

Review of MS acp-2017-1094:

“Estimates of Exceedances of Critical Loads for Acidifying Deposition in Alberta and Saskatchewan” by P.A. Makar *et al.*

submitted for publication in *Atmospheric Chemistry and Physics*.

General remarks:

This rather extensive paper reports on detailed deposition calculations for the two Canadian provinces as well as their use in exceedance calculations for different sets of critical loads (CLs) for terrestrial and aquatic ecosystems. As the larger part -- and more of the novel material -- is concerns atmospheric depositions, I suggest to change the title to “Estimates of Acidifying Deposition and Critical Load Exceedances in Alberta and Saskatchewan” (or similar), and also to restructure the paper accordingly, i.e. first depositions, and then CLs and their exceedances. With respect to depositions I suggest to move some of the material to the ‘Supporting Information’, since it’s mostly material taken from existing literature. The paper (and the reviewer) would have benefitted if the authors had carefully read the paper before submission: there are close to 30 references that are there and not cited or cited and not in the reference list (see below); also, the equation numbering is quite faulty in some parts. Furthermore, the definition of critical load exceedances (in case of non-exceedance) requires some attention (see below). Apart from this, I consider the material of paper suitable for publication, after the authors have also taken into consideration the (often minor) remarks/corrections listed below.

Detailed remarks:

Note: ‘ $X \rightarrow Y$ ’ means: replace ‘ X ’ by ‘ Y ’ (in the text).

Title:

See ‘General remarks’ above.

Abstract:

P[age]1, L[ine]33: Suggest to change ‘protocols’ to ‘methods’ (throughout the paper!), as ‘protocol’ has its own meaning in the context of CLRTAP!

P1, L34: Delete ‘forest and’: forests are terrestrial ecosystems!

P2, L2: Delete ‘emissions and’.

P2, L7: ‘was shown to have’ \rightarrow ‘has’ (otherwise it sounds the authors have shown that in this paper).

P2 L11: ‘primary particle dust particles’ \rightarrow ‘primary dust particles’.

Introduction:

P2, L17: Delete ‘regional and’.

P2, L18: The reference to the CLRTAP Manual sh/could be simplified (throughout the whole paper!), as it’s always the same source. Just call it always CLRTAP (2017) -- with the text in the References as is now under ‘CLRTAP, 2004’ -- since 2017 is the (last) time you accessed it.

P3, L 16: ‘Estimates of critical loads’ \rightarrow ‘Critical loads’.

P4, L1-2: No! If BC_{dep} is greater than $S_{dep} + N_{dep}$, a large part of that BC_{dep} could be taken up by forests and harvested (i.e. taken away) and thus not be available for neutralising the S and N deposition; and a case could also be made for the converse -- If the statement were true, then the CL would be equal to BC_{dep} !

P4, L5: ‘emissions levels’ \rightarrow ‘emission levels’.

P4, L7: What is ‘alkylation’? (I guess the authors mean ‘alkalinisation’?).

P4, L24: ‘aquatic and terrestrials’ \rightarrow ‘aquatic and terrestrial ecosystems’.

P4, L25: Insert ‘deposition’ after ‘surface’.

Methodology:

P5, L15: ‘sum of in equivalents of’ → ‘sum of’; the criteria is generally reported as ‘molar Bc:Al (or Al:Bc) ratio! That’s way the factor 3/2 appears in eq.4 to convert it to equivalents!

P5, L19: There is no ‘level of protection’ defined for CLs.

P5, L19-20: It’s the chemical criterion (here Bc:Al ratio) that *defines* the critical ANC leaching – the user does not *specify* the critical ANC leaching (in the case described here), he just computes it!

P5, eq.2: ‘(CL_{max}(S)/(1-f_{de}))’ → CL_{max}(S)/(1-f_{de}); i.e. drop the superfluous parentheses.

P5, eq.4: Delete the superfluous parentheses (twice); only the square brackets are needed.

