# *Additional Preliminary Comments from Dr. Charles Driscoll on EPA's Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter – Ecological Criteria (First External Review Draft)*

Chapter comments on $NO_x$-$SO_2$ secondary standard ISA

Charles Driscoll

Overall Comments. I am generally supportive of the $NO_x$-$SO_2$ secondary standard ISA. It is a very long and comprehensive document. The writers of this document synthesize previous analysis from the 2008 ISA and present additional literature since that time. Overwhelmingly the most recent literature is consistent with the 2008 ISA, and affirms the conclusions of that document. In some cases, additional important detail is now available from this recent literature. I have reviewed chapters 1, 2, 3, 4, 5, 7, 8, 10, 12,13 and 14. The ISA is well written and well organized. I have no major concerns with the ISA. I have some general suggestions and comments below for Chapters 4, 12 and 13. In an accompanying spreadsheet I have provided detailed comments and suggestions for the chapters I have read. In many cases this is calling attention to additional recent literature that may be relevant to what is presented.

*Chapter 4. Please comment on the accuracy, clarity, level of detail and relevance of the discussion regarding indicators, processes, models, monitoring and characterization of national scale sensitivity.*

I am supportive of Chapter 4 which summarizes soil biochemical response to atmospheric sulfur and nitrogen deposition. The chapter is complete I see inadequacies with the presentation on indicators, processes, models, monitoring or national sensitivity. It might be useful to contrast the sensitivity to atmospheric sulfur and nitrogen deposition across different regions. Also it would be useful to comment if there are differential effects of oxidized vs. reduced nitrogen. A few suggestions are provided on the detailed comments.

*Chapter 12. Please comment on the accuracy, clarity, level of detail and relevance of information presented on the biological effects of nutrient-enhanced sulfide phytotoxicity and nutrient enhanced mercury methylation.*

I am generally supportive of this chapter. Chapter 12 adequately covers the accuracy, clarity, level of detail and relevance of information presented on the biological effects of nutrient-enhanced sulfide phytotoxicity and nutrient enhanced mercury methylation. I have a few comments on the text and suggest consider additional references. The major concern I have with the chapter is with the apparent inconsistency on the role of sulfate inputs in driving changes in methyl mercury production and bioaccumulation. The chapter could be greatly improved and clarified with some additional text at the start of the methylmercury section. There are four environmental factors that potentially limit the production of methylmercury: inorganic mercury supply, oxygen or redox status, labile organic carbon and sulfate. Any one of these factors can limit methylmercury production. One idea that does not come across in the text is that at low sulfate concentrations, such as those in forested areas in the upper Midwest, methylation may be sulfate limited and therefore readily responsive to inputs of sulfate. In regions with higher inputs of sulfate, maybe methylation is not sulfate limited but limited by other factors. The conceptual model proposed by Gilmour (2011) (below) might be useful to the general reader to understand this concept. Methylation is likely stimulated by sulfate additions at low sulfate concentrations, but can be limited at high sulfate inputs due to high production of sulfide. This "Goldilocks" zone can be altered by different environmental factors.

*Chapter 13. Please comment on the accuracy, clarity, level of detail and relevance of information presented on modification of ecosystem response due to changes in temperature and precipitation.*

It is a tremendous challenge to try to link climate change effects with linkages to effects on atmospheric sulfur and nitrogen deposition. This short chapter is an attempt to do this. I can imagine there are many important dimensions of climate change and atmospheric deposition that interconnect. Climate is fundamental to biogeochemical processes and phenomena, so it is not surprising that changes in temperature and precipitation will alter ecosystem response to atmospheric deposition of nitrogen and sulfur. That said I don't think this chapter should be long, particularly given the length of the document. I have a couple of suggestions. First the chapter overwhelmingly focuses on nitrogen. I think the chapter could benefit from a few more examples of how climate change interacts with sulfur effects. Rice et al. (2014) conducted a nice analysis of latitude variation in sulfate recovery from acid deposition. This study is discussed in chapter 4. Rice et al. (2014) indicate in their paper that hydrology and specifically runoff is an important controller of sulfate recovery in watershed because drainage flushes the accumulate sulfur from soil. If precipitation and runoff patterns change under a future climate, this important process will be affected. Along a similar line, Mitchell and Likens (2011) observed that following decades of changes in stream sulfate concentrations and fluxes that have been driven by atmospheric deposition, variation is stream sulfate in now being controlled by variations in precipitation inputs. Increased variation in precipitation and water stage will increase wetting and drying cycles that promote mineralization of sulfate from soil and subsequent methylation.

A second point this might be considered is the recent hypothesis that forested watersheds are undergoing an oligiotrophication phenomenon that is driven by climate change (Duran et al. 2016). Also Sabo et al. (2016) recently report long-term declines in δ15N in tree rings in the Adirondacks, which may be an indicator of a long-term oligiotrophication.

