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

## Anonymous Referee #2

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General Comments:

This paper makes an important contribution to the ever evolving acid rain literature and is definitely within the scope of HESS. Its evaluation of the limitations of the F-factor is clear and for the most part complete. The results are well presented and support the paper's central conclusions. The paper is a pleasure to read – clear, concise, generally properly referenced, and the language is almost flawless (there are a few wordy bits). I recommend publication in HESS.

As a widely recognized environmental issue within Europe and North America, "acid rain" has been with us since the 1970s. The scientific rationale behind political action to reduce European and Canadian emissions of acidifying pollutants has been largely

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based on surface water or forest soil critical loads and exceedances. Much of the early credit for this must go to A. Henriksen (Norway) who developed the Steady-State Water Chemistry Model (SSWC) in the 1980s – a conceptually and computationally simple model that relates an aquatic ecosystem's critical load to the weathering rate of its drainage basin expressed in terms of the base cation flux. Simplicity was achieved through adoption of the F-factor which "corrects" the weathering flux of base cations for that proportion of the flux originating from soil leaching. In Henriksen's view of the time, the F-factor should have a value between zero (no soil exchange contribution) and one (all base cations from the soil).

Computationally, the F-factor was the ratio of the change in surface water base cations (relative to pre-industrial conditions) to the change in acid anions, but because little or no information is available for pre-industrial conditions, it has been estimated from contemporary water chemistry information through a number of empirical equations as discussed in the current paper. Using modelled time-series data (1800 to 2100) for Swedish catchments, this paper presents a comprehensive comparison of the different computed values for F and how they very with time, shows how the assumptions that underlay the F-factor's original conceptualization fail under current conditions of acidification recovery, and point out the implications for continued use of the SSWC in emission reduction policy. Lest the reader thinks this completely refutes the past use of the SSWC to instruct policy development, this paper points out that during the time of its introduction and use to set emission reduction targets in the 1980s and 1990s, the different equations for calculating F gave similar values and hence similar critical load and exceedance values.

## Specific Comments:

1) The entire paper depends on using simulated time series data for 0.5 m depth soil water chemistry produced by the biogeochemical model SAFE. The authors offer this as "a plausible time series of annual runoff chemistry". I applaud the author's direct acknowledgement that "use of the SAFE model does not provide us with the real truth". Nevertheless, the "plausibility" of this simulated data should be better supported by comparing them to measured data or at least supplying citations to papers that have demonstrated the goodness-of-fit between SAFE output and measured runoff chemistry. In addition, the version of SAFE used to generate the test data set did not consider SO4 adsorption/desorption in the soils. The authors should note what implications this may have.

2) Equation (1) is the simplest possible, albeit most commonly used conceptual formulation for F. Henriksen originally noted that the denominator contains the change in acid anions, but in a time when ecosystems acidification was progressing almost entirely in response to sulphate deposition (i.e., the 1980s), he quickly simplified it to the  $\Delta$ SO4 term. The authors should at least acknowledge the existence of a  $\Delta$ NO3 term. and given the rapidly changing DOC levels observed in many European and North American surface waters, the possibility of a heretofore unexplored term for changing organic anion.

3) Contemporary records of base cation deposition refute the assumption needed to specify Equation (4). The authors should briefly note the implication.

4) Page 3927, line 7: There is no Posch et al. (1993) in the reference list.

5) Section 4.1: I ask the authors to include a short paragraph that discusses what it means to have F-factor values >1. Clearly, something else is influencing  $\Delta$ BC beyond  $\Delta$ SO4. What are the implications.

6) Page 3934, lines 8 and 9: my immediate response to these two lines was "huh?". On further reflection I think I get it, but a more complete development of this point would be useful.

7) Page 3934, line 13: reference to Equation (9) cannot be correct; presumably is should say Equation (11).

8) Figure 6 (and discussion in Section 4.3): Why has FAB (rather than the SSWC)

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been used to calculate critical load exceedances? There is an equation for the present day (S+N) exceedance calculated using the SSWC critical load value, i.e.,  $Ex(S+N) = S^{*}dep + Nleaching - CL$  where Nleaching is the product of the lake NO3 concentration and regional runoff. If Figure 6 is to stand, additions to and extra explanations in the text will be required to deal with the use of FAB.

9) References: Is the journal name in the Curtis et al. citation correct?

Interactive comment on Hydrol. Earth Syst. Sci. Discuss., 6, 3917, 2009.