P5, L30: Bc is already explained above (line 15).

P5, L31: Insert ‘annual’ after ‘long-term’.

P6, L1: ‘due to other forms of removal (e.g., harvesting)’ → ‘due to, e.g., harvesting’.

... and all variables in the text should be in *italics* if they are so in the equations (also further below)!

P6, L8: ‘Q’ is already defined on line 1.

P6, eq.6: Y does not stand for Ca+Mg+K+Na-Cl, but Σ_Y stands for the sum of base cations minus chloride (Ca+Mg+K+Na-Cl).

P6, L28: ‘(i)’ → ‘(subscript i)’, etc.

P7, L4: What is ‘charge x mole equivalent’? moles? moles of charge? ...

P7, L22: There is a change in font size from that line onwards – Any reason?

P8, L1: ‘In some instances, S deposition (or N) must be reduced to achieve non-exceedance’ What do the authors want to say? As it stands, it’s trivial/obvious.

P8, Figure 1: (a) Why is the slope of the Critical Load Function (CLF) shown as 45°? This is a special case only for f_{de}=0 (see eq.2); (b) The point (N₀,S₀), computed in eqs.12,13 should be shown on the Figure; (c) It should be indicated in the Figure how the quantity E₀ is derived, i.e. where N_A and S_A are located on the CLF.

P8, L5: ‘denotes ecosystem’: No, it does not denote ecosystems, it denotes ‘the case for which’.

P8, L15: E₀, as a negative quantity, cannot be a distance, only a positive quantity can; e.g. |E₀| = -E₀ (thus it would be better to define E₀ as a positive quantity and make it -E₀ in eq.11)!

P8, L15: There are no ‘exceedance lines’ – what you mean is the critical load function.

P8, eq.15: In fact, N_A is the N_{dep}-value on the CLF for a given S_{dep}, and S_A the S_{dep}-value on the CLF for a given N_{dep}. It can be easily shown that S_{dep}-S_A is always greater than (or equal to) N_{dep}-N_A, or, to express it in positive terms (i.e. distances): S_A-S_{dep} ≤ N_A-N_{dep}. Thus eq.15 simplifies to:

$$(15) \quad E_0 = \begin{cases} S_{dep} - CL_{max}(S) & \text{for } N_{dep} \leq CL_{min}(N) \\ m(N_{dep} - CL_{max}(N)) & \text{for } CL_{min}(N) < N_{dep} < CL_{max}(N) \end{cases}$$

... and eq.16 becomes superfluous.

P9, L5: Insert ‘critical loads’ after ‘estimated’.

Note: For computing E₀ a different distance measure is used than for computing positive exceedances. This is not really faulty, but peculiar, and should at least be mentioned (and maybe ‘justified’). More generally, the authors should give reasons why they map negative exceedances, as policy makers might not be so much interested in them; generally, they are ‘happy’ when there is non-exceedance (however small) ... But it makes ‘nice’ maps; and maybe there is another reason as well ...

P9, L11: ‘... in order to obtain data for critical load estimates’: Only for that purpose?

P9, L15/16: ‘... and other related information’: What else c/would that be?

P9, L16/17: ‘estimates ... were conducted’: Do you really conduct estimates?

P9, L20: Delete ‘lowest’.

P9, L21: ‘for grid cells containing *smaller* number of lakes, the ... most sensitive lake was used’: Smaller than what? (20? ...). And, by the way, percentiles can be computed for any number ...

P9, L22/23: 45 km² grid cell – What’s that? If it’s a 45 km × 45 km grid cell its size is 2025 km²; if it’s area is 45 km², what are the lengths of the sides? – It’s no problem to call it a 45 km grid cell in the former case ...

P10, L5: Only BC deposition is needed for CL calculations.

P10, L6: Critical alkalinity leaching is not an input, but calculated from, e.g. a critical Bc:Al ratio.

