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

Comments of Bob Howarth: I reviewed chapters 4, 7, and 10. Overall, I chapter 10 to be the best written, and I am only suggesting modest additions there. Chapter 4 started strong, but it is a long chapter, with a lot of redundancy, and the level of integration suffers towards the end. Chapter 7 is the weakest of the three, by far, and I believe needs some substantial revision and updating.

Chapter 4:

Page 4.2, lines 12-20: you could add that Hong et al. (2011) and (2013) show N deposition and total nitrogen inputs for all counties across the US, and provide an on-line link for individuals to evaluate how this has changed over time. The N deposition is based on the CMAQ model.

Hong, B., D. Swaney, and R. W. Howarth. 2011. A toolbox for calculating net anthropogenic nitrogen inputs (NANI). *Environmental Modeling and Software* 26: 623–33.

Hong, B., D.P. Swaney, and R.W. Howarth. 2013. Estimating net anthropogenic nitrogen inputs (NANI) to US watersheds: comparison of methodologies. *Environmental Science & Technology* 47: 5199−5207

Page 4.5, table 4.1: I suggest adding increased nitrous oxide (N2O) fluxes, and tying this to global warming.

Page 4.11, lines 5-23, and table 4.3 on pages 4-13 to 4-19: Bettez et al. (2013) demonstrated 25% to 30% higher N deposition to forests 10 m away from roads on Cape Cod, compared to hundreds of meters away from the road; the higher deposition was associated with a 2- to 4-fold greater rate of nitrate leaching from the soils. 73% of the deposition was retained in the forest away from roads, compared to 58% retention at 10 from the road.

Bettez, N., R. Marino, R.W. Howarth, and E.A. Davidson. 2013. Roads as nitrogen deposition hot spots. *Biogeochemistry* 114: 149-163.

Page 4.26, lines 26-34: it should be noted that increased nitrification will result in more formation of nitrous oxide, and more release of this greenhouse gas to the atmosphere.

Page 4.27, lines 14-20: need to point out that denitrification is only occurring in flooded soils, which leads to anoxia, and the spatial variation is tied to variation in flooding.

Page 4.27, lines 23-25: I consider the conclusion from Bouwman et al. (2013) that there is more denitrification in terrestrial ecosystems than in groundwater or riparian systems to be rather speculative, and not based on actual data; I would delete this statement.

Note also that for the major watersheds of the northeastern US, van Breemen et al. (2002) concluded that more denitrification probably occurs in riparian wetlands and in first-order streams than in terrestrial ecosystems.

Van Breemen, N., E. W. Boyer, C. L. Goodale, N. A. Jaworski, K. Paustian, S. Seitzinger, K. Lajtha, B. Mayer, D. van Dam, R. W. Howarth, K. J. Nadelhoffer, M. Eve, and G. Billen. 2002. Where did all the nitrogen go? Fate of nitrogen inputs to large watersheds in the northeastern USA. *Biogeochemistry* 57&58: 267-293.

Page 4.29, lines 1-6, and page 4.30, lines 1-9: this material seems redundant with the discussion earlier in the chapter, and should be moved up and better integrated. It seems tacked on at the moment.

Section 4.3.7, which begins on page 4.35: this section on how N effects decomposition is fine and reasonably up to date on the details. But, what is the intended take-home message here? The forest is getting lost in the trees…..

Section 4.3.8, which begins on page 4.43: the take-home message from this section is clear: mineralization increases with increasing N deposition. However, this seems at odds with the more qualified section on decomposition (which says sometimes rates increase, sometimes they go down).

Page 4.44, lines 11-12: Casson et al. (2014a) is cited as saying that most of the N mineralized in their forest site becomes nitrate. I believe this is at odds with a rather large body of evidence that ammonium is the more general product in forests. Referring only to this one study or one site, without the broader context, is misleading.

