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Evaluation of atmospheric nitrogen deposition model performance in the context of U.S. critical load assessments

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HIGHLIGHTS

- A novel performance assessment approach was developed to inform critical load applications.
- Model estimates were compared to wet and bulk inorganic N deposition measurements.
- Model bias and error were expressed as a percentage of regional critical load values.
- Bias was large relative to or even exceeded critical loads in some cases.
- This approach may help assess confidence in critical load and exceedance calculations.

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ABSTRACT

Air quality models are widely used to estimate pollutant deposition rates and thereby calculate critical loads and critical load exceedances (model deposition > critical load). However, model operational performance is not always quantified specifically to inform these applications. We developed a performance assessment approach designed to inform critical load and exceedance calculations, and applied it to the Pacific Northwest region of the U.S. We quantified wet inorganic N deposition performance of several widely-used air quality models, including five different Community Multiscale Air Quality Model (CMAQ) simulations, the Tdep model, and 'PRISM x NTN' model. Modeled wet inorganic N deposition estimates were compared to wet inorganic N deposition measurements at 16 National Trends Network (NTN) monitoring sites, and to annual bulk inorganic N deposition measurements at Mount Rainier National Park. Model bias (model − observed) and error (|model − observed|) were expressed as a percentage of regional critical load values for diatoms and lichens. This novel approach demonstrated that wet inorganic N deposition bias in the Pacific Northwest approached or exceeded 100% of regional diatom and lichen critical load values at several individual monitoring sites, and approached or exceeded 50% of critical loads when averaged regionally. Even models that adjusted deposition estimates based on deposition measurements to reduce bias or that spatially-interpolated measurement data, had bias that approached or exceeded critical loads at some locations. While wet inorganic N deposition model bias is only one source of uncertainty that can affect critical load and exceedance calculations, results demonstrate expressing bias as a percentage of critical loads at a spatial scale consistent with calculations may be a useful exercise for those performing calculations. It may help decide if model performance is adequate for a particular calculation, help assess confidence in calculation results, and highlight cases where a non-deterministic approach may be needed.

1. Introduction

Air quality models are frequently used to estimate pollutant deposition rates and thereby calculate critical loads and critical load exceedances. A critical load is a threshold pollutant deposition rate

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that induces a specific ecological change (Burns et al., 2008; Nilsson and Grennfelt, 1988). A critical load exceedance occurs if ambient deposition is greater than a critical load, and implies ecological changes specified by the critical load are likely. Exceedance calculations are used in international agreements addressing transboundary pollution in Europe (CLRTAP, 2004), and are used by air quality managers to inform decision making in many countries, including the United States (U.S) (USFS et al., 2011). When modeled deposition rates are used to calculate critical loads or exceedances, it is important to consider if model operational performance is adequate for these specific applications. If model bias (model − observed) is large relative to critical load values, calculated critical loads may have large uncertainty, and exceedance calculations could yield false positive or false negative exceedance determinations if model bias is not considered.

Critical load calculations may use modeled deposition rates, deposition measurements, or experimental pollutant additions to define deposition rates associated with ecological changes (Pardo et al., 2011; Vries et al., 2015). Critical load exceedances are calculated as:

$$\text{Exceedance} = P_{dep} - CL \quad (1)$$

where P_{dep} is a pollutant deposition rate, CL is a critical load for the pollutant, and both have the same units (typically kg pollutant ha^{-1} yr^{-1}). Equation (1) may be applied to a single location such as a watershed, or, more frequently, to each grid cell of a Eulerian air quality model within a region of interest. Equation (1) can be parameterized using either a deterministic approach, where discrete values are used for P_{dep} and CL in each grid cell, or a non-deterministic approach, where Monte Carlo or other methods generate a probability distribution of deposition rates, critical load values, and exceedances within each grid cell (Heywood et al., 2006; Page et al., 2008; Skeffington, 2006).

In the United States (U.S.), law does not require critical load and exceedance calculations, but government agencies use deterministic exceedance calculations to protect ecosystems from adverse effects of atmospheric nitrogen deposition. The National Park Service (NPS), U.S. Forest Service (USFS), and U.S. Fish and Wildlife Service calculate deterministic nitrogen deposition exceedances within individual protected land units to evaluate the potential impacts of proposed emissions sources under the Clean Air Act (U.S. Forest Service et al., 2011). USFS also calculates deterministic exceedances within individual USFS land units when developing forest management plans (U.S. Forest Service, 2016). Both USFS and NPS also use exceedances to help monitor air quality status and trends at land-unit and regional scales. At the national scale, the U.S. Environmental Protection Agency (USEPA) has evaluated the adequacy of current secondary National Ambient Air Quality Standards (NAAQS) for nitrogen oxides in part based on deterministic exceedance calculations (USEPA, 2009, 2008).