References

Durán, J., J. L. Morse, P. M. Groffman, J. L. Campbell, L. M. Christenson, C. T. Driscoll, T. J. Fahey, M. C. Fisk, G. E. Likens, J. M. Melillo, M. J. Mitchell, P. H. Templer and M. A. Vadeboncoeur.

2016. Climate change decreases nitrogen pools and mineralization rates in northern hardwood forests. Ecosphere 73.doi: e01251.10.1002/ecs2.1251.

Gilmour, C.C. (2011) A Review of the Literature on the Impact of Sulfate on Methylmercury in Sediments and Soils. Report for the South Florida Water Management District. 67 p.

Sabo, R. D., S. E. Scanga, G. B. Lawrence, D. M. Nelson, K. N. Eshleman, G. A. Zabala, A. A. Alinea and C. D. Schirmer. 2016. Watershed-scale changes in terrestrial nitrogen cycling during a period of decreasing atmospheric nitrate and sulfur deposition. Atmospheric Environment. DOI: 10.1016/j.atmosenv.2016.08.055.

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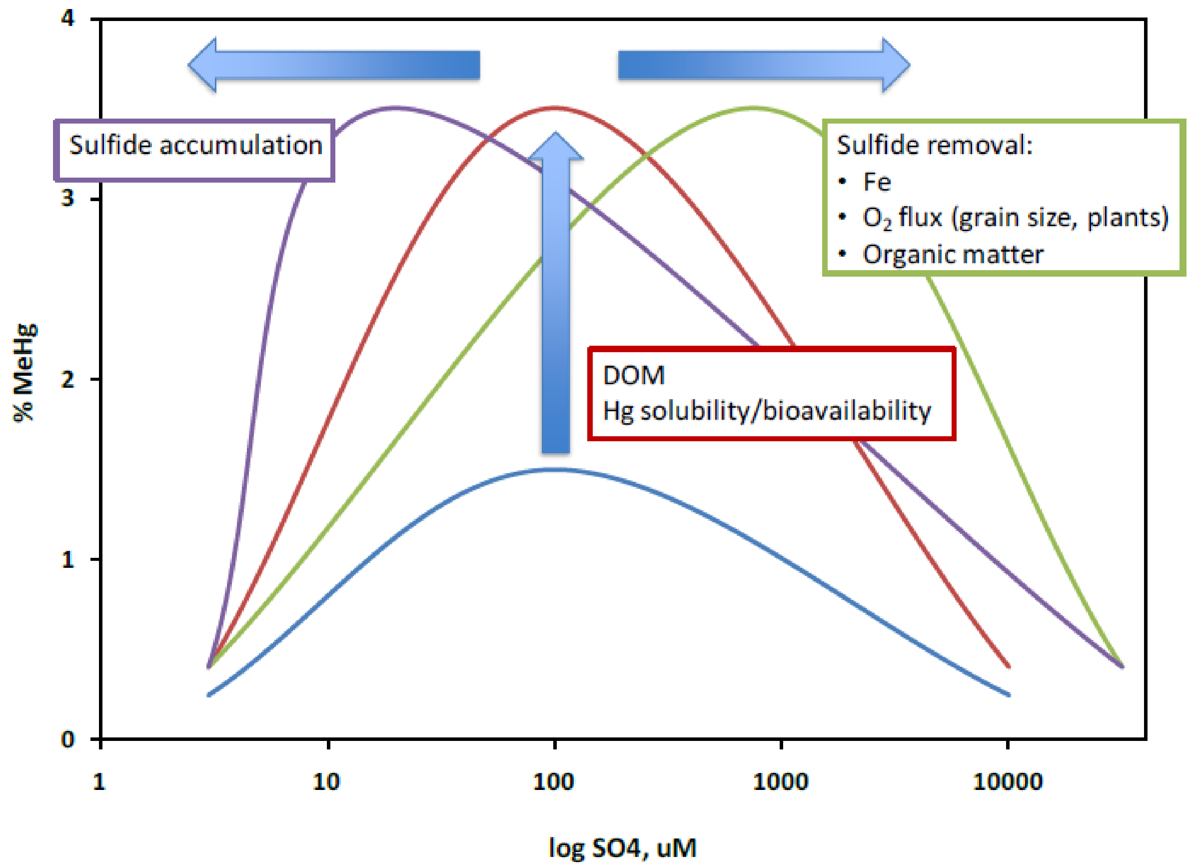