Page 4.48, lines 5-9: Some awkward writing here. First the report states that the reasons for increases in DOC are unclear, and while there is some thought it could be from recovery from acidification, in may also be from land use changes or climate change (and the land use and climate change ideas need some referencing, if they are to remain here). So my conclusion is no one really know. And then the report goes on and talks about a "second mechanism" for the increased DOC, tying it to N deposition. This really is the 4th mechanism the report lays out, if land use and climate change have any validity at all.

Page 4.48, lines 9-35: this needs some tightening up. It reads like a "he said, he said" sort of thing. I suggest instead emphasizing the meta-analysis, which starts on line 36, and shortening the material on lines 9-35 to support the meta-analysis.

From here on, I only very quickly skimmed chapter 4, as I was running out of the time I could devote to this chapter. My overall take is that the chapter could be substantially tightened up and shortened to reduce redundancy and make the take-home messages more clear.

Chapter 7:

Page 7.1, lines 1-12: There is a mis-match here between freshwater and estuarine coverage. Specifically, indicators of eutrophication, monitoring, and water quality criteria are all discussed for freshwaters, but not for estuaries. This is very strange, since N pollution and eutrophication is a far larger problem in estuaries than in freshwaters.

Page 7.2, lines 26-29: It may be worth noting that N-limitation in freshwater systems is more prevalent in oligotrophic systems, such as the high-altitude lakes where N deposition is the major N input (P limitation is more prevalent in mesotrophic freshwater lakes).

Page 7.3, lines 27-34: It is OK to talk about total N inputs, as this section does, but it should also emphasize where N depositional inputs are greatest. I suggest looking at Hong et al. (2011) and (2013), who show N deposition and total nitrogen inputs for all counties across the US.

Hong, B., D. Swaney, and R. W. Howarth. 2011. A toolbox for calculating net anthropogenic nitrogen inputs (NANI). *Environmental Modeling and Software* 26: 623–33.

Hong, B., D.P. Swaney, and R.W. Howarth. 2013. Estimating net anthropogenic nitrogen inputs (NANI) to US watersheds: comparison of methodologies. *Environmental Science & Technology* 47: 5199–5207

Page 7.4, lines 14-21: In Howarth et al. (1996) and Howarth et al. (2012), we showed across a large range of watersheds that 75% of N inputs are retained in the watershed or denitrified, and 25% are exported to surface waters. This was true no matter what the dominant N input was, including N deposition.

Howarth, R.W., G. Billen, D. Swaney, A. Townsend, N. Jarworski, K. Lajtha, J. A. Downing, R. Elmgren, N. Caraco, T. Jordan, F. Berendse, J. Freney, V. Kueyarov, P. Murdoch, and Zhu Zhao-liang. 1996. Riverine inputs of nitrogen to the North Atlantic Ocean: Fluxes and human influences. *Biogeochemistry* 35: 75-139.

Howarth, R. W., D. Swaney, G. Billen, J. Garnier, B. Hong, C. Humborg, P. Johnes, C. Morth, and R. Marino. 2012. Nitrogen fluxes from large watershed to coastal ecosystems controlled by net anthropogenic nitrogen inputs and climate. *Frontiers in Ecology & Environment* 10: 37-43.

Page 7.6, lines 6-16: this section on apportionment of N loads to deposititon vs. other N sources should include the work of Hong et al. (2011) and Howarth et al. (2012), which make this apportionment for all counties in the US.

Also, please note that the SPARROW model results presented here used wet deposition only as their measure of N deposition, thereby underestimating total deposition, particularly near urban areas where dry deposition of gases from combustion can be higher, according to the CMAQ model and several studies reviewed in Howarth (2008a) and Howarth (2008b). As discussed in those two reviews, the SPARROW model attributed a large amount of nitrogen to non-point source urban influences; this is most likely the dry deposition of exhaust N gases, and often doubles the importance of N deposition as an N source to higher-order river systems.

Howarth, R. W. 2008a. Coastal nitrogen pollution: A review of sources and trends globally and regionally. *Harmful Algae* 8: 14-20.

