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Interactive comment on “Assessment of excess N₂ and groundwater N₂O emission factors of nitrate-contaminated aquifers in northern Germany” by D. Weymann et al.

Anonymous Referee #3

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The manuscript "Assessment of excess N₂ and groundwater N₂O emission factors of nitrate contaminated aquifers in northern Germany" uses a valuable data set to address important topics about N-cycling in aquifers. The manuscript will be of interest to readers of BG. Revisions are needed, however, before the paper will be suitable for publication. Some important details should be added (e.g. What are the excess air concentrations? What is the depth to the water table at these sites? How were the values calculated in Table 2?). The results section should be expanded, and the results should be more thoroughly discussed in the context of existing studies. At the same time, some tangential or tenuous points can be eliminated or reduced in length. Lastly, attention should be given to carefully defining the topic and objectives of this

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work. For example, the title includes "excess N₂" which is hardly mentioned in the conclusions; the abstract lacks a final sentence that would give the reader a strong sense of the main point(s) of the paper; and the conclusion does not address many of the issues raised in the discussion section. Specific suggestions to remedy these and other issues are given below.

P1264 L18-20 - Suggest replacing the sentence "According to denitrification intensity..." with a more straight-forward definition of EF₂. L25 - The Abstract seems to lack a concluding statement that highlights the most important finding(s) of this study and gives the reader a sense of the implications of these results. The current closing sentence is a site specific result with unclear implications.

P1265 L3 - Why "layers"? Suggest a more general term like "zones". L6-16 - The introduction jumps too quickly to details of methods of study without first establishing why this sort of study and this particular study are important contributions. Much of this information could probably be omitted or moved to the methods section. L21 - suggest "result from" in place of "are associated with" L21-26 - This is a good concise statement of the context of this study in the existing literature.

P1266 L6 - omit "presumably" L9 - spell out "IPCC" L12 - Suggest "Typically" in place of "Principally" L15 - "and" instead of "und" L18 - "reactions" instead of "change"

P1267 A map would be helpful to show the locations of the sites. L18-21 - This sentence is confusing. "respectively" appears twice. L23 - Please provide depth to the water table at these sites.

P1268 L2 - Suggest "pumping" instead of "suction". It seems unlikely that reducing the rate of pumping would make much difference. Perhaps move this comment to below the discussion of comparison of results from different pump types. L6 - Suggest using SI units instead of inches, and "in these wells" instead of "here" L11 - "proves" is too strong a word. Maybe "indicates" or "implies". Comparing treatments is a good approach, but the reader needs to know the vertical distance from pump to water table

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at each site to evaluate the validity of extrapolating the comparison from one site to another. L27 - What does "quantitatively removed" mean? Please clarify.

P1270 L5 - Suggest omitting "Principally" L9 - "in excess of..." L10 - Suggest "near the water table" instead of "at the groundwater surface"

P1271 L12 - Please clarify which value is meant by "this value". L24 - Suggest "to assess" instead of "assessed"

P1272 L4 - What are the units for these standard deviations? L8 - Please explain or provide a reference for "Gaussian error propagation" L10 - Suggest "Initial NO₃- concentration at a given location on the aquifer surface is defined by the NO₃- concentration of the recharging water before alteration by denitrification in groundwater." L13-18 - Suggest revising this sentence for clarity. L23 - Suggest omitting "is generally correlated with excess N₂ in denitrifying aquifers"

P1273 L7 - Suggest defining EF₂ with an equation in the same fashion as EF₁. L16 - Suggest simplifying this header L18 - Suggest "Groundwater temperatures at these sites were relatively constant..." L22-24 - Is there any obvious explanation of why pH varies among these sites?

P1274 L2 and Table 2 - please explain the meaning of these mean, min and max values. Are they all "means from the minimum and maximum values" (P1271, L25)? This is confusing. For example, some readers will likely assume that the "min" values of excess N₂ are calculated using Eqs 1 and 3. L2-3 - It seems unnecessary and convoluted to say that lowest values of excess N₂ coincided with low RP, and that high values of excess N₂ coincide with high RP. This is more a mathematical implication of the definition of RP than a result of this study. L9 - Should this say "denitrification is complete in parts of the Fuhrberg aquifer"? L12-13 - Is this difference statistically "significant"? What is the p-value? L14-15 - Suggest "The presence of excess N₂ demonstrates that..." L21 - These EF values were highly variable among the sites, or within each site? Please explain. L22-23 - Suggest "was in agreement" L25 - By

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definition, $EF2 > EF1$ at sites with denitrification, so this seems unnecessary to report as a result. Is there something more to say about this? L25-26 - Should this say "Among the sites"?

P1275 L13-26 - The first half of this first paragraph of discussion repeats much introductory material. Suggest omitting or moving to the introduction.

P1276 L3 - Please relate these values to the "UN2" term defined by equation 4. L7 - "the uncertainty of RP is relatively small and does not affect L1-14 - At this point in the discussion, it is distracting to read a defense of results that were given earlier in a separate section. I suggest including these calculations in the results section and including error bars on Figs 2 & 3 to show the reader the potential importance (or lack of importance) of the error associated with assuming total dissolution of entrapped bubbles. Also, Figure 3 should be introduced in the results section. The "relationship" shown in Figure 3 is mentioned here, but it hasn't been mentioned previously and it is not explained here. I agree that the topic of uncertainty in these estimates worthy of discussion, but a discussion section should provide a broader perspective on how these results are relevant to the existing literature. For example, what does this say about previously published values of excess N₂ and associated denitrification rates? Are the errors small enough that we can trust the published values, or should we be concerned about a potentially significant bias? L14-26 - This paragraph seems tenuous. If it is an important point, maybe the data (e.g. sum of partial pressures versus hydrostatic pressure) should be shown.

P1277 L1-20 - The point made by this paragraph seems very weak. Giving this much space and attention to a comparison with a previous modeling estimate (not even real data) distracts from more important points raised by this study. One sentence in the results section should be sufficient for this point. Section 4.2 - Here it would be nice to see some comparisons with previous studies. Have these sorts of correlations been seen elsewhere? A broader context would make these arguments more compelling.

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P1278 L3 - Does "partial" refer to a part of the entire data set or a part of the Fuhrberg data set? L5 - Temporal and spatial or just spatial? Suggest omitting "which implies that the relationship between reaction progress and residence time was more variable." L13 - "found" instead of "evaluated" L16-17 - Is it possible that N₂O correlates with pH because N₂O correlates with NO₃⁻ and NO₃⁻ correlates with pH? Acidification due to greater fertilizer applications explains the NO₃⁻ / pH correlation. L19-20 - Omit "This regulation is illustrated... in our study."

P1279 Section 4.3 - Again, a more thorough comparison with existing studies would strengthen this discussion section. Is the result of higher emissions between RP=0.2-0.6 a new finding? Does it agree or disagree with previous results in other studies? L27 - The assertion that emission is negligible at RP close to zero appears not to hold true at Thulesfelde. Is there anything unique about this site that would explain the difference?

P1280 L13-14 - Earlier in the paper (P1274 L23) a comment is made that EF₂ values are similar to EF_{5-g}. Given the large range of both EF estimates among the sites, can a stronger case be made that one is any better than the other? Or are these EF values more or less random. Are EF values worth calculating, or should a different approach be used? L26 - "was" instead of "could be"

P1281 L7 - "EF(1)" instead of "EFs(1)" L17 - Are there any important conclusions about uncertainty of excess N₂ / N₂O / initial NO₃⁻ estimates? Any conclusions about regulating factors for N₂O production?

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