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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric inorganic nitrogen input via dry, wet, and sea fog deposition to the subarctic Western North Pacific Ocean" *by* J. Jung et al.

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This is an interesting and relevant study that is presented in this manuscript from an extended cruise on the Western North Pacific Ocean. The authors measured inorganic nitrogen in aerosol concentrations, in sea fog droplets, and in rainwater. They use the "small is beautiful" approach with intensive analyses and interpretation of the few samples that could be collected. They state that this is the first study on the topic (p. 19,110, l. 18–22) and hence provides an essential first information on the relevance of fog in overall nitrogen deposition rates, which the authors quantify at 17% of total inorganic nitrogen deposition (TIN, see their Table 4).

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1 Major Points

Nevertheless, I recommend to consider the following major points in the revision before finally accepting the paper for ACP:

1. The convention that the authors use in their definition of total deposition is not in perfect agreement with conventional usage. Conventionally, dry deposition is considered the sum of gaseous dry deposition plus aerosol particulate matter deposition. Here it is only the latter, whereas many other studies (namely terrestrial studies) neglect aerosol deposition. My suggestion is to make an estimate of gaseous dry deposition in order to obtain the correct relative deposition rates in the manuscript, namely in Table 4. On terrestrial ground in the mid-latitudes it is often the case that (gaseous) dry deposition is on the order of two thirds of total deposition, and hence the relative relevance of fog is lowered if gaseous dry deposition is also considered. It may be that it can be safely assumed that gaseous dry deposition is a small component in the total deposition because of low (background) concentrations; but if it is like this, then it should be stated in the manuscript. For reference our deposition estimates for some Swiss localities can be found in Burkard et al. (2003) and Eugster et al. (1998).

2. Statistics. This is a flaw that is found throughout the scientific literature and hence is not specific to this paper, but I feel obliged as a reviewer to make sure statistics are correctly used. In this particular case it is obvious that wherever the standard deviation of reported numbers is larger than the mean – notably for concentrations which cannot be negative – then this is an indication that the wrong statistics are used. Mean and standard deviation are the two parameters of a normal distribution; roughly 68% of the data lie within the mean $\pm$ 1 standard deviation, and 16% are **smaller** than mean – 1 standard deviation. This is impossible for cases where the standard deviation is larger than the mean and simply indicates

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that the distribution of the data is not following a standard normal distribution. In such cases the mean may still be meaningful (as the sum divided by the number of samples), but the standard deviation is no statistical parameter for the actual distribution. Hence, either an appropriate model distribution for the data must be found and the respective parameters reported, or (more simple) robust statistics using the empirical data distribution should be used (reporting median, interquartile range, and possible 95% interval if this is relevant for the reader to know). The case is very obvious on page 19,098, but also elsewhere (abstract: numbers for wet and sea fog deposition, but also Tables 1 and 4). You can use a statistical test to test your data for normal distribution (e.g. Shapiro-Wilks test) to convince this reviewer that he's wrong.

3. Following up on the question of the correct distribution of your data the t-test used on page 19,098, l. 8 may not be the appropriate test statistics. Please update according to your finding and reply to the previous point. In many cases the t-test is rather robust to the violation of the assumption of normal distribution, so it is not impossible that your results with the correct statistical test are similar to what the t-test provided, but for the sake of scientific correctness I encourage you to use the appropriate statistical test that applies to your empirical data distribution (maybe a nonparametric test is needed if you cannot define the general statistical distribution model for your data).

4. The influence of the vessel's exhaust is minimized in their sample strategy by using conditional data selection with two criteria: (1) wind direction is from within $\pm 100^{\circ}$ from the bow direction, and (2) wind speed exceeds 1 m s^{-1}. This appears to be a sound approach for data selection in order to minimize the contamination from the sampling vessel. The only concern I have here is with respect to the fog droplet sampling which unfortunately is a passive system (no aspiration as e.g. a CASCC would do), so there might be cases where initially the conditions were within the rejection range defined by the authors, but later they were OK and fog

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might have been collected on the collection mesh that has previously received some contamination from soot from the vessel's enginge. I assume the authors have carefully avoided such conditions. They state in Section 2.3 their careful procedure to clean an prepare the sampling tissue, but there is no mention what and how it was assertained that there was no contamination of the kind I could imagine as explained above. Maybe add a short statement saying exactly how you did this, or provide a statement that such conditions are unlikely (maybe they can be neglected since wind was never low? However, I imagine that fog and low wind speeds correlate in such cases).