Howarth, R. W. 2008b. How important is atmospheric deposition as a source of nitrogen to coastal marine ecosystems in the northeastern United States? Pages 47-65 in A. Desbonnet and B. A Cost-Pierce (eds.), *Science of Ecosystem-Based Management.* Springer, NY.

Page 7.6, lines 17-21: This section on trends in N deposition seems rather outdated, relying heavily on references from 2008. Also, it should be noted that the spatial pattern of N deposition across the US has changed dramatically over the past few decades, as NOx emissions have gone down in urban areas and along highways, and ammonia emissions from ag areas have increased.

Page 7.6, lines 21-28: this section on N retention refers largely to literature on forests. Much of the N deposition on watershed falls on urban/suburban lands or ag lands, where retention is less.

Page 7.13, lines 1-8: this paragraph is rather qualitative, and could be strengthened by the observation from Howarth et al. (1996, 2012) that on average for large watershed with mixed land uses, 75% of N inputs are retained in the watershed and 25% exported downstream.

Page 7.14, lines 1-33: these two paragraphs give an awful lot of detail to just two rather micro-oriented studies, while also ignoring a large number of other studies. I suggest greatly shortening these, and brining in other new papers that use similar approaches.

Page 7.14, line 34, through page 7.15, line 10: this discussion on effects seems way out of place here, and also too limited in the examples shown.

Page 7.17, lines 1-16: The Goodale et al. (2009) study did indeed find high N retention, but measured in small forested catchments. This should be contrasted with the work of Boyer et al. (2002) and Howarth et al. (2006) which found far lower retention at larger spatial scales, including the Susquehanna, that included mixed land uses.

Boyer, E. W., C. L. Goodale, N. A. Jaworski, and R.W. Howarth. 2002. Anthropogenic nitrogen sources and relationships to riverine nitrogen export in the northeastern USA. *Biogeochemistry* 57&58: 137-169.

Howarth, R. W., E. W. Boyer, R. Marino, D. Swaney, N. Jaworski, & C. Goodale. 2006. The influence of climate on average nitrogen export from large watersheds in the northeastern United States. *Biogeochemistry* 79: 163-186.

Section 7.2.3.3, page 7.17, lines 17-22: this very, very short section of 5 lines should be deleted. Note that in the surface waters of the US, DIN is overwhelmingly dominated by nitrate (which should be explicitly stated). So the first part of this section should simply be a sentence in the previous section. The last sentence is out of place here in any case, and I believe should be deleted.

Page 7.18, lines 14-42: This long discussion is focused on one paper, and does not do an adequate job of describing the larger, related research. A few key points: The IPCC study referred to here is the 4th assessment from 2008; the 5th assessment from 2013 gives a far higher value, based on many published papers, but notably that of Crutzen et al. (from 2007 or 2008, I think). Also, N2O is produced during both denitrification and nitrification, and the study referenced here failed to point that out. And third, it is not true that this study found higher denitrification rates than say we would have predicted from the prior work of Seitzinger, or from the Howarth et al. (1996) synthesis.

Page 7.18, lines 23-30: the Mulholland et al. (2009) paper is an important one, but this summary should be better integrated with prior and subsequent research (Seitzinger, Alexander, Howarth, etc.).

Page 7.18, lines 31-33: the Bellinger et al. study may be new, but it's conclusions are not. Again, the report is doing a poor job of here of setting things up in the broader context. Kelly et al. made this same point in a comparative paper on N retention in lakes, which was published back in the 1980s I believe.

Page 7.18, lines 33-36: the details of this study on the relationship of nitrification to denitrification are simply not important here, and there are dozens of other papers on this topic, if we really want to get into it.

Page 7.18, line 36, through page 7.19, line 23: Here two the report relies heavily on just two studies, and by the same group. There is a much larger, and older body of literature here. The present coverage is misleading in that regard.

Page 7.19, lines 28-33: the report needs to specify whether mass unit ratios or molar unit ratios are being used, and someone should go back through the original papers and make sure what is being reported here is accurate in terms of mass or molar, since both are commonly used in the literature.

