Review of MS acp-2017-1094:


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.
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 ‘alkylization’? (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 Be:Al (or Al:Be) 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 Be: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 ΣY 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 (N_{A},S_{A}), computed in eqs.12,13 should be shown on the Figure; (c) It should be indicated in the Figure how the quantity E_{0} 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: E_{0}, as a negative quantity, cannot be a distance, only a positive quantity can; e.g., |E_{0}| = –E_{0} (thus it would be better to define E_{0} as a positive quantity and make it –E_{0} 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:
\[ E_{0} = \begin{cases} S_{dep} - CL_{max}(S) & \text{for } N_{dep} \leq CL_{min}(N) \\ m\left(N_{dep} - CL_{max}(N) \right) & \text{for } CL_{min}(N) < N_{dep} < CL_{max}(N) \end{cases} \]
… and eq.16 becomes superfluous.
P9, L5: Insert ‘critical loads’ after ‘estimated’.

Note: For computing E_{0} is 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’.
Results:
P25, L1: Insert ‘in’ after ‘result’.
P25, L3: ‘input emissions’ ➔ ‘emission inputs’.
P25, 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, L6: ‘sampling for’ ➔ ‘sampling to monitor’.
P36, L18: ‘Columbia’: NO! Columbia is 100 times larger (about 1.14 ×10^6 km^2)! 
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, 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’ $\rightarrow$ ‘sulphur’ or ‘S’.

Figure 6: Caption: ‘Nitrogen’ $\rightarrow$ ‘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!