Detailed Comments

| page | line/location | comment |
|---|---|---|
| lxvi | Table ES-1 | typo second line in first category "alteration" |
| lxxiii | 28 | here and throughout the document inorganic Al is probably not the best term, because soil/sediment Al is largely in an inorganic form and is non-toxic. Probably should use dissolved inorganic Al |

| page | line/location | comment |
|---|---|---|
| 1-47 | 15 | Need to clarify the uniits of Bc:Al. eq/mol? |
| 1-56 | 5 | Should be PnET-BGC |
| 1-57 | 10 | Statement is not correct. Some models, such as PnET-BGC predict all acid base species, including pH, species of Al, in addition to ANC |
| 1-57 | 11 | Should provide some additional clarification to statement. ANC is a human chemical contruct to enhance understanding of acid-base processes and does not have any direct relevance to biological impacts. |
| 1-62 | 14 | Seems to be a mistake. "maintain an ANC of 74 eq/ha-yr…" 74 eq/ha-yr is a loading not an ANC value. Clarify. |
| 1-73 | 3 | Susceptible to eutrophication or experiencing eutrophication? |
| 1-73 | 8 | Water quality has diminished or deteriorated, rather than decreased |
| 1-74 | | Would it make more sense to place the order of the wetland section before the marine section to follow the flow from the atmoshere to uplands to surfacewaters to wetlands to marine ecosystems? |
| 2-1 | | Give an example of a species represented in NOz |
| 2-1 | 29 | Is the first phrase in the sentence begining "Excess NH3" linked to the phrase that follows? By nitrification is the author referring to oxidation of NH3? Nitrification of or nitrification in? A strangely phrased sentence. |
| 2-4 | Table 2-1 | These are U.S. emissions correct? This should be clarified in the table title. |
| 2-16 | 13 | transported from , correct? |
| 2-66 | | Would it be possible to show a map of % dry nitrogen deposition for the CONUS similar to 2-26. Even better if this cound be shown for both oxidized and reduced nitrogen. |
| 2-66 | 26 | parks (lower case) |
| 2-77 | 6 | Most surface waters are net sources of atmospheric carbon dioxide, so I don't think this statement is correct or relevant |
| 2-82 | Figure 2-34 | In Figure 2-34 it would be helpful to clarify what the data points for transference ratios represent |
| 2-18 | | Background values for ambient air concentrations are not that helpful for an effects document. It would be better to provide specific concentrations in precipitation and total deposition of backgound values to place current deposition values in perspective. The % contribution as background deposition is also not particularly useful and this % changes with the period of observation. Absolute background deposition values would be most helpfu. |
| 4-19 | Sulfate | I am concerned that this section does not address the pH-dependent adsorption of sulfate. In sites where the pH is changing this can influence the retention/ release of soil sulfate. See Fakhraei, H., C. T. Driscoll, J. R. Renfro, M. A. Kulp, T. Blett, E. F. Brewer, and J. Schwartz. 2016. Critical Loads and Exceedances for Nitrogen and Sulfur Atmospheric Deposition in Great Smoky Mountains National Park, USA. Ecosphere, 7(10). doi:e01466. 10.1002/ecs2.1466. |
| 4-4 | 17 | The most important process of soil acidification is the leaching of cations with drainage waters. The quantity of precipitation and runoff is an important determinant. van Breemen, N., J. Mulder, and C. T. Driscoll. 1983. Acidification and alkalization of soils. Plant and Soil 75:283-308. |

