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Interactive comment on "Dynamics of the chemical composition of rainwater throughout Hurricane Irene" *by* K. M. Mullaugh et al.

Anonymous Referee #2

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General Comments:

I find that the article by Dr. Mullaugh et al. is of sufficient quality and scientific rigor that it is appropriate for publication by the ACP. I do think that it is scientifically interesting, new, and appropriate for publication, but with some changes and clarification.

I feel that it needs some rewrite, particularly as it refers to the meteorological implications and clarifications. The chemistry and field work sections are fine, but more clarification is necessary with respect to the meteorology and readability. I have detailed these in my specific comments.

Specific Comments.

Pt. 1. One of my more important comments is the use of the term "air mass". This isn't

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the appropriate term. What I think you mean to say is "air parcel" mostly. A hurricane isn't an air mass specifically, but a subtropical low pressure system operating within a maritime tropical air mass. An air parcel is a smaller body of air, which is the term you should use throughout. Pt. 2. My second major comment is about the idea of high concentration with low rainfall and suppressed concentrations with high rainfall (dilution). I think some of the conclusions and implications need to be reevaluated based on total mass deposition (by component) during the A-K periods. This will move you farther to the implications, given that you are looking at total deposition and get away from some of the dilution/concentration problems with highly variable rainfall. Pt. 3. A few simple things; define the hydrogen peroxide chemical notation Page 4, define RSD, page 4. Only one (or a few) detection limits are reported through section 2, and you might consider listing them all. A table of RSD and detection limits would work very well. Pt. 4. Figures: all of the figures are listed with the same problem; e.g. "total rainfall / mm" meaning the units are in millimeters. I think it would be best for all the figures to be listed using parentheses, "total rainfall (mm)", which I think is more standard for atmospheric journals. Pt. 5. Figures again: I would suggest that the authors consider using radial wind direction figures (i.e. like a pollution roses) for figures 3-6 to show the different concentrations with respect to the wind direction. The concentration is defined along the radius, and the angle defines the wind direction. This would be a great addition to the paper, and make understanding by the reader much easier. I would think pairing these with the time graphs would work well. The same might be considered for figure 1 and a wind rose with speed information incorporated into the same graphic. Pt. 6. Figure 7 F might be best as figure 1. I think the atmospheric scientists would like to see it early. That would define how the hurricane moved, and if you bring all of Figure 7 to the start of the paper, the reader will understand right from the beginning that the early air parcel movement (or "streamlines" if you prefer) came in over the water, and the latter over the land. Pt. 7. Table 1, units and throughout. The air pollution audience will be expecting mg/liter as a unit, to compare it to the numbers they know, rather than micro molar. I think that would be better for understanding, at

least from that audience. Again, clarity and understanding. Also, I assume the sulfate column is measured sulfate and not NSS sulfate. If so, I would add in this column here and mark it as a calculated value. Pt. 8. Page 1, Line 19. How did you clean the glassware for the basic analytes? Just a short description of the washing method. Pt. 9. Abstract: "the second when...". I don't think that this statement imparts what happened. I think it should read something like "when the back trajectories showed that large volumes of marine surface air was lifted" or something like that. Pt. 10. Page 5, Line 5: the NADP could provide the 2011 Na values for each week over the year. Pt. 11. Page 6, Line 4: you might want to justify your selection of 500m for back-trajectories. This much below the likely cloud bottom, and trajectories are really poor at low altitudes (below 100 m). Pt. 12. Page 6, L 14: I would add in a reference here to Figure 1A also. Pt. 13. Page 7, L 9: reference to p value. I would put in the range of the correlation coefficients here. I am unclear as to what the p value is referencing. Pt. 14. Page 7, L 16: versus what? The NADP values? Other? Likely needs a reference. Pt. 15. Page 7, line 23: space missing in reference list. Pt. 16. Page 8, line 1, 15: spaces are missing in two reference lists. Pt. 17. Page 8, L 3: Sulfur is primarily used as a tracer for coal combustion, but NOx is formed with any combustion, so this is a mixed signal. Perhaps further clarification of the sentence. Pt. 18. Page 8, Lines 10-16: This idea is somewhat unclear. Are you arguing that the H2O2 that you measured was formed from photochemical reactions during this hurricane? If so, then I do not agree. The conditions during a hurricane would not favor photochemistry due to low radiation levels with cloud cover and high turbulence, both of which are not common during typical SE photochemical episodes. Perhaps I am misunderstanding what your point is? Clarification of the idea is needed. Pt. 19. Page 8, L 19: elevation. Same air mass comment again. What I think you mean to say is that the elevation of the air parcel? Pt. 20. Page 8, Lines 25-28. Same comment again. I think you mean to say that the trajectories show surface air lifting during this period. Pt. 21. Page 8, beginning discussion. I would suggest adding a sentence or two here that talks about how big the storm was (spatial extend, strength, maximum

