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1 Pedon-scale silicate weathering: comparison of the PROFILE model
2 and the depletion method at 16 forest sites in Sweden

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13

14 **Abstract**

15 Weathering of soil minerals is important for the recovery from acidification and for the
16 sustainability of forestry. However, there is still substantial uncertainty about its absolute rate.
17 This study presents a harmonized comparison of field weathering rates estimated with the
18 mechanistic model PROFILE and the depletion method for 16 intensively sampled soil
19 profiles across Sweden representing different site conditions. In general, a correspondence in
20 total weathering rates was found between the two methods except in rare cases where either
21 method yielded deviating results. The weathering rate was higher according to the depletion
22 method than according to PROFILE for Mg, while PROFILE produced higher weathering
23 rates for the other base cations. The Spearman rank correlation (ρ) between the two methods
24 indicated significant correlation for Ca ($\rho=0.44$, $p=0.04$) and non-significant correlation for
25 Mg ($\rho=0.51$, $p=0.09$), Na ($\rho=0.25$, $p=0.34$), K ($\rho=0.07$, $p=0.80$), and the sum of the base
26 cations ($\rho=0.11$, $p=0.67$). The variation in weathering rates with depth showed opposite
27 gradients in the upper 50 cm, which reflects the conceptual differences between the methods.
28 This study shows the potential of using multiple methods to identify a probable weathering
29 rate, if harmonized input data is used. Furthermore, it highlights the importance of making
30 comparisons for individual elements in order to interpret differences between methods.
31 Regardless of the method used, weathering rates were below or at the same level as the losses
32 caused by whole-tree harvesting, particularly in southern Sweden, indicating a risk of
33 negative effects on soils and waters.

34

35 **1. Introduction**

36 Chemical weathering is important for the nutrient sustainability of forests, the neutralization
37 of acidifying compounds and the quality of water transported from upland soils to surface
38 water systems downstream. The historical deposition of acid compounds, which culminated
39 around 1980 (Schöpp et al., 2003), caused substantial leaching of base cations from soils to
40 waters, leading to base cation depletion and acidification of soils. A reduction of acid
41 deposition to levels below the total of base cation weathering and base cation deposition is a
42 prerequisite for the recovery of soils from acidification. However, recovery will also depend
43 on the export of nutrients from the forest ecosystem through harvesting, which has increased
44 recently owing to the increased focus on using harvest residues for bioenergy, e.g. whole-tree
45 harvesting. While active measures, such as ash recycling, are recommended in e.g. Sweden
46 (Swedish Forest Agency, 2008) a basis for such activities is an assessment of the mass
47 balance of input and output of mineral nutrients in the ecosystem. Thus, there is a demand for
48 robust estimates of the release rate of mineral nutrients by weathering for predictions of future
49 recovery from acidification and for optimizing forest management policies.

50 Proposed methods for determining weathering rates include the mass balance approach using
51 experimental data (Bain et al., 1994), the mass balance approach using catchment modelling
52 (Cosby et al., 2001), the depletion method using an immobile element as an internal standard
53 (Brimhall & Dietrich, 1987; Olsson & Melkerud, 1989), and the process oriented model
54 PROFILE where weathering rates are estimated for soil pedons (Sverdrup & Warfvinge,
55 1993). The various methods are conceptually different and are intended for different spatial
56 scales; from the soil pedon at site level to the entire soil deposit at the catchment level.
57 Furthermore, the estimates consider different time perspectives; from long-term historical
58 averages to present day weathering and steady state weathering.