Pge 7.19, lines 31-33: Although I have worked on N vs P limitation of net primary production in aquatic ecosystems for over 35 years, I have no idea what this sentence means…… and I know that at least one of the papers referenced (Schindler 1980) does not support limitation by anything other than P alone. This part of the report needs to be much, much clearer.

Page 7.20, lines 1-2: delete "N/L"

Page 7.20, lines 8-17: delete this paragraph, which states some highly controversial ideas, and does so with no referencing. I for one do not agree with it as written.

Page 7.20, lines 18-26: one take a whole paragraph to discuss this one, single paper? I do not see the importance of this study, and suggest it be deleted.

Page 7.20, lines 27-37: I like this paragraph, and fully agree with it. However, here still the report is relying on just one paper. This general point of N limitation in oligotrophic settings and P limitation in more productive lakes has been made by others.

Page 7.21, ines 14-28: There is a fundamental confusion here. The Crowley et al paper dealt with both forests and lakes, and the summary in this report has confused the two: the reported failure of N deposition to drive limitation towards P was for forests, not lakes. The Crowley et al. paper states that lakes were always P limited, regardless of N deposition.

DUE TO TIME CONSTRAINTS, I ONLY QUICKLY SKIMMED PAGES 7.22 TO 7.41.

I was surprised by the large coverage of acid monitoring, relative to very sparse coverage of either nitrogen or eutrophication. Given the national and regional networks for monitoring of surface nutrients and eutrophication, I think is imbalance should be addressed.

Page 7.42, lines 14-28: the Han and Allen (2008) comparison is important, but it is not the only such comparison. The report should, at a minimum, also look at Alexander et al. (2002), Biogeochemistry, 57/58: 295-339.

Page 7.42, lines 24-25: Using a larger data set, Howarth et al. (2012) said 25% is exported.

Page 7.42, lines 29, to pge 7.43, line 4: why pick these particular 3 European studies? On the smaller scale work reported here, there are many, many other recent studies, particularly in Europe. I suggest taking a look at the European Nitrogen Assessment as a start, including the chapter by Billen et al. (2011).

Billen, G., M. Silvestre, B. Grizzetti, A. Leip, J. Garnier, M. Voss, R. Howarth, F. Bouraoui, H. Behrendt, A. Lepisto, P. Kortelainen, P. Johnes, C. Curtis, C. Humborg, E. Smedberg, O. Kaste, R. Ganeshram, A. Beusen, & C. Lancelot. 2011. Nitrogen flows from European regional watersheds to coastal marine waters. Pages 271 -297 in M.A. Sutton et al. (editors), *The European Nitrogen Assessment: Sources, Effects, and Policy Perspectives*. Cambridge University Press.

Page 7.43, lines 5-13: It is fine to use the Haag et al. (2012) study on the Baltic Sea, but please note that was based on earlier work that included watersheds from the US (Howarth et al. 1996, 2006; Boyer et al. 2002), and there are also more recent papers that include the US (Howarth et al. 2012; Hong et al. 2012). All of these papers make the same point, so it is unfortunate to make it seem like only the Swedish study exists.

Page 7.43, lines 17-23: why focus on this single study as the modeling paper on N cycle within lakes? There must be other examples. I am pretty sure I've seen work on how high N loads favor production of volatile organic toxins from cyanobacteria in lakes, for example, which would seem important to mention here.

PAGES 7.44 THROUGH 7.50: Again, time constraints have allowed me to only skim these pages. However, much of the material seems redundant with what I read in chapter 4.

Table 7.6, page 7.52: Only 10 states are listed. Does this mean the other 40 states do not have N criteria for freshwaters? That is simply astounding, if true, and deserves to be highlighted in this report.

Also, as I mentioned above, this report MUST also address N criteria from estuaries and coastal waters.

Pages 7.53 and 7.54: the charts are nice looking, but they come with no real explanation. It is important to explain why the EPA is specifying the regional differences in sensitivity to N that are implies in these charts.