| page | line/location | comment |
|---|---|---|
| 4-22 | Base cations | I am not sure what science should be included in a section on base cations. There is no mention of the calcium silicate experiment conducted at Hubbard Brook. There has been quite a bit of relevant literature from this experiment. Shao, S., C. T. Driscoll, C. E. Johnson, T. J. Fahey, J. J. Battles, and J. D. Blum. 2016. Long-term responses in soil solution and stream-water chemistry at Hubbard Brook after experimental addition of wollastonite. Environmental Chemistry, 13(3), 528-540. doi:10.1071/EN15113.; Battles, J. J., T. J. Fahey, C. T. Driscoll, J. D. Blum, and C. E. Johnson. 2014. Restoring soil calcium reverses forest decline. Environmental Science & Technology Letters 1:15-19.; Johnson, C. E., C. T. Driscoll, J. D. Blum, T. J. Fahey, and J. J. Battles. 2014. Soil chemical dynamics after calcium silicate addition to a northern hardwood forest. Soil Science Society of America Journal 78:1458-1468.; Green, M. B., A. S. Bailey, S. W. Bailey, J. J. Battles, J. L. Campbell, C. T. Driscoll, C. Eagar, L. Lepine, G. E. Likens, S. V. Ollinger, and P. G. Schaberg. 2013. Decreased water flowing from a forest amended with calcium silicate. Proceedings of the National Academy of Sciences 110:5999-6003.; Nezat, C. A., J. D. Blum, and C. T. Driscoll. 2010. Patterns of Ca/Sr and 87Sr/86Sr variation before and after a whole watershed CaSiO3 addition at the Hubbard Brook Experimental Forest, USA. Geochimica et Cosmochimica Acta 74:3129-3142 DOI: 3110.1016/j.gca.2010.3103.3013. |
| 4-26 | Aluminum | I don't know how the ISA can say there have been no papers on aluminum leaching. Above I provide studies from the wollastonite addition experiment. Also see Fakhraei, H., and C. T. Driscoll. 2015. Proton and aluminum binding properties of organic acids in surface waters of the Northeastern, USA. Environmental Science & Technology 49:2939-2947.; Fuss, C. B., C. T. Driscoll, and J. L. Campbell. 2015. Recovery from chronic and snowmelt acidification: Long-term trends in stream and soil water chemistry at the Hubbard Brook Experimental Forest, New Hampshire, USA. Journal of Geophysical Research Biogeosciences 120:2360-2374.; Driscoll, C. T., K. M. Driscoll, H. Fakhraei, and K. Civerolo. 2016. Long-term temporal trends and spatial patterns in the acid-base chemistry of lakes in the Adirondack region of New York in response to decreases in acidic deposition. Atmospheric Environment, 146, 5-14. doi:10.1016/j.atmosenv.2016.08.034. |
| 4-48 | 1-2 | Do we know that decreases in pH supress DOC production? It is difficult to separate this effect from abiotic effects. There should be some text added on changes in partitioning of DOC with changes in soil pH which could drive changes in DOC mobility. |
| "4-48 | Dissolved organic carbon | It would be good to add in some text on the acid base chamistry of organic acids and potential changes in dissolved organic matter quality. Fakhraei, H., and C. T. Driscoll. 2015. Proton and aluminum binding properties of organic acids in surface waters of the Northeastern, USA. Environmental Science & Technology 49:2939-2947. |
| 4-55 | Soil monitoring databases | I mentioned the long-term wollastonite addition study at Hubbard Brook above. There is also a long-term soil solution data base; see Fuss, C. B., C. T. Driscoll, and J. L. Campbell. 2015. Recovery from chronic and snowmelt acidification: Long-term trends in stream and soil water chemistry at the Hubbard Brook |

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| | | Experimental Forest, New Hampshire, USA. Journal of Geophysical Research Biogeosciences 120:2360-2374. |
| 4-56 | 4 | I do not understand the sentence on the Lehman et al. 2008 study. This should be rewritten so it is clearer. |
| 4-64 | Models | I don't understand how you could say there have been no applications on PnET-BGC since 2008. See the following references. Fakhraei, H., C. T. Driscoll, J. R. Renfro, M. A. Kulp, T. Blett, E. F. Brewer, and J. Schwartz. 2016. Critical Loads and Exceedances for Nitrogen and Sulfur Atmospheric Deposition in Great Smoky Mountains National Park, USA. Ecosphere, 7(10). doi:e01466. 10.1002/ecs2.1466.; Pourmokhtarian, A., C. T. Driscoll, J. Campbell, K. Hayhoe, A. M. K.Stoner, M. B. Adams, D. Burns, I. Fernandez, M. J. Mitchell and J. B. Shanley. 2016. Modeled Ecohydrological responses to climate change at seven small watersheds in the northeastern U.S. Global Change Biology 23: 840-856. doi:10.1111/gcb.13444.; Zhou, Q., C. T. Driscoll, and T. J. Sullivan. 2015. Responses of 20 lake-watersheds in the Adirondack region of New York to historical and potential future acidic deposition. Science of the Total Environment 511:186-194.; Zhou, Q., C. T. Driscoll, T. J. Sullivan, and A. Pourmokhtarian. 2015. Factors influencing critical loads and target loads for the acidification of lake-watersheds in the Adirondack region of New York. Biogeochemistry 124:353-369.; Zhou, Q., C. T. Driscoll, S. E. Moore, M. A. Kulp, J. R. Renfro, J. S. Schwartz, M. J. Cai, and J. A. Lynch. 2015. Developing critical loads of nitrate and sulfate in the Great Smoky Mountains National Park, United States. Water Air and Soil Pollution 226:1-16.; Fakhraei, H., C. T. Driscoll, P. Selvendiran, J. V. DePinto, J. Bloomfield, S. Quinn, and C. Rowell. 2014. Development of a total maximum daily load (TMDL) for acid-impaired lakes in the Adirondack region of New York. Atmospheric Environment 95:277-287.; Bytnerowicz, A., M. Fenn, S. McNulty, F. Yuan, A. Pourmokhtarian, C. T. Driscoll, and T. Meixner. 2013. Interactive effects of air pollution and climate change on forest ecosystems in the United States: Current understanding and future scenarios. Pages 333-369 in R. Matyssek, N. Clarke, P. Cudlin, T. N. Mikkelsen, J.-P. Tuovinen, G. Wieser, and E. Paoletti, editors. Developments in Environmental Science. Climate Change, Air Pollution and Global Challenges. Elsevier Physical Sciences Series.; Pourmokhtarian, A., C. T. Driscoll, J. L. Campbell, and K. Hayhoe. 2012. Modeling potential hydrochemical responses to climate change and rising CO2 at the Hubbard Brook Experimental Forest using a dynamic biogeochemical model (PnET-BGC). Water Resources Research 48, W07514:13pp.; Wu, W., and C. T. Driscoll. 2010. Impact of climate change on three-dimensional dynamic critical load functions. Environmental Science & Technology 44:720-726.; Fenn, M. E., C. T. Driscoll, Q. Zhou, L. E. Rao, T. Meixner, E. B. Allen, F. Yuan, and T. J. Sullivan. 2015. Use of combined biogeochemical model approaches and empirical data to assess critical loads of nitrogen. Chapter 10. In W. De Vries, J.-P. Hettelingh, and M. Posch, editors. Critical Loads and Dynamic Risk Assessments: Nitrogen, Acidity and Metals for Terrestrial and Aquatic Ecosystems. Springer, Dordrecht, The Netherlands. |
| 4-65 | 16 | The sentence starting with Zaehle (2013) is not clear and should be rewritten. |

