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

Charge Question #7. Review of Chapter 7 – Scientific evidence on aquatic biogeochemical response in freshwater and estuarine systems to nitrogen and sulfur deposition. Lead discussants are: Drs. Robert Howarth, Elizabeth Boyer, Douglas Burns, and William McDowell.

Chapter 7 characterizes scientific evidence on aquatic biogeochemical response in freshwater and estuarine systems to nitrogen and sulfur deposition.

a. *Please comment on the accuracy, clarity, level of detail, and relevance of information on biogeochemical processes and chemical indicators presented in the chapter as well as the adequacy of the discussion of monitoring, models, and national-scale sensitivity.*

b. *Please provide suggestions that may further improve the utility of this chapter as the foundation for linking biogeochemical alterations associated with nitrogen and sulfur deposition to biological effects in aquatic systems.*

I find that this chapter does a good job of summarizing and updating information on the response of fresh waters and estuaries to atmospheric nitrogen and sulfur deposition. I agree with the statements that causal relationships between atmospheric deposition and alteration of biogeochemistry in fresh waters and estuaries are well supported. These statements were also included in the 2008 ISA, and additional research since that time has further confirmed these relationships. There is a more recent body of literature on the role of nitrogen in causing lowered pH values in estuaries through enhanced decomposition of organic matter. In systems where atmospheric nitrogen deposition is a significant component of the load delivered to the estuary, a causal link to atmospheric deposition is clear. I have a few comments to improve accuracy and clarity of the information presented in the chapter that are detailed below. I also suggested in a few places where references to more recent studies (2014 and later) would help reinforce some key conclusions.

- Page 7-4, line 19 – there is also DNRA, which should probably be mentioned at least briefly.
- Table 7-1 – Burns et al., 2009 paper shows 40-53% of direct NO3 deposition in suburban stream during storm runoff. Also Sebestyen et al., 2008, Water Resources Research, p 8 – 48% direct atmospheric NO3 at peakflow.
- Page 7-6, line 18 – has reduced N also increased in terms of absolute concentrations or loads in some areas (e.g. – Iowa). Seems worth mentioning beyond the relative comparison.
- Page 7-8, line 2 – an important point here is that atmospheric nitrate was about half of stream nitrate during storms (more than the forested watershed) because of rapid shunting

of runoff through storm sewers.

- Page 7-9, line 2 – not sure I agree that there have been no studies of S sources since the ISA 2008. See Kang et al., 2014, dx.doi.org/10.1021/es502563n | Environ. Sci. Technol. 2014, 48, 11259−11267.
- Section 7.2.3.2 – There are now at least few studies that show temporal decreases in surface water nitrate concentrations that are consistent with declines in N deposition. The recent work of Keith Eshleman in Maryland. A recent paper by Driscoll et al., 2016, Atmospheric Environment shows declines in nitrate concentrations in many Adirondack lakes. Also, Strock et al., 2014 for lakes in New England and New York show a recent accelerating decline in nitrate concentrations.
- Page 7-16, line 31 – Sebestyen et al. 2008, Water Resour. Res. is also an important paper for showing the dominant direct role of atmospheric nitrate in snowmelt runoff at Sleepers R. VT.
- Section 7.2.3.8 – the base cation surplus of Lawrence should be mentioned here because it was designed to address the deficiency of using ANC as an indicator when DOC concentrations are elevated.
- Page 7-33, line 19 – there are two more recent studies that have looked at alkalinity trends in larger rivers and found a role for atmospheric deposition as well as other factors, Stets et al., 2014, Science of the Total Environment 488–489 (2014) 280–289, and Kaushall et al., 2013, dx.doi.org/10.1021/es401046s | Environ. Sci. Technol. 2013, 47, 10302−10311. These studies are relevant in this section of text.
- Pages 7-34 to 7-35 – much of this material would fit better in the DOC section.
- Page 7-35, line 20 – there is a study by Mitchell et al., 2011, Biogeochemistry (2011) 103:181–207 that is relevant here because they look at S budgets across several watersheds in the NE US and SE Canada. This study should be cited here and perhaps in other sections of the text such as the sulfate section.
- Page 7-37, line 16 – a recent paper by Fuss et al., 2015, J. Geophys. Res. Biogeosci., 120, 2360–2374, doi:10.1002/2015JG003063. examined trends at HBEF.
- Section 7.2.4.4 – paper by Rice et al., 2014, ES&T, Environ. Sci. Technol. 2014, 48, 10071−10078, is important to discuss in this section.
- Section 7.2.4.6 – work by Tom Clair for Atlantic Canada, 2011**,** Can. J. Fish. Aquat. Sci. 68: 663–674 should be discussed in this section. Also, there have been a number of papers on Ca in Canadian lakes by Norman Yan and colleagues. They describe the low Ca concentrations that are now evident in these lakes despite some recovery trends in lake ANC. There seems to be a hysteresis in the recovery that has implications for lake biota.
- Section 7.2.4.7 – two important European trends papers may have been published later than this review was performed, but they give broad views of trends across Europe (Garmo et al., 2014, Water Air Soil Pollut (2014) 225:1880) and the UK (Monteith et al., 2014, Ecological Indicators 37 (2014) 287–303).
- Section 7.2.5 – the forSAFE model has been used frequently for acid rain and critical loads studies, especially in Europe and is deserving of mention here. See: Zanchi et al., 2016, *Hydrology* **2016**, *3*, 11; doi:10.3390/hydrology3010011, and several references therein.

- Page 7-68, line 7 – Zhou et al., 2014, Limnol. Oceanogr., 59(2), 2014, 373–384, add further insights regarding the role of climate.
- Section 7.3.2.3 – would be helpful in this section to indicate briefly why estuarine/coastal acidification may be important from an ecosystem effects perspective since I don't believe this topic is discussed in Ch. 8.