Page 7.55, lines 5-7: I disagree that dissolved oxygen is the widespread indicator of problems with eutrophication in estuaries. Chlorophyll, which can be measured by satellite, is a broader indicator.

Page 7.55, lines 19-21: I find it a little puzzling to give Elser et al. (2007) as the only reference that estuaries tend to be N limited. While I fully agree they tend to be N limited, there are many, many earlier papers that make this case, and I believe more convincingly than Elser et al. (2007). I suggest referencing the NRC 2000 report and Howarth & Marino (2006), as well as Vitousek and Howarth (1991) at the very least for this point.

Howarth, R. W. & R. Marino. 2006. Nitrogen as the limiting nutrient for eutrophication in coastal marine ecosystems: Evolving views over 3 decades. *Limnol. Oceanog*r. 51: 364-376.

Page 7.55, line 30, to Page 7.56, line 5: The story of acidification is a little more complicated than stated here, not just algae dying and decomposing: production of carbon by seagrasses is also important, and respiration by living algae and seagrasses during the night can also contribute to acidification. Note that in a eutrophic seagrass bed we studied in Massachusetts, there was a very pronounced diel pattern in pH, with moderate pHs during the day, but by dawn, very acidic waters driven by this respiration by living algae and seagrasses overnight. See:

Howarth, R.W., M. Hayn, R.M. Marino, N. Ganju, K. Foreman, K. McGlathery, A.E. Giblin, P. Berg, and D. Walker. 2014. Metabolism of a nitrogen-enriched coastal marine lagoon during the summertime. *Biogeochemistry* 118: 1-20.

Page 7.56, lines 8-10: I disagree that deposition is typically less than half the N inputs to estuaries. As I discussed in Howarth (2008a, 2008b), I think the evidence points to N deposition being perhaps half of the total N load to Chesapeake Bay and Barnegat Bay, and quite likely more than that New England estuaries such as Casco Bay and Great Bay. Also, keep your Tampa Bay example in mind….

Page 7.56, lines 28-31: The Howarth et al. (2002) reference is fine, but I suggest also adding in Howarth (2008a) and (2008b). And as I stated just above, I disagree with characterizing the atmospheric input to estuaries as generally less than half.

Pages 7.57, 3-15: As with earlier in the Chapter, keep the regional differences in mind: yes, NOy deposition is going down across the US, and NHy deposition up. But that means lower deposition overall throughout most of the East coast. Also, this section seems very dated, with the most recent references from 2008. Surely, there has been new work on the change in deposition since then? For instance, hasn't the Chesapeake Bay Program reported decreased N loads from the Susquehanna River over the past decade, due to lower atmospheric deposition?

Table 7.8, starting on page 7.59: As per my 2008 critique, I believe the SPARROW model underestimates the importance of atmospheric deposition, since it relies on wet deposition as the surrogate for total deposition; the relative importance of dry gaseous N deposition in urban and suburban areas (near NOx emission sources) is far higher than in rural areas, introducing a major bias to the SPARROW results.

Also, note that while Boyer et al. (2002) do indeed give an average relative importance of atmospheric deposition of 31%, the values for watersheds in northern New England were substantially higher; there, atmospheric deposition dominated. This is important to point out in the table. (and Boyer et al. 2002) are probably underestimating N deposition in urban and suburban areas).

Page 7.60, line 13, through page 7.61, line 2: Again, I believe many of these underestimate the importance of atmospheric deposition. Note in particular that I am quoted as saying the importance is 25% in Howarth (2007); that paper was actually published in 2008 (it is 2008b above), and I said up to 50%.

Pages 7.61 through 7.62: I have again skimmed this work; a lot of studies are included, but the selection of papers seems random and not comprehensive, and the section is written in a non-synthetic

way. Are all of these studies of equal value and validity? I do not think so, and yet that is the impression one gets.

Page 7.62, lines 24-27: the statement on the importance of lightning as an N source just before landfall by a hurricane needs to be qualified, or deleted. Otherwise, the reader may come away with the thought that lightning rather than human activities is a general problem, which is simply not the case. Lightning dominated for a very, very brief time. This is interesting, but not important.