| page | line/location | comment |
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| 5-10 | Table 5-2 | In Battles et al. 2014 sugar maple response to changes in Ca/Ali in soil solutions are reported. |
| 5-13 | 1 | Is there any evidence for episodic acidification in soil? |
| 5-26 | 17 | Clarify the species abundance increase with increasing pH. |
| 5-30 | 11 | Are the units here inappropriate for pH? Should be unitless, correct? |
| 5-32 | 17 | driven? |
| 5-32 | 18 | Is this statement that the highest atmospheric fate factor occur on the west coasts globally relevant or only for the temperate zone in the Northern Hemisphere? |
| 5-34 | Acidification models | There must be a bias against PnET-BGC. As mentioned in chapter 4 PnET-BGC has been used to evaluate acid rain effects on soil acidification. |
| 5-43 | Impacts of ambient deposition | It would be helpful to indicate the year/ period over which these assessments were made, as deposition has changed markedly in recent years. |
| | | There is a new paper on the recent response of red spruce to decreases in acid deposition that may be of interest. Wason, J. W., Dovciak, M., Beier, C. M. and Battles, J. J. (2017), Tree growth is more sensitive than species distributions to recent changes in climate and acidic deposition in the northeastern United States. J Appl Ecol. doi:10.1111/1365-2664.12899 |
| 7-5 | Table 7-1 | There have been several other studies documenting nitrogen deposition to watersheds and surface waters before 2008. For example Castro, M. S., C. T. Driscoll, T. E. Jordan, W. G. Reay, and W. R. Boynton. 2003. Sources of nitrogen to estuaries in the United States. Estuaries 26:803-814. ; Driscoll, C. T., D. Whitall, J. Aber, E. Boyer, M. Castro, C. Cronan, C. L. Goodale, P. Groffman, C. Hopkinson, K. Lambert, G. Lawrence, and S. Ollinger. 2003. Nitrogen pollution in the northeastern United States: Sources, effects, and management options. BioScience 53:357-374.; Castro, M. S., and C. T. Driscoll. 2002. Atmospheric nitrogen deposition to estuaries in the mid-Atlantic and Northeastern United States. Environmental Science & Technology 36:8. |
| 7-8 | 8 | Sulfate is not highly mobile in unglaciated watersheds with soils elevated in clay and amorphous iron and aliminum. |
| 7-12 | 13 | Is part of the sentence missing here? "Residence times were relatively long in fine detritus, insects and particulate N." |
| 7-18 | 3 | Nitrogen can also be released back to the atmosphere as N2. |
| 7-18 | 6 | I don't think the sentence about reposition is correct, at least over relatively short time frames. Denitrification produces N2O and N2, both of which have long atmospheric desidence times. |
| 7-18 | 22 | 2009 is not a new study. |
| 7-23 | 32 | There have been some recent studies that could be used to update the research on watershed 1 at Hubbard Brook. Johnson et al. 2014 characterized the response of exchangeable calcium and aluminum to the wollastoite treatment. Johnson, C. E., C. T. Driscoll, J. D. Blum, T. J. Fahey, and J. J. Battles. 2014. Soil chemical dynamics after calcium silicate addition to a northern hardwood forest. Soil Science Society of America Journal 78:1458-1468. Shao et al. 2016 |

