**Hydrology and
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Interactive comment on "Surface water acidification and critical loads: exploring the F-factor" *by* L. Rapp and K. Bishop

CJ Curtis (Referee)

c.curtis@ucl.ac.uk

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General comments

This paper is a valuable contribution to discussions about the validity of critical load model assumptions under the current situation of declining acid deposition across much of Europe. In particular, it develops the definition and interpretation of the mysterious "F-factor" that is a key component of static critical load models and highlights the shortfalls of static SSWC family of models and the assumptions they require.

The paper is highly significant because of the major reductions that have been achieved in acid deposition across Europe but the continued exceedance of critical loads in some

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sites in several countries which require further reductions that are difficult (expensive) to achieve. Hence understanding the uncertainties and reliability of calculated critical loads in these most sensitive unprotected sites is of paramount importance.

The paper is technically very good and the only revisions I would recommend are for further development of some of the discussion, especially regarding the necessary assumptions of static models used for large numbers (thousands) of sites often with only a single spot chemistry sample used for critical loads, compared with the more realistic but much more data intensive dynamic acidification models. In particular, the international critical loads mapping exercise co-ordinated by the CCE for the UNECE CLRTAP has moved towards simple dynamic modelling (e.g. VSD model) and empirical approaches based on upscaling from sites where data are available for full dynamic model application to sites with only water chemistry and catchment attribute data.

Another issue worthy of further discussion is the related issue of critical load exceedance calculated using SSWC or FAB and the steady-state assumptions which mean that current exceedance of critical load does not necessarily indicate current damage. This is again related to the dynamic nature of "F" and is closely linked to the issue of temporally varying critical loads and F. Some of these issues have been discussed in a previous paper on this subject: Curtis, C.J., Reynolds, B., Allott, T.E.H. and Harriman, R. (2001) Critical load exceedance and biological damage: a re-interpretation. Water, Air and Soil Pollution: Focus 1 (1-2), 399-413.

Specific comments PAGE 3919 Definition of critical load (lines 1-5) is very loose – why not give the full standard definition of Nilsson & Grennfelt?

Line 6: ambiguous – specify that deposition is declining rather than increasing towards the critical load, i.e. critical loads were exceeded in the past and still are – this provides the introduction to the key point which follows that as exceedance approaches zero, uncertainties become more important especially as further emission reduction become harder and more expensive to achieve.

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PAGE 3921 Line 17: I would go further and say that the term [BC*]0 effectively defines the critical load, as it provides the critical level of base cation leaching to maintain the critical ANC value, i.e. it is the fundamental basis of the SSWC critical load.

PAGE 3922 Line 1: Agree that reference to nitrate may not be necessary in Swedish context but perhaps worth a mention that in other countries e.g. UK, Italy, nitrate may be a major acid anion contributing to acidification.

PAGE 3927 Line 14: Worth mentioning the long-term increasing trends in DOC observed across much of N Hemisphere here (e.g. Monteith D.T. et al 2007. Dissolved organic carbon trends resulting from changes in atmospheric deposition chemistry. Nature 450: 537-541.). This is also likely to affect calculations of pre-industrial chemistry even though not directly referred to in SSWC model – affects calculation of alkalinity from ANC (equation 15)

PAGE 3928 Lines7-8: the restriction of 0<=F<=1 is explicit in some of the original papers describing its calculation and is easy to understand; F=0 implies all S deposition is accompanied by hydrogen ions into surface waters so each unit of sulphate contributes one unit of acidity with no ion exchange in soils. F=1 implies all hydrogen ions are exchanged in soils for base cations with no net contribution of acidity or decline in ANC in surface waters per unit deposition of S. Values of F outside the range 0-1 should not be possible IF the assumptions of the steady-state models are valid? The theoretical implications of computed values outside this range must be discussed further in Section 4.1, especially the meaning of F>1.

PAGE 3930 Lines 1-3: this is a key point of the paper, demonstrating the inadequacy of the SSWC model in the context of declining deposition.

Section 3.2: More discussion is required here about the limitations of a simple steady-state model like SSWC compared with more realistic but data-intensive models like SAFE. It might be construed that a key conclusion from this paper is that the assumptions of the steady-state models are clearly inappropriate for a situation of changing

deposition, especially when declining, and this has major controversial implications for the widespread use of critical load models around Europe.

PAGE 3932 Lines 15-20: this is a key conclusion from the paper – the assumptions of the steady-state models are not valid during the recovery phase when F can be <0! And yes – the pre-industrial BC concentration is assumed to be constant under a theoretical steady-state.

Technical corrections PAGE 3919 Line 17: no capital in "The US"

Lines 18-19: A key component. . .. IS the F-factor in the Steady State Water Chemistry Model. (otherwise current sentence does not make sense)

Line 21: Warfvinge & Sverdrup reference should be 1992a not b in first mention

PAGE 3921 Line 5: Hettelingh

Line 20-24: remove extra commas after [SO4*]0 (3 times)

PAGE 3924 Line 4 remove extra comma

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