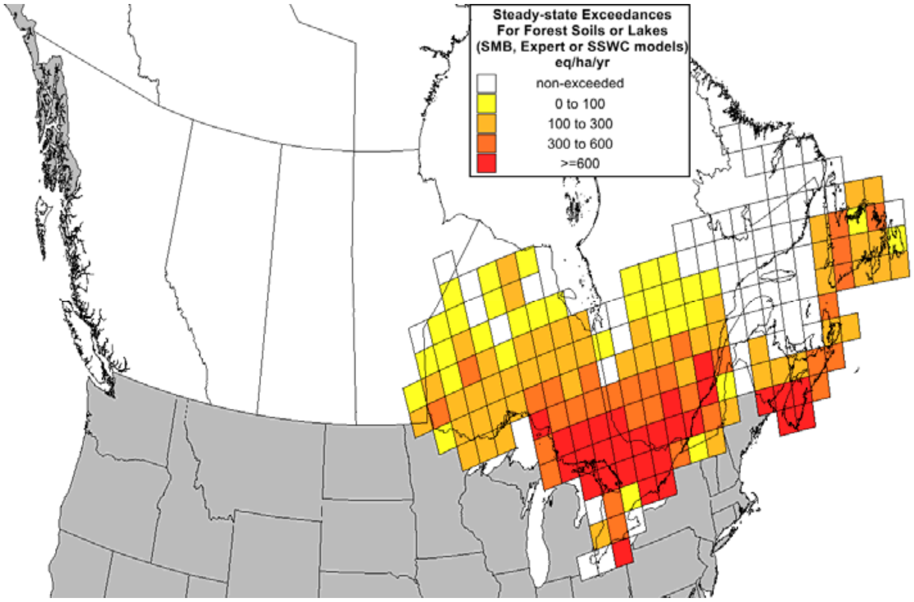


Figure 14. Measured 1994-98 t-S+N ADCL exceedance field for Canada [eq ha^{-1} y^{-1}] (based on Figure 8.8b of Jeffries and Ouimet [2005]).

for which no network data were available and hence were not included in the calculations that produced Figure 14 (Jeffries and Ouimet, 2005, p. 347).

Acid deposition models like AURAMS can also be used to simulate future-year emission scenarios so that the changes in air quality and acid deposition resulting from changes in emissions can be assessed (e.g., Moran, 2005). AURAMS is now being applied to simulate such scenarios, and the methods described in this paper are being used to assess the environmental impacts from changes in acid deposition predicted by AURAMS along with human health impacts due to changes in air pollutant concentrations.

6. SUMMARY AND CONCLUSIONS

The acid-deposition critical-load approach provides a valuable tool to assess the environmental impact of S and N emissions. This approach has been used in studies of acid-deposition impacts in Canada since the 1990s, but the results of recent research have expanded and enhanced its usefulness. Specifically, a combined aquatic-forest ADCL map is now available for most of sub-Arctic Canada, thus allowing the acid-deposition issue to be considered at the national scale for multiple ecosystems.

To calculate ADCL exceedance fields, which identify areas at risk due to acid deposition, measured or modelled estimates of annual total deposition of S species, oxidized N species, and reduced N species are also required. Measurements of course provide “ground truth” at instrument locations, but acid-deposition models have the twin advantages of being able to provide estimates of acid-deposition levels (a) for areas where few or no measurements exist and (b) for future conditions if future levels of acidifying emissions can be specified.

This paper provides a demonstration of the usefulness of regional acid-deposition models for calculating ADCL exceedance fields. The EC AURAMS model has been run for one full year (2002) and the model’s performance has been evaluated. It was found that AURAMS was able to predict annual S and N concentration and deposition fields important for acid deposition with considerable skill. A number of the evaluation results are shown in this paper.

Two Canada-wide ADCL exceedance calculations were then performed based on the AURAMS annual total deposition predictions, one for annual sulphur-only total deposition and one for annual t-S+N total deposition. Consistent with recent measurement-based ADCL exceedance fields, sizeable areas of eastern Canada were predicted to be in exceedance of ADCL values for both calculations, but so too were some parts of western Canada. This latter prediction, though preliminary, is significant because acid deposition was not thought to be an environmental concern in this region and SO_2 and NO_x emissions are projected to increase in western Canada over the next few decades. Unfortunately, atmospheric acid deposition measurements are not available for the predicted exceedance areas in western Canada, but new lake-chemistry data that should provide some “ground truth” are being collected in this region.

It is also evident that ADCL exceedance results must be interpreted carefully since arguments can be advanced for specifying either one of two atmospheric acidity inputs in the calculations, i.e., t-S total deposition or t-S+N total deposition. The former allows an assessment of current ongoing damage whereas the latter also identifies areas that are potentially at risk of damage from acid deposition if current levels continue into the future.

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