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| | | provided an updated response of soil and stream water to the treatment. Shao, S., C. T. Driscoll, C. E. Johnson, T. J. Fahey, J. J. Battles, and J. D. Blum. 2016. Long-term responses in soil solution and stream-water chemistry at Hubbard Brook after experimental addition of wollastonite. Environmental Chemistry, 13(3), 528-540. doi:10.1071/EN15113. |
| 7-24 | 11 | ANC is generally more stable than pH because it is insensitive to changes in CO2 |
| 7-25 | 14 | The most important process driving episodic acidification is the dilution of base cations. |
| 7-25 | 27 | Again dissolved inorganic aluminum or inorganic monomeric aluminum. |
| 7-25 | 30 | I would phrase this to say in the absense of inputs of strong acids (e.g., sulfuric acid, nitric acid) |
| 7-26 | Surface water DOC | Gerson et al. (2016) discuss the implications of enhanced mobilization of DOC for changes in the nutrient status of Adirondack lakes. Gerson, J. R., C. T. Driscoll, and K. M. Roy. 2016. Patterns of nutrient dynamics in Adirondack lakes recovering from acid deposition. Ecological Applications, 26(6), 1758-1770. doi:doi:10.1890/15-1361.1, Fakharaei and Driscoll (2015) characterized the acid base and aluminum binding properties of dissolved organic matter. They also suggest that the quality of dissolved organic matter is changing with the enhanced mobilization. Fakhraei, H., and C. T. Driscoll. 2015. Proton and aluminum binding properties of organic acids in surface waters of the Northeastern, USA. Environmental Science & Technology 49:2939-2947. |
| 7-27 | Eutrophication monitoring | Yanai et al. (2013) conducted a detailed time series analysis of nitrogen budgets for a forest reference watershed at Hubbard Brook, They found decreases in stream nitrate precede decreases in atmospheric deposition. Yanai, R. D., M. A. Vadeboncoeur, S. P. Hamburg, M. A. Arthur, C. Fuss, P. M. Groffman, T. G. Siccama, and C. T. Driscoll. 2013. From missing source to missing sink: Long-term changes in the nitrogen budget of a northern hardwood forest. Environmental Science & Technology 47:11440-11448. |
| 7-29 | Acidification monitoring | Driscoll et al. (2016) conducted detailed time-series analysis of changes in the acid-base status of Adirondack lakes. Driscoll, C. T., K. M. Driscoll, H. Fakhraei, and K. Civerolo. 2016. Long-term temporal trends and spatial patterns in the acid-base chemistry of lakes in the Adirondack region of New York in response to decreases in acidic deposition. Atmospheric Environment, 146, 5-14. doi:10.1016/j.atmosenv.2016.08.034.Fakhraei et al. (2016) conducted an analysis of trends in stream chemistry at the Great Smoky Mountain National Park. Fakhraei, H., C. T. Driscoll, J. R. Renfro, M. A. Kulp, T. Blett, E. F. Brewer, and J. Schwartz. 2016. Critical Loads and Exceedances for Nitrogen and Sulfur Atmospheric Deposition in Great Smoky Mountains National Park, USA. Ecosphere, 7(10). doi:e01466. 10.1002/ecs2.1466. |
| 7-35 | Northeastern US | Fuss et al. 2015 examined long-term trends in soil solutions and surface waters at the Hubbard Brook Experimental Forest NH. They found rates of ANC increase under year around conditions were similar to snowmelt, although the ANC during snowmelt was 10 µeq/L lower that yearly averages. |
| 7-35 | 35 | You probably only need to say acid-sensitive once in the sentence. |