P10, L14: ‘(a) a critical Bc:Al ratio of 10 and a K_{gibb} of 3000.0 were used’: This is not a *simplifying* assumption! Furthermore, provide units for this numbers!

P10, L16: Replace ‘invariant’ by ‘spatially uniform’.

P10, L16/17: ‘the equivalent of the $(CL_{max}(S)/(1-f_{de}))$ term ...’: Although I can infer what you want to say, it’s confusing for the non-expert. What you are doing is modelling denitrification as a constant flux N_{de} , (i.e. $CL_{min}(N) = N_i + N_u + N_{de}$) and not with the fraction f_{de} , as in eq.2 – but this has to be explained (better)!

P10, L17: Why the somewhat ‘awkward’ number 35.7? Why not 36, or 40? Tell the reader that it comes from a nice round number in a different unit and an assumed soil depth of 0.5 m (this is a simplifying assumption that should be mentioned!)

P10, L18: ‘weathering ... was assumed to be dependent on temperature’: This is not really a simplifying assumption to me.

P10, eq.17: (a) the minus sign should be a plus sign! (b) Why do you insert the numbers (2 times 35.7) for N_i and N_{de} , but not for $(Bc:Al)_{crit}$ and K_{gibb} ? (c) Some reason should be given why this is now called $CL(S+N)$, and not $CL_{max}(N)$ as in eq.2.

P10, eq.18: The square brackets are superfluous.

P10, L23: ‘ $Bc_{we} = 0.75 BC_{we}$ ’: this is another simplifying assumption, that should be mentioned above.

P10, L27: (a) I guess it’s 2.5 km × 2.5 km (or 2.5 km for short) grid cells. (b) Delete ‘lowest’.

P10, L31: Parentheses around $S_{dep} + N_{dep}$ superfluous.

P11, eq.19: This is a ‘dangerous’ equation! What if $N_{dep} < N_i + N_{de}$? The remaining N-sink can **not** compensate any S deposition! Maybe it does never happen in AL and SK (?), but it has to be said that the (potentially) remaining N sink is not used to compensate S_{dep} , as the equation does as it stands!

P12, L1: ‘permutations’ seems a strange expression in this context!

P12, L11: ‘Soils’ → ‘Soil’.

P12, L20: How deep was the rooting zone?

P12, L27: Comparing $CL_{max}(S)$ with $CL(S+N)$ does not make much sense, as the latter includes N-terms; thus $CL(S+N)$ could be compared to $CL_{max}(N)$.

P14, L4: ‘(equations 1 through 16)’: This is misleading; e.g. eqs.1-4 describe the CLs for terrestrial ecosystems. Improve the citation of equation numbers in the whole paragraph! By the way: the exceedance calculations given in eqs.11-16 are not fully correct for FAB CL functions, as also the first segment of the CLF is tilted.

P14, L6: ‘predictive maps’: wouldn’t ‘interpolated maps’ be more appropriate?

P14, L6/7: ‘four target variables’: In the next 2 lines only 3 variables are mentioned (BC, DOC, SO_4^{2-}); what’s the 4th one?

P14, L16: ‘wetlands’ → ‘peat’.

P19, L1: ‘Yao’ → ‘Yau’.

P21, L20 – P22, L4: This paragraph goes into very much detail ... Maybe to Supp Info?

P22, L7: Isn’t sulphate an anion?

P22, L12: ‘Manzaono’ → ‘Manzano’.

P22, L13 and eqs: The equation numbers are wrong: On page 11 you had already eq.19!!

P22, L14: ‘used in equation (18)’: Which eq.18?

P22, L24: Delete ‘in 2014’ (?)