5. Usage of the term "subarctic" Pacific Ocean: there is a good overview provided by Bailey (1996) who defines the subarctic region as a subdivision of the polar domain, but only for land surfaces. Nevertheless, in the eastern Siberian and Alaskan region the subarctic region does not extend to latitudes below 50° N. On his map of the Ecoregions of the Oceans in that book your cruise is entirely in the "temperate" domain (as I would have expected). So my suggestion is to avoid confusion and **not** use the term subarctic for your study; any reader in my domain would expect a cruise going up further North in order to touch the subarctic region. Use the term "temperate" instead.

2 Minor Points

1. Parameters vs. variables. You use the word parameter for variables, which unfortunately is quite widespread in meteorology and other disciplines, although parameter in science is actually a pseudo-constant, not a variable. My suggestion is to call variables "variables" and reserve the word parameters for e.g. model parameters of a statistical distribution etc. (mean and standard deviation are such parameters).

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2. Section 3.7 (Deposition flux estimates) is a methods section and should be moved to an appropriate position in Section 2 (Methods)

3. Section 3.1, meteorological conditions: in Bruijnzeel et al. (2005) we tried to group published literature according to naming conventions for fog types, and distinguished between “sea fog” (fog forming from atmospheric vapor when warm air rests over cold ocean) and “steam fog” (fog forming from evaporation from a warm ocean into a cold atmosphere). The figure that I made to clarify the usage of words was also reprinted in the introductory paper of the special issue of the 2007 fog conference [Eugster (2008)], downloadable from http://www.die-erde.de/DIE_ERDE_2008_1-2_Eugster.pdf. I know that it is difficult to establish a nomenclatura in fog types, but if you agree with what was published earlier, it would help newcomers to try to use these wordings.

4. p. 19,093, l. 6: it is unclear why you call this impactor a “virtual” impactor. As I understand your description it is a “real” impactor, that is, the particles impact on the filter. It the terminology established to call this a virtual impactor? Please clarify.

5. p. 19,094, l. 8–9: was the rain sampler opened manually? Most likely so, but it would help to explicitly state that this was not an automatic procedure.

6. p. 19,095, l. 16–22: sounds perfect, but be aware of the fact that the assignment of FM-100 signals droplet sizes is somewhat more complex than the standard procedure that we, you and others have used so far. Please have a look at Spiegel et al. (2012) and then decide. My best guess is that in your case the Mie scattering effect is within the uncertainty of your deposition estimates anyway, but only you know exactly how you operated the FM-100.

7. p. 19,110, l. 6–8: to obtain such an estimate you most likely must have made an assumption on the C:N ratio to yield an estimate for N requirements based on

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C flux data. Please specify what assumptions you have made, which literature values (with references) were available for making such an assumption, or specify what other basis you used for this estimate

8. Table 1: you honestly mention that you used a value of 0 for cases with negative concentrations or concentrations below detection limit. This however is not the standard approach as I understand; see for example Helsel (1990). Your numbers should not change much, but for the sake of scientific correctness I recommend using the procedure suggested by Helsel.

9. Table 2: caption does not mention that also seawater ratios are given.

10. Fig. 8: I am surprised how large the droplets are in your fog! But that's probably the characteristics of this fog and is fine. But I find the labels on the x-axis misleading where you label 20 μm with 2, and 40 μm with 4, a factor 10 off, which should be corrected

11. Fig. 10: nice overview, but since there is a different reference for aerosol concentrations than rainwater/fog water, the circles are misleading in that they imply a much higher aerosol concentration than there actually is. Would it be possible to draw the circles to scale, and since for aerosols they would be utterly tiny, use a zoom and show the two circles that you present now in a graphical display that clearly indicates the reader that these two were magnified (e.g. small box removed to the left and lines connecting it to the same display at correct size).

3 Typographical Details

- p. 19,090, l. 11: delete “s” in reveal (it is plural)
- p. 19,091, l. 7: add “s” to constituents (plural)

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- p. 19,109, l. 23: “co-limitation” is typically written with a hyphen

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