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Authors: Brown, Thomas C., and Froemke, Pamela

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Nationwide Maps of Atmospheric Deposition Are Highly Skewed When Based Solely on Wet Deposition

We appreciate the value of the recent article by Brown and Froemke (2012) in providing an updated assessment of nonpoint-source threats to water quality in the United States. However, we highlight some concerns with the measure chosen to quantify atmospheric deposition in the United States. Our primary concern is that the selective use of only atmospheric wet deposition of nitrate plus sulfate as the chosen index of the level of atmospheric deposition perpetuates a long-standing impression that atmospheric deposition is low and of minimal environmental and ecological importance throughout the western United States. Brown and Froemke's figure 2i shows varying levels of elevated deposition in the eastern half of the country, with no areas showing elevated deposition in the West.

The source of the deposition data in Brown and Froemke's article is the National Atmospheric Deposition Program National Trends Network (NADP/NTN), which measures atmospheric wet deposition. The long-term data from the NADP have proven invaluable in advancing our understanding of deposition effects and temporal deposition trends in the United States. However, as in the Brown and Froemke article, NADP data have been frequently used as the sole measure of atmospheric deposition without acknowledging the limitations of measuring only wet deposition. Actual air pollution inputs can be dramatically underestimated when dry deposition is ignored, particularly in areas with semiarid or arid climates.

Dry deposition and deposition in fog or cloudwater—both of which can be major atmospheric deposition inputs—are not measured by the NADP/NTN. The Clean Air Status and Trends Network (CASTNet) is a dry deposition network, with monitoring sites collocated at a subset of the NADP/NTN sites. However, there are insufficient data points in the CASTNet network to create dry deposition maps. Dry deposition fluxes of nitrogen (N) and other pollutants are important in all regions of the United States but can constitute as much as 85 percent or more of the N deposition inputs in arid regions.

Brown and Froemke considered both the acidification and nutrient effects of atmospheric deposition on water quality. The authors mentioned the impressive reductions in sulfate deposition in the eastern United States in past decades. The deposition of nitrate is also decreasing in some areas. However, in the United States, ammonium is becoming an increasingly larger fraction of total atmospheric N deposition, with increasing ammonium deposition trends in many areas (Lehman et al. 2005). However, Brown and Froemke did not include ammonium in their atmospheric deposition metric.

Wet plus dry deposition of both reduced (i.e., ammonia, ammonium) and oxidized (i.e., nitric acid vapor, various nitrogen oxides, nitrate) N forms should be included in any assessment of N deposition effects on water quality, either as a nutrient effect or when considering acidification effects. However, it does not seem appropriate to include sulfate deposition when evaluating nutrient effects on water quality.

We argue that N deposition is not well represented unless dry deposition of N in its various gaseous and particulate forms, including reduced forms of N (e.g., ammonium and ammonia), and, in some cases, cloudwater deposition are also included. Otherwise, a skewed N deposition map will result. An example of a more appropriate portrayal of N deposition in the United States is the total (wet + dry) N deposition map recently published in *BioScience* (Baron et al. 2011, figure 3c).

Finally, the use of the sum of nitrate and sulfate in wet deposition expressed on a mass basis (kilograms per hectare) as a measure of deposition inputs (Brown and Froemke 2012) is also highly unconventional. Each of these ions has a unique molecular weight and on a chemical basis are not appropriately summed unless they are first converted to moles or equivalents (the latter is more appropriate when considering acidification effects; e.g., equivalents per hectare per year).

MARK E. FENN, ANDRZEJ BYTNEROWICZ, AND DANIEL LIPTZIN

Mark E. Fenn (mfenn@fs.fed.us) and Andrzej Bytnerowicz are affiliated with the US Department of Agriculture Forest Service's Pacific Southwest Research Station, in Riverside, California, and Daniel Liptzin is affiliated with the Department of Natural Resources and the Environment at the University of New Hampshire, in Durham.

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Improved Measures of Atmospheric Deposition Have a Negligible Effect on Multivariate Measures of Risk of Water-Quality Impairment: Response from Brown and Froemke

In their comment on our article on nonpoint-source pollution threats to water quality (Brown and Froemke 2012), Fenn and colleagues pointed out that we used a simplified measure of atmospheric deposition as one of our nine water-quality stressors. Our atmospheric stressor ignored dry nitrogen (N) deposition and the contribution of ammonium to total N. Furthermore, we used a simplified approach for combining nitrate and sulfate to capture acidic deposition.

As we stated in our article, we avoided the complications of computing more refined measures of our stressors because prior research had shown that

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the additional precision afforded by using more refined measures added little to the resultant multivariate measure of risk—principally because of the generally high correlation between a simplified measure and its related more refined measure. An additional reason for not including dry deposition was that the more comprehensive and spatially detailed estimates, from the Community Multiscale Air Quality model (CMAQ; *www.cmaq-model.org*), appeared to rely on a greater degree of modeling than we were comfortable with and, furthermore, were available for only one year: 2002 (see Baron et al. 2011).

Nevertheless, it is useful to determine whether including dry deposition and wet ammonium deposition in the measures of atmospheric N deposition and refining our measure of acidic inputs would significantly alter our estimates of the relative risk of water-quality impairment. In addition, it would be useful to see the effect of using PRISM (Parameter-elevation Regressions on Independent Slopes Model) precipitation data to distribute deposition estimates across the landscape, which would allow for greater spatial precision than what is available with the National Atmospheric Deposition Program (NADP) interpolation procedure that we relied on (Baron et al. 2011, Latysh and Wetherbee 2012).

In our article, we used subsets of nine stressors to estimate a relative risk value for each of three water-quality problems (sediment, nutrients, and toxics) for each of the 15,272 fifth-level watersheds covering the coterminous United States. The risk values for the three problems were then combined to yield a single risk value for each watershed. Wet nitrate (NO_3^-) deposition was one of the stressors affecting the nutrient problem, and the sum of wet nitrate plus wet sulfate (SO_4^{2-}) deposition was one of the stressors affecting the toxics problem. The data for these measures were taken from the NADP data set for the years 2000–2006. New measures we explore here are, for the nutrient problem, nitrate plus ammonium (NH_4^+) wet deposition (both incorporating PRISM precipitation data) plus dry deposition, all expressed in kilograms (kg) of N per hectare (ha) and, for the toxics problem, nitrate plus sulfate deposition expressed in equivalents per ha. The ammonium data were taken from the NADP Web site for the years 2000–2006, and the dry deposition data were taken from the CMAQ site mentioned above for 2002.

Adding dry N to wet N deposition has a substantial effect, increasing the median total N deposition across the watersheds from 3.5 kg per ha for wet only to 7.7 kg per ha for wet plus dry deposition. Across the full set of watersheds, the correlation of the new N measure with our original measure is $r = .86$. The correlation of acidic inputs in terms of equivalents with our original measure is $r = .98$. Finally, the correlation of the original scale values of risk with those computed when the new atmospheric deposition measures are incorporated is $r = .996$. The effect on the risk values of the change to the new deposition measures is very small, largely because in this framework, dry deposition affects only one of five stressors of one of three problems. Details on the new measures are available from the authors.

Therefore, we agree that our original measures of atmospheric deposition lacked completeness and precision, but we confirm that moving to more complete and refined deposition measures has a negligible impact on our multivariate, multiproblem characterization of the risk of water-quality impairment.

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THOMAS C. BROWN AND
PAMELA FROEMKE
Thomas C. Brown (thomas.brown@colostate.edu) and Pamela Froemke (pfroemke@fs.fed.us) are with the US Forest Service's Rocky Mountain Research Station, in Fort Collins, Colorado.

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