Results:

P25, L1: Insert 'in' after 'result'.
 P25, L3: 'input emissions' → 'emission inputs'.
 P26, L4: Sub-header: 'oil sands' → 'Oil Sands region' (?)
 P27, L28: 'simulation, of' → 'simulations by'.
 P31, L30/31: merge lines.
 P33, L13: Delete parentheses around ' $BC_{dep}-S_{dep}-N_{dep}$ '.
 P34, L16: 'sampling for' → 'sampling to monitor'.
 P35, L9: 'critical load exceedances' → 'critical loads and their exceedances'.
 P35, L13: Sub-header: 'Exceedances with respect to' → 'Exceedances of'.
 P36, L18: 'Columbia': **NO!** Columbia is 100 times larger (about 1.14×10^6 km²)!
 P38, L3: 'have increased in size relative to' → 'are larger than'.
 P40, L1: Sub-header: 'Exceedances with respect to' → 'Exceedances of'.
 P40, L6: 'equation (7)': No, it's equ.(5), I presume.
 P40, L6: 'Canada-Wide' → 'Canada-wide'.
 P40, L11: 'superimposed in' → 'superimposed on'.
 P40, L20: 'to be in exceedance' → 'to be exceeded'.

Discussion:

P46, L12: 'improve the bias and correlation' → 'reduce the bias and improve the correlation'.
 P46, L30: 'expected to occur' → 'expected to occur or has occurred'.
 P47, L4: 'of Figure 17(b)': Or 18(b)? As said in the caption of Fig.20

Author Contribution:

P51, L7: 'Lakes and Forest' → 'lakes and forest'.

References:

The following **references** are cited in the text, but **missing** here:

- NPRI, 2013
- Aherne, 2013
- Aklilu, 201x
- ECCC, 2014
- Nasr et al., 2010
- Whitfield et al., 2010
- Pregitzer et al., 1990
- Stockwell et al., 1989
- Gong et al., 2003a
- Gong et al., 2003b
- Gong et al., 2015
- Makar et al., 2017
- Wesely et al., 1989 [or is this the same as Wesely, 1989?]
- Slinn, 1982
- Jacobson, 2003 [or should it be Jacobson, 1999, which is given, but not cited?]
- Watmough et al., 2015 [or should it be 2014?]
- Whaley et al., 2017

The following **references** are **superfluous**, as they are not cited in the text:

- Brook et al., 1999
- Dasch and Cadle, 1986
- Ellsworth and Reich, 1993
- Environment and Climate Change Canada, 2017 [I guess that's ECCC, 2017, cited in the text!?!]
- Henriksen, 1984
- Hicks et al., 1987
- Hosker and Lindberg, 1982
- Jacobson, 1999 [maybe 2003? -- see above]
- Meyers et al., 1998

- Sverdrup and De Vries, 1994
- Voldner et al., 1986
- Wesely and Hicks, 2000

Figures:

Figure 1: Improve as suggested above.

Figure 2: Caption: ‘Lake (S_{dep})’ is a somewhat strange notation. Why not give the equation number used to calculate the CL. Same for ‘Forest ($S_{dep}+N_{dep}$)’.

Figure 4: Caption: Replace ‘ $S_{dep}+N_{dep}$ ’ by ‘ S_{dep} and N_{dep} ’ [and add ‘(FAB model)’ after it].

Figure 5: Caption: ‘Sulphur’ → ‘sulphur’ or ‘S’.

Figure 6: Caption: ‘Nitrogen’ → ‘nitrogen’ or ‘N’.

- Plate (i): Isn’t it the **sum of** particulate nitrate (dry), gaseous organic nitrate (dry), etc? And not **each of!**?

Figure 13: Caption: Add that Alberta and Saskatchewan are shown in the maps (?) [also in other Figures!]

Figure 14: Caption: Add that Alberta is shown in the maps (?) [also in other Figures!]

- Year after Wang *et al.* is missing.

Figure 15: Caption: Add ‘(see Figure 1)’ to explain the regions 1,2,3,4! The term ‘region’ in this context is a bit confusing – e.g., ‘cases’ would be clearer, to distinguish from geographical regions.

Figure 19: Caption: Add ‘(see Figure 1)’ to explain regions 1,2,3,4!

Figure 20: Caption incomplete!