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| 7-36 | 4 | Actually in some surface waters there have been marked decreases in dissolved inorganic aluminum see Driscoll, C. T., K. M. Driscoll, H. Fakhraei, and K. Civerolo. 2016. Long-term temporal trends and spatial patterns in the acid-base chemistry of lakes in the Adirondack region of New York in response to decreases in acidic deposition. Atmospheric Environment, 146, 5-14. doi:10.1016/j.atmosenv.2016.08.034. |
| 7-38 | Central and Southern Appalachans | Rice et al. (2014) examined the source-sink behavior of sulfate in forested watersheds across a latitudinal gradient under conditions of decreases in sulfate deposition. Rice, K. C., T. M. Scanlon, J. A. Lynch, and B. J. Cosby. 2014. Decreased atmospheric sulfur deposition across the southeastern US: when will watersheds release stored sulfate? Environmental science & technology 48:10071–10078. |
| 7-43 | Freshwater acidification models | Fakhraei et al. (2016) conducted hindcast and forecast simulations with PnET-BGC on streams in the Great Smoky Mountain National Park. Fakhraei, H., C. T. Driscoll, J. R. Renfro, M. A. Kulp, T. Blett, E. F. Brewer, and J. Schwartz. 2016. Critical Loads and Exceedances for Nitrogen and Sulfur Atmospheric Deposition in Great Smoky Mountains National Park, USA. Ecosphere, 7(10). doi:e01466. 10.1002/ecs2.1466. |
| 7-52 | 4, Table 7-6 | Aren't criteria for ammonia targeted to free ammonia? If so this should be clarified? I thought New York had criteria for nitrate and free ammonia. |
| 7-68 | Nutrient enhanced coastal acidification | Does phytoplankton uptake of ammonium or nitrate influence the acid-base chemistry of estuarine waters? |
| 7-73 | 10,11 | Is there a differentiation between alkalinity and ANC? If so what is it? |
| 8-39 | 19 | Another recent study showing lack of fish recovery despite improvements in water chemistry is Baldigo et al. 2016. Baldigo, B. P., K. M. Roy, and C. T. Driscoll. 2016. Response of fish assemblages to declining acidic deposition in Adirondack Mountain lakes 1984-2012. Atmospheric Environment, 146, 223-235. doi:10.1016/j.atmosenv.2016.06.049. |
| 8-60 | Target and dynamic critical loads | Other recent analyses of dynamic critical loads in the Adirondacks and the Great Smoky Mountains National Park include: Fakhraei, H., C. T. Driscoll, P. Selvendiran, J. V. DePinto, J. Bloomfield, S. Quinn, and C. Rowell. 2014. Development of a total maximum daily load (TMDL) for acid-impaired lakes in the Adirondack region of New York. Atmospheric Environment 95:277-287. and Fakhraei, H., C. T. Driscoll, J. R. Renfro, M. A. Kulp, T. Blett, E. F. Brewer, and J. Schwartz. 2016. Critical Loads and Exceedances for Nitrogen and Sulfur Atmospheric Deposition in Great Smoky Mountains National Park, USA. Ecosphere, 7(10). doi:e01466. 10.1002/ecs2.1466. |
| 12-4 | 9 | Sulfate in not particularly mobile in soils will a high clay content and high in amorphous iron and aluminum oxides. |
| 12-5 | 28 | Soil content of amorphous iron and aluminum oxides is also an important controller of sulfate adsorption |
| 12-6 | 12 | Flocculant layer? |

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| 12-6 | 28 | This statement is not correct. The quantity of sulfate retained in lake sediments is highly variable and depends on th mean depth and hydraulic residence time of the lake. Kelly, C. A., J. W. M. Rudd, R. H. Hesslein, D. W. Schindler, P. J. Dillon, C. T. Driscoll, S. A. Gherini, and R. E. Hecky. 1987. Prediction of biological acid neutralization in acid-sensitive lakes. Biogeochemistry 3:129-140. |
| 12-23 | Zones of high methylmercury fractions across the landscape | Yu et al. found no variation in concentrations of total mercury, methylmercury and %MeHg with pH in Adirondack lakes. Yu, X., C. T. Driscoll, M. Montesdeoca, D. Evers, M. Duron, K. Williams, N. Schoch, and N. C. Kamman. 2011. Spatial patterns of mercury in biota of Adirondack, New York lakes. Ecotoxicology 20 1543-1554 Selvendiran et al. 2008 found methylmercury and %MeHg greatly increase in waters draining wetlands compared with drainage from uplakes in the Adirondack landscape. Selvendiran, P., C. T. Driscoll, J. T. Bushey, and M. R. Montesdeoca. 2008. Wetland influence on mercury fate and transport in a temperate forested watershed. Environmental Pollution doi:10.1016/j.envpol.2007.12.005: 154:46-55. |
| 12-24 | 24 | base of the foodchain |
| 12-27 | 26,33 | Onondaga Lake |
| 12-27 | Seasonality and temperature | Selvendiran et al (2008) and Gerson and Driscoll (2016) observed marked increases in concentrations of methylmercury and %MeHg in Adirodack streams during the summer low flow period. Selvendiran, P., C. T. Driscoll, J. T. Bushey, and M. R. Montesdeoca. 2008. Wetland influence on mercury fate and transport in a temperate forested watershed. Environmental Pollution doi:10.1016/j.envpol.2007.12.005: 154:46-55. Gerson, J. R., C. T. Driscoll. 2016. Is mercury in remote forested watershed of the Adirondack Mountains responding to recent decreases in emissions? Environmental Science and Technology, 50, 10943-10950. doi:10.1021/acs.est.6b02127 |
| 12-28 | Total mercury concentrations | Gerson and Driscoll (2016) recently examined factors driving long-term decreases in total mercury, methyl mercury, but not %MeHg in an Adriondack stream and lake. They found that these changes were consistent with decreases in atmospheric mercury deposition, but methylmercury concentrations and %MeHg were not affected by long term decreases in sulfate or changes in hydrology. Gerson, J. R., C. T. Driscoll. 2016. Is mercury in remote forested watershed of the Adirondack Mountains responding to recent decreases in emissions? Environmental Science and Technology, 50, 10943-10950. doi:10.1021/acs.est.6b02127 |
| "12-29 | pH | Yu et al. found no variation in concentrations of total mercury, methylmercury and %MeHg with pH in Adirondack lakes, but bioaccumulation of MeHg at the base and through the food chani was enhanced with decreases in lake pH. Yu, X., C. T. Driscoll, M. Montesdeoca, D. Evers, M. Duron, K. Williams, N. Schoch, and N. C. Kamman. 2011. Spatial patterns of mercury |
| 12-31 | 5 | Dittman et al. 2010 also found increases in MeHg and total Hgwith increases in the hydrophobic and high molecular weight fraction of dissolved organic carbon. Dittman, J. A., J. B. Shanley, C. T. Driscoll, G. R. Aiken, A. T. Chalmers, J. E. Towse, and P. Selvendiran. 2010. Mercury dynamics in relation to dissolved organic carbon concentration and quality during high flow events in three |