Page 7.62, line 31, to page 7.63, line 4: this paragraph on the influence of climate change is very brief, and very light on referencing. In Howarth et al. (2012), we demonstrated larger N fluxes (larger percent delivery of human N inputs) in wetter climates with more discharge, across 150 different watersheds. And in Howarth et al. (2011), we review several mechanisms whereby the climate change that has already occurred over the past decade or so has made estuaries and coastal marine ecosystems more sensitive to the adverse effects of N pollution. See also Naijar et al. (2010) for a review of possible climate change impacts on Chesapeake Bay, including how these interact with N pollution, and Howarth et al. (2000) for a study on the Hudson River estuary in NYC harbor.

Howarth, R. W., G. Billen, F. Chan, D. Conley, S. C. Doney, J. Garnier, and R. Marino. 2011. Coupled biogeochemical cycles: Eutrophication and hypoxia in coastal marine ecosystems. *Frontiers in Ecology & Environment* 9: 18-26.

Howarth, R. W., D. Swaney, G. Billen, J. Garnier, B. Hong, C. Humborg, P. Johnes, C. Morth, and R. Marino. 2012. Nitrogen fluxes from large watershed to coastal ecosystems controlled by net anthropogenic nitrogen inputs and climate. *Frontiers in Ecology & Environment* 10: 37-43.

Howarth, R. W., D. Swaney, T. J. Butler, and R. Marino. 2000. Climatic control on eutrophication of the Hudson River estuary. *Ecosystems* 3: 210-215.

Naijar, R., C. R. Pyke, M. B. Adams, D. Breitburg, C. Hershner, M. Kemp, R. Howarth, M. R. Mulholand, M. Paolisso, D. Secor, K. Sellner, D. Wardrop and R. Wood. 2010. Potential climate-change impacts on the Chesapeake Bay. *Estuarine and Coastal Shelf Science* 86: 1-20.

Page 7.63, lines 5-10: As I wrote above, I find it a little puzzling to give Elser et al. (2007) as the only reference that estuaries tend to be N limited. Here, I suggest deleting Elser et al. (2007), and instead simply use NRC 2000, or perhaps that plus Howarth and Marino 2006.

Page 7.63, lines 11-26: this is a long, qualitative discussion (particularly when it comes to atmospheric deposition of N), and based heavily on a 2002 reference. I suggest using more recent material, and making this more quantitative, or deleting.

Page 7.65, lines 1-15: here, we are returning to the style earlier in the chapter of having large paragraphs devoted to describing one or two studies, often site specific as in the case. There is a very much larger body of literature out there, making the choice of these particular studies very puzzling. The studies are not unique…. What is the point that EPA is trying to develop here?

Page 7.65, lines 16-22: There is a large body of literature on the importance of denitrification, and the two studies cited here fail to do that justice; in fact, the study cited that claims denitrification is not important is very much at outlier: denitrification is a very important sink for N in most estuaries. See the 1996 review by Nixon et al. (Biogeochemistry, 35: 141-179). For an example of its importance in a recent study, see Hayn et al. (2013).

Hayn M., R.W. Howarth, R. Marino, N. Ganju, P. Berg, K. Foreman, A.E. Giblin, and K. McGlathery. 2013. Exchange of nitrogen and phosphorus between a shallow lagoon and coastal waters. *Estuaries & Coasts* doi10.1007/s12237-013-9699-8

Page 7.65, lines 23-27: this brief paragraph focuses equally on denitrification and ANAMOX, but ignores the dissimilatory reduction of nitrate to ammonium (DNRA), a process that was little understood in estuaries as of 2008. The paragraph needs to be expanded to include this, and to better discuss denitrification and its controls; I would not expand on ANAMOX, as this is generally not an important process in most esturairies. And this should definitely include the Nixon et al. (1996), Biogeochemistry 35: 141-179, review which is a critical part of our understanding (for the time before DNRA was fully appreciated).