59 Several attempts have been made to quantify weathering rate uncertainties through an
60 ensemble approach (Kolka et al., 1996; Starr et al. 1998; Sverdrup et al., 1998; Whitfield et al.
61 2006; Koseva et al., 2010; Klaminder et al., 2011; Futter et al., 2012). Sverdrup et al. (1998)
62 compared rates from six different approaches in two catchments in Gårdsjön, Sweden. The
63 variation in base cation weathering rates (Ca, Mg, Na and K) was 30-39 and 47-81 meq m⁻² y⁻¹
64 in the two catchments respectively, indicating relatively low uncertainties. On the other
65 hand, Klaminder et al. (2011) demonstrated a wide span for Ca and K weathering rates, 7-150
66 meq m⁻² y⁻¹ and 2-32 meq m⁻² y⁻¹ respectively, at a site in northern Sweden where ten studies
67 were compared, and concluded that estimated weathering rates were too uncertain to be used
68 in assessments of sustainable harvesting. They revised their conclusions slightly in Futter et
69 al. (2012), where they pointed out that much of the variation was related to input data and
70 known conceptual differences between the methods, for example that different estimations
71 were valid for different soil depths. They concluded that at least three independent estimates
72 should be used when making management decisions. This is in line with the conclusions by
73 Whitfield et al. (2006) who compared weathering estimates from five approaches in five

74 catchments in Canada. The three soil profile-based approaches gave similar results, with low
75 weathering rates ranging from 3 to 13 meq m⁻² y⁻¹. However, the two catchment-based
76 methods gave one order of magnitude higher rates. Koseva et al. (2010) compared PROFILE
77 weathering estimated with catchment mass balance calculations for 19 sites. The PROFILE
78 weathering rates were in most cases within the range of the catchment mass balance
79 weathering rates, but PROFILE weathering rates were generally lower, as expected when
80 comparing soil pedon weathering rates with catchment weathering rates. Although they did
81 not present any direct uncertainty measures based on this, they used it to demonstrate the
82 reliability of PROFILE. A critical aspect when comparing weathering estimates is the quality
83 of the input data, boundary conditions and assumptions made, e.g. the maximum soil depth
84 considered and corrections for coarse fragments and organic matter. The stone and boulder
85 content may amount to >50%_{vol} in many forest soils (Stendahl et al., 2009) and the way it is
86 treated will have a strong influence on estimated pedon-scale weathering rates. It is crucial
87 that comparisons of weathering estimation methods are harmonized with regard to
88 assumptions and input data, and they should be carefully evaluated when published results
89 from different studies are synthesized.

90 In this study we estimated the base cation weathering at the pedon-scale by the depletion
91 method and PROFILE for 16 intensively sampled soil profiles representing a wide range of
92 soil conditions in Sweden. Both methods have been frequently used to estimate weathering
93 rates on local, regional and national scale for assessments of sustainable harvesting (Olsson et
94 al., 1993; Sverdrup & Rosén, 1998; Akselsson et al., 2007). The depletion method quantifies
95 the loss of mobile elements since the parent material was deposited, whereas PROFILE
96 estimates the release of elements due to the dissolution of soil minerals at steady state. Hence,
97 the two methods consider very different time perspectives. Despite the different time
98 perspectives of the methods the comparison was justified by the relatively young age of the
99 profiles (10 000 – 16 000 years). The methods were applied in a harmonized way using input
100 data from the same forest sites and the same pits, as well as using the same assumptions.
101 Furthermore, the estimated weathering rates were compared with estimated base cation losses
102 at whole-tree harvesting on a selection of the sites, in the same way as in e.g. Olsson et al.
103 (1993) and Klaminder et al. (2011). This simplified mass balance calculation puts the
104 difference in weathering rates between the two methods in a sustainability perspective. The
105 objectives of this study were to: (i) compare how the two methods ranked the sites with
106 regards to Ca, Mg, K, and Na weathering, (ii) compare how weathering intensity vary with
107 depth in the uppermost soil profile for the two methods, (iii) investigate the causes for
108 deviating results between the two methods, and (iv) put the results in a sustainability
109 perspective by comparing the estimated weathering rates with base cation losses associated
110 with whole-tree harvesting.

111 The comparison can shed light on how the total weathering rate and weathering intensity
112 within the soil profile evolve over time and can be seen as a robustness test of the two
113 methods.