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| | | northeastern U.S. streams. Water Resources Research 46, W07522, doi:10.1029/2009WR008351. |
| 12-32 | 14 | Iron oxides can form surface complexes with inorganic mercury. |
| 12-32 | Nitrate | Matthews et al. (2013) document the application of calcium nitrate as a whole - lake sediment treatment to limit the production of methyl mercury in a mercury contaminated lake. Matthews, D. A., D. B. Babcock, J. G. Nolan, A. R. Prestigiacomo, S. W. Effler, C. T. Driscoll, S. Todorova, and K. M. Kuhr. 2013. Whole-lake nitrate addition for control of methylmercury in mercury-contaminated Onondaga Lake, NY. Environmental Research 125:52-60. |
| 12-44 | Sulfur oxide effects on methylmercury | This recent article was mentioned above but it is also relevant here. Gerson and Driscoll (2016) recently examined factors driving long-term decreases in total mercury, methyl mercury, but not %MeHg in an Adriondack stream and lake. They found that these changes were consistent with decreases in atmospheric mercury deposition, but methylmercury concentrations and %MeHg were not affected by long term decreases in sulfate or changes in hydrology. Gerson, J. R., C. T. Driscoll. 2016. Is mercury in remote forested watershed of the Adirondack Mountains responding to recent decreases in emissions? Environmental Science and Technology, 50, 10943-10950. doi:10.1021/acs.est.6b02127 |
| 12-76 | Sensitive ecosystems | Evers et al. (2011) was not peer reviewed but includes relevant information on ecosystem mercury sensitivity. This analysis was done for the Great Lakes drainage area. The authors found that lands with forest cover had greater mercury concentrations in game fish than lands in agricultural cover. This was attributed to the enhanced deposition of mercury by forests, and abundance of wetlands and low productivity. Evers, D. C., J. G. Wiener, C. T. Driscoll, D. A. Gay, N. Basu, B. A. Monson, K. F. Lambert, H. A. Morrison, J. T. Morgan, K. A. Williams, and A. G. Soehl. 2011. Great Lakes Mercury Connections: The Extent and Effects of Mercury Pollution in the Great Lakes Region. Biodiversity Research Institute. Gorham, Maine. Report BRI 2011-18. 44 pages. |
| 12-77 | 7 and elsewhere throughout the chapter | The authors indicate that mercury associates with thiosulfate groups in organic molecules. Mercury also associated with reduced sulfur groups, sulfhydryl groups. |
| 13-1 | 15 | data are |
| 13-1 | 12 | Earth systems |
| 13-2 | Figure 13.1 | Should clarify the significance/meaning of "+" and "-". |
| 13-5 | 25 | Need to be clear by what is meant by units, molar or mass units presumably. |
| 13-9 | 18 | I am not clear on the statement that acidification driven changes in nitrogen occur at higher levels of nitrogen addition than for initial changes to the carbon cycle. Is there a reference for this? Is this statement true? |
| 13-10 | 31 | Recently, Warren et al. (2016) suggested that mobilization of dissolved organic matter may help mitigate the effects of temperature increases on brook trout survival. Warren, D. R., C. E. Kraft, D. C. Josephson, and C. T. Driscoll. 2016. Acid rain recovery may help to mitigate the impacts of climate change on thermally |

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| | | sensitive fish in lakes across eastern North America. Global Change Biology. doi:10.1111/gcb.13568. |
| 14-2 | 8,9 | Probably among would be better than between. |
| 14-4 | 5 | Rather than "leaked" how about transported? |
| 14-4 | 21 | How about this ? "; however it is difficult to quantify these impacts due to data limitations." |
| 14-8 | 32 | Should use among rather than between. Also this sentence is confusing and should be rewritten. |
| 14 | | Caputo et al. (2017) conducted a benefits analysis on how reductions in acid deposition would improve the recreational fishery in the Adirondacks.Caputo, J., C. Beier, H. Fakhraei and C. T. Driscoll. 2017. Impacts of acidification and potential recovery on the expected value of recreational fisheries in Adirondack lakes (USA). Environ Sci Technol. 51: 742-750.doi: 10.1021/acs.est.6b05274 |