Page 7.65, lines 28 through line 2 on page 7.57: delete this paragraph, as it contributes little or nothing towards understanding the larger issues upon which this report should be focused. It is very detailed microbiology, taken out of the context of why this might matter in terms of N deposition and estuaries.

Page 7.66, lines 3-11: Since the section title says “sediment processes and DO,” I suggest qualifying in the first sentence that you are talking about DO in the water column (since estuarine sediments are almost always largely devoid of oxygen below 1 mm or so). Also, most of this section is actually on water-column hypoxia, and not sediment processes at all; so I suggest changing the titel to “DO and hyopoxia.”

Also, the causes of low DO need to be broadened: it is not simply about the decomposition of dead organic matter. The respiration of living algae and seagrasses can also lower DO to very low levels, as seen in the deeper portions of the Hudson River estuary where phytoplankton are mixed into the dark

for long periods of time (Howarth et al. 1996), and as seen every night in a eutrophic seagrass-dominated system on Cape Cod (Howarth et al. 2014).

Howarth, R.W., R. Schneider & D. Swaney. 1996. Metabolism and organic carbon fluxes in the tidal, freshwater Hudson River. *Estuaries* 19: 848-865.

Howarth, R.W., M. Hayn, R.M. Marino, N. Ganju, K. Foreman, K. McGlathery, A.E. Giblin, P. Berg, and D. Walker. 2014. Metabolism of a nitrogen-enriched coastal marine lagoon during the summertime. *Biogeochemistry* 118: 1-20.

Page 7.68, lines 11-23: acidification is driven not simply by decomposition of dead material, but can also be the result of CO2 produced by respiration of algae and seagrasses. Howarth et al. (2014) demonstrated high acidiy (low pH) every night in a eutrophic seagrass system. This was driven by respiration of the living plants and algae; during the day, as CO2 was drawn down through primary production, the pH steadily climbed and was no longer as acidic, although the net effect was to drive acidification, due to slow exchange of CO2 with the atmosphere.

Howarth, R.W., M. Hayn, R.M. Marino, N. Ganju, K. Foreman, K. McGlathery, A.E. Giblin, P. Berg, and D. Walker. 2014. Metabolism of a nitrogen-enriched coastal marine lagoon during the summertime. *Biogeochemistry* 118: 1-20.

Section 7.3.3.1, pages 7.68 to 7.70: this section of modeling is strong, a welcome reprieve after reading the earlier material in this chapter.

Section 7.3.3.2: this section too is well done.

Section 7.4.2, pages 7.77 to 7.78: This summary on the N biogeochemistry in estuaries is very short, certainly in comparison to the summary on freshwaters that precedes it. Given that N pollution is the major cause of harm to the majority of the estuaries in the US (NRC 2000; Bricker et al. 2007), this section really needs to be strengthened and expanded.

Chapter 10:

Page 10.2, lines 2-6: the CO2 that drives acidification is not just from decomposition of dead material; during the night, respiration of living algae and seagrasses can also drive substantial acidification. See Howarth et al. (2014).

Howarth, R.W., M. Hayn, R.M. Marino, N. Ganju, K. Foreman, K. McGlathery, A.E. Giblin, P. Berg, and D. Walker. 2014. Metabolism of a nitrogen-enriched coastal marine lagoon during the summertime. *Biogeochemistry* 118: 1-20.

Page 10.2, lines 27-30: I agree with this statement that estuaries tend to be N limited, but I would not use Elser et al. (2007) as the sole reference: that was largely a meta-analysis of short-term bioassays, and the bioassay approach has been heavily criticized by many. I suggest adding two references that address this concern, and yet still conclude that estuaries are largely N limited: our NRC (2000) report, and Howarth & Marino (2006). Further, note that both NRC (2000) and Howarth & Marino (2006) stated that some estuaries are P limited, or colimited by N & P, or switch seasonally between N and P, although N limitation alone dominates.

Howarth, R. W. & R. Marino. 2006. Nitrogen as the limiting nutrient for eutrophication in coastal marine ecosystems: Evolving views over 3 decades. *Limnol. Oceanog*r. 51: 364-376.