114

115 **2. Material and Methods**

116 *2.1. Sites and soil profile data*

117 The 16 sites, which are part of the NORDSOIL database (Raulund-Rasmussen & Callesen,
118 1999), are located at latitudes 56-68 °N across Sweden on podzolised glacial till without
119 stratigraphic layering (Fig. 1, Table 1). All sites are located above the highest Quaternary
120 shoreline and it was assumed that no redistribution of soil material has occurred since
121 deglaciation. The parent material is of granitic composition with varying mineralogical
122 composition. Among the sites 12 were used previously in weathering studies by Olsson &
123 Melkerud (1989, 1991, 2000) and Olsson et al. (1993). For each site, a profile was used that
124 had complete soil chemical and physical data necessary to apply the two methods. Sampling
125 was made at 10 cm depth intervals from the top of the mineral soil to the maximum mineral
126 soil depth (42-263 cm, Table 1). Volumetric samples were taken by a core sampler for each
127 10 cm layer in the A, B and uppermost C horizon and bulk density was determined after
128 drying and weighing. Grain size distribution data was determined for 10 cm layers, although it
129 was missing for the A horizons and ca 25% of the other layers. In those cases the grain size
130 distribution was estimated based on the layers above and below. For each site the stone
131 content was estimated based on the soil texture class to 10-40%_{vol}. An analysis of the total
132 geochemical content of the bulk soil material <2 mm was made for the samples from each 10
133 cm layer throughout the profile by extracting a representative subsample (20 g) that was
134 homogenized by dry grinding in an agate mortar after which 0.125 g was fused with 0.375 g
135 lithium metaborate (LiBO₂) at 1000 °C and dissolved in nitric acid. The content of major
136 elements and trace elements was quantified by plasma emission spectrometry analysis (ICP-
137 AES). The analytical data were normalized with respect to dry matter weight.

138

139 *2.2. The depletion method*

140 The basic principle of the method is the long-term depletion of mobile (weatherable) elements
141 and the concurrent enrichment of immobile (inert) elements in weathered soil layers during
142 soil development. The concentration of the mobile and the immobile element in a weathered
143 and an unweathered soil layer are utilized to estimate the loss of the mobile element in the
144 weathered layer. The idea to use immobile elements to estimate element losses was first
145 proposed by Marshall & Haseman (1942) and the theoretical framework was formalized by
146 Brimhall & Dietrich (1987) and Brimhall et al. (1991). Their framework focuses on the
147 relative mass loss (or gain) of elements from layers in the original soil pedon and they define
148 the fractional mass loss as:

149
$$\tau_{j,w} = \frac{C_{j,w} C_{i,p}}{C_{j,p} C_{i,w}} - 1 \quad (1)$$

150 where C denote concentrations (%), i denotes the immobile element, j denotes the mobile
 151 element, w denotes a weathered horizon, and p denotes the unweathered parent material.
 152 Values of $\tau < 0$ indicates net mass loss, $\tau > 0$ net mass gain, and $\tau = 0$ no change.

153 The weathered amount of the mobile element from a soil layer (g m^{-2}) can be expressed based
 154 on the same principles (Olsson & Melkerud 1989):

155
$$W_{j,w} = \frac{d_w \rho_w}{100} \left(C_{j,p} \left(\frac{C_{i,w}}{C_{i,p}} \right) - C_{j,w} \right) \quad (2)$$

156 where d is layer depth (m), and ρ is bulk density (g m^{-3}).

157 Zirconium, which is predominantly found in the weathering resistant mineral zircon (ZrSiO_4),
 158 was used in this study as the immobile element, since its redistribution within the profile is
 159 negligible (Hodson, 2002). The depletion method assumes that there is no weathering of the
 160 immobile element, that there is no weathering below a certain soil depth, and that the soil
 161 profile was developed from homogenous regolith. One critical aspect of the method is to
 162 define a reference soil layer to represent the unweathered parent material.

163 In this study we used equation (2) to estimate the loss of mobile base cations. The reference
 164 soil layer used (Table 2) was the uppermost layer that met the following criteria: (1) it was
 165 located in the soil C horizon, (2) the ratio between the mobile element and Zr was stable
 166 below this layer, and (3) the Zr gradient was stable below this layer. The long-term
 167 weathering rate was calculated by dividing the total weathered amount by the soil age, i.e. the
 168 age since deglaciation given by the National Atlas of Sweden (Fredén, 2009; Table 1).