Although modeled deposition rates are widely used in the U.S. to calculate critical loads and exceedances (Ellis et al., 2013; Lee et al., 2016; Pardo et al., 2011), for many models used in U.S. calculations, operational performance either has not been quantified at all, or has not been quantified specifically to inform critical load and exceedance calculations. Model simulations are typically run at regional or national spatial scales, and performance statistics are calculated across the entire domain to describe overall model performance (Appel et al., 2011; Ellis et al., 2013; Simon et al., 2012). In contrast, critical loads and exceedances may be calculated at scales ranging from local to national. For example, exceedances are calculated for individual national parks and national forests using only a small subset of grid cells within a model domain. Performance statistics are also typically calculated on seasonal time scales for ammonium and nitrate deposition separately (Lee et al., 2016; Simon et al., 2012), whereas critical loads and exceedances are calculated on annual time scale, in units of kg total N ha^{-1} yr^{-1} or kg wet inorganic N ha^{-1} yr^{-1}. Widely-used performance statistics such as normalized mean bias and normalized mean error (Simon et al., 2012) can also be difficult to interpret when the spatial scale of statistics does not match that of calculations. Thus, although conventional performance analyses provide relevant information about a model's ability to simulate deposition processes, there is also a need to quantify operational performance specifically to inform critical load and exceedance calculations.

The objective of this study was to quantify nitrogen deposition operational performance of several air quality models widely used in U.S. critical loads assessments (Table 1), using an approach specifically designed to inform critical load and exceedance calculations. Analyses focused on the Pacific Northwest U.S., where both deposition rates and critical loads are low and of similar magnitude (1–5 kg N ha^{-1} yr^{-1}) (Figs. 1 and 2). We calculated annual wet inorganic N deposition model bias and error at 17 regional monitoring sites, and expressed bias and error as a percentage of regional critical load values for diatoms and lichens. We only quantified model wet inorganic N deposition bias because necessary measurement data are not available for other deposition components. Results suggest expressing model bias as a percentage of critical load values may be informative when selecting a model to use in critical load or exceedance calculations or interpreting calculation results.

2. Methods

2.1. Model descriptions

2.1.1. CMAQ

We evaluated the N deposition performance of five Community Multiscale Air Quality Model (CMAQ) (Byun and Schere, 2006) simulations. CMAQ is an Eulerian model that requires gridded meteorological data, pollutant emissions, and chemical and meteorological initial and boundary condition inputs, which are used to simulate atmospheric physical and chemical processes, and predict pollutant concentrations and deposition. CMAQ has been widely used in U.S. critical loads assessments (Geiser et al., 2010; Pardo et al., 2011), and in the evaluation of the adequacy of secondary NAAQS for protecting against adverse ecological effects of N deposition (Greaver et al., 2012; Scheffe et al., 2014; USEPA, 2008).

CMAQ simulations evaluated in this study vary based on model version, model domain, grid scale, time period, model inputs (including meteorology and emissions), and chemical and atmospheric processes. Each model configuration is described in Appendix B. Three CMAQ simulations were specific to the Pacific Northwest. AIRPACT-3 (2008, 12-km grid) and AIRPACT-4 (2013–2014, 4-km grid) are CMAQ simulations run to generate hourly forecasts of pollutant concentration and deposition for the Pacific Northwest (http://lar.wsu.edu/airpact). Accumulated hourly AIRPACT precipitation and deposition forecast outputs were summed to calculate annual precipitation and N deposition rates at monitoring sites (Fig. 1). EPA-PNW is a 2008 4-km regional simulation conducted by USEPA. Unlike AIRPACT-3 and AIRPACT-4, the EPA-PNW is retrospective, accounts for bi-directional NH_3 exchange and uses lightning strike data to estimate lightning-generated NO_x.

We also evaluated two CMAQ data sets generated by USEPA specifically to provide a time series of deposition estimates for the continental U.S. for use in critical loads assessments (EPA-UNADJ and EPA-ADJ). Each predicts annual deposition for the continental U.S. on a 12-km grid for each year in 2003–2012, and accounts for