Also, in this regard, David Schindler and others have argued that only a whole-ecosystem experiment can demonstrate N limitation, and he further stated there have been so such experiments in estuaries. We address this concern in Howarth et al. (2011). And in the Howarth et al. (2014) study (see above), we present data from an inadvertent whole-ecosystem N addition that clearly shows the estuary to be N limited.

Howarth, R. W., G. Billen, F. Chan, D. Conley, S. C. Doney, J. Garnier, and R. Marino. 2011. Coupled biogeochemical cycles: Eutrophication and hypoxia in coastal marine ecosystems. *Frontiers in Ecology & Environment* 9: 18-26.

Page 10.4, lines 1-12: I read this after writing the comments immediately above; these lines are well done, but I still suggest integrating in some of my thoughts from just above.

Page 10.4, lines 22-28: this section on the importance of atmospheric deposition relies on chapter 7, where I have argued major revisions are needed. So this summary should also be updated. And the use of only referenced from 2008 for the shift in rates of deposition is problematic; there is a much larger, more recent literature out there on decreased NOy deposition and increased NHy deposition.

Section 10.1.2, beginning on page 10.5: this short, qualitative summary is completely redundant with chapter 7. I suggest deleting it, or updating it after chapter 7 is revised.

Page 10.8, lines 5-10: It is worth noting that the most heavily N-loaded estuary in North America (the lower Hudson, NYC harbor estuary) has few if any negative consequences, due to the very rapid flushing of this ecosystem: phytoplankton are flushed away as fast as they can grow.

Howarth, R. W., D. Swaney, T. J. Butler, and R. Marino. 2000. Climatic control on eutrophication of the Hudson River estuary. *Ecosystems* 3: 210-215.

Howarth, R. W., R. Marino, D. P. Swaney & E. W. Boyer. 2006. Wastewater and watershed influences on primary productivity and oxygen dynamics in the lower Hudson River estuary. Pages 121-139 in J. S. Levinton & J. R. Waldman (eds.), *The Hudson River Estuary*, Cambridge Univ. Press.

Page 10.9, lines 9-10: I disagree with the suggestion that DIN is a reliable indicator of N inputs or eutrophication. In the highly eutrophic seagrass system we have studied on Cape Cod, DIN levels are always very low (< 1 uM) despite the very high N load; the DIN is simply taken up too rapidly by the primary producers. See Howarth et al. (2014), referenced above.

Page 10.22, lines 18-20: low dissolved oxygen results not just from decomposition of dead organic matter: the respiration of living algae and seagrasses can also lower DO to very low levels, as seen in the deeper portions of the Hudson River estuary where phytoplankton are mixed into the dark for long periods of time (Howarth et al. 1996), and as seen every night in a eutrophic seagrass-dominated system on Cape Cod (Howarth et al. 2014). In this seagrass system, hypoxia is common at dawn after hours of darkness, while oxygen levels are supersaturated at the end of the daylight period.

Howarth, R.W., R. Schneider & D. Swaney. 1996. Metabolism and organic carbon fluxes in the tidal, freshwater Hudson River. *Estuaries* 19: 848-865.

Howarth, R.W., M. Hayn, R.M. Marino, N. Ganju, K. Foreman, K. McGlathery, A.E. Giblin, P. Berg, and D. Walker. 2014. Metabolism of a nitrogen-enriched coastal marine lagoon during the summertime. *Biogeochemistry* 118: 1-20.

Page 10.26, lines 23, through page 10.27, line 18 (and Table 10.4): In the Howarth et al. (2014) study referenced above, we observed a significant dieback of seagrasses in a eutrophic harbor in 2010, and clearly tied it to the elevated nitrogen load.

Page 10.44, lines 11-20: add reference to Howarth et al. (2014), referenced above, where we showed a very pronounced diel pattern in pH in an N-enriched seagrass ecosystem: pH's were very acidic every dawn at the end of the dark period.