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winds), peak winds experienced at Wilmington, etc. so that the reader has a better idea of how close the station was to the center, high winds and the like. Pt. 22. Page 9, Lines 13-16: Good discussion of the idea of low volume/high concentrations. The opposite is also likely happening with periods of high rainfall; dilution. You do bring this up later. Therefore, I would recommend adding a figure based on deposition. What I am thinking is a bar chart showing deposition of individual components during the A-K periods. This will integrate this idea and show when the important depositions occurred during these extreme amounts of rain. Total mass deposition by period. This would again be important in Lines 27-29 same page. Pt. 23. Page 9, L 24: Title. I am not comfortable with the Washout label. Much of the deposition is washout rather than rainout. Therefore, I am not sure what you mean with this label. Pt. 24. Figure 1. I would convert the axis to mile per hour (or a second Y axis), for the understanding of the reader. Most still use miles per hour, and are more comfortable with it. In the same vein, mm to inches in Figure 1 b. I would also suggest that you add in mm/sample bar graph under the integrated cumulative rainfall, so the reader has a better idea of when high and low volumes occurred. I would also label F1a as "surface wind direction" for reader clarity. I would also mark in the time axis of F1a when landfall occurred. Pt. 25. Table 2: reference where the annual values are from. Again, the ammonia values are available for the area from NADP (Beaufort, I believe). Pt. 26. Page 10, lines 10-12: again, the air mass comment. "the back trajectories indicate that the air over Wilm. at the time was originally at the surface and was lifted" or something like that. Pt. 27. Page 10, Lines 18-20. The sentence "the increased..." needs some clarification. Pt. 28. Figure 5, Page 10 lines 26-27. This seems a bit problematic to me. NNS was high during the low volume early rainfalls with maritime air. Then you argue that it goes back up due to air moving over land. This point can be clarified by a deposition plot rather than concentration plots (you stay away from the dilution/concentration problems mentioned earlier). I again would argue for addition of a total deposition plot for A-K for the ions (or major ions if space is an issue). I would expect to see little deposition early of continental ions and heavy depositions later. This point is again shown in Page 11,

lines 1-2. Your concentration plots do not show this for NSS. Deposition plots would likely show your point very clearly. This would also (likely) clarify your argument in lines 11-18. Pt. 29. Table 2: I would add "Concentration" to the column labels. Pt. 30. Page 11: I would bring in data from a nearby NADP site for comparison. You would then be able to show in table 2 (hopefully) high deposition rates of the other components. As it reads, only the CI value is high. Pt. 31. Page 12, Lines 3-5: I would say that this increase is only theorized at this point. Some are projecting this, but it is still speculative. Pt. 32. Page 12, Lines 9-12; same air mass and elevation point as before. Pt. 33. Page 12, lines 11-12. "but a simple.." again this washing out is unclear. Most of the components were washed out, and you have no way to tell if they were washed out or rained out (in cloud). I think you are arguing for not simple dilution? If this is the case, I do agree with this either. All of the concentrations are depressed during high rainfall. Total mass deposition plots will clear this up, I believe. Pt. 34. Figure 7. I would identify, somehow, along the track, when the samples were taken. This will relate to the reader when the samples were taken to the hurricane position. Pt. 35. Figures: some points have error bars and some do not. If you have multiple samples per point, then they should all have error bars. Error bar definition should be stated in the captions. âĂČ

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