169

170 2.3. The PROFILE model and input data preparation

171 PROFILE is a steady state soil chemistry model, originally developed to calculate the effect
 172 of acid rain on soil chemistry (Sverdrup & Warfvinge, 1993). PROFILE includes process
 173 oriented descriptions for chemical weathering of minerals, leaching and accumulation of
 174 dissolved chemical components and solution equilibrium reactions. The weathering module is
 175 a central part of the model. Weathering rates are calculated using transition state theory, based
 176 on the geochemical properties of the soil system, climate, deposition and tree uptake. Soil
 177 physical properties, such as soil mineralogy, soil moisture content, bulk density and exposed
 178 mineral surface area, have been identified as the input parameters with the highest influence on
 179 the output in several studies (Jönsson et al., 1995; Hodson et al., 1996; Zak et al., 1997).

180 In PROFILE, the soil is divided into soil layers with different properties, preferably based on
181 the naturally occurring soil horizons. Soil chemistry and weathering rates are calculated for
182 each layer, and the soil chemistry of each layer is affected by the layer above, through vertical
183 water flow.

184 The estimated soil chemistry and weathering rates describe the situation at steady state, i.e.
185 the soil chemistry and weathering rates that will eventually evolve, with the deposition, tree
186 uptake, climate conditions and soil properties that are given as input to the model (Sverdrup &
187 Warfvinge, 1993). Thus, the model ignores development over time, e.g. changes in soil
188 properties due to weathering.

189 Detailed soil data was available for the chosen profiles from the NORDSOIL database and
190 was used in the input data preparation for the PROFILE model runs. Total elemental content
191 for the different soil layers was used, along with information about predominant minerals and
192 their compositions compiled in previous studies (Warfvinge & Sverdrup, 1995; Akselsson et
193 al., 2004), for calculating soil mineralogy with the A2M model (Posch & Kurz, 2007). The
194 modeled mineralogical composition for the sites is given in Table 3. The specific surface area
195 of minerals, i.e. the weatherable area, was calculated using an empirical algorithm (Warfvinge
196 & Sverdrup, 1995; Holmqvist et al., 2002) using data on the grain size distribution and bulk
197 densities for each layer (data from 50 cm given in Table 1). Data on gravel content were only
198 available for a few of the sites, for the rest of the sites the gravel content was estimated based
199 on grain size distribution (Table 1) and sieving curves (SGU, 1994). The estimated specific
200 surface area was corrected for the stone volume in the soil by subtracting the stone volume
201 from the soil volume, assuming that the weathering from stones is negligible due to the low
202 specific surface area as compared with fine soil. Soil moisture class was given for each site
203 according to a classification system with seven classes (Table 1). Soil moisture was allocated
204 to the soil moisture classes, according to methods in Warfvinge & Sverdrup (1995).
205 Concentration of DOC, CO₂ pressure and Al solubility constants for different layers were
206 derived from Martinson et al. (2003).

207 Information on tree species and mean annual production from the NORDSOIL database
208 (Table 1) was used for estimating net uptake according to methods described in Akselsson et
209 al. (2007), using base cation concentrations in stem biomass compiled in Akselsson (2005).
210 Mean annual temperature and precipitation was derived from the NORDSOIL database (Table
211 1), whereas runoff was derived from national maps from the Swedish Meteorological and
212 Hydrological Institute (SMHI) (Raab & Vedin, 1995). Deposition values were derived from
213 modeled deposition values for 1998 by the MATCH model (Langner et al., 1996).

214

215 *2.4. Comparison with base cation losses at whole-tree harvesting*

216 The base cation losses were estimated for stem (log) only and whole-tree harvesting for the
217 eight spruce sites. The calculations were based on the mean annual stem biomass production

218 for each site (Table 1) and the amount of stems, branches and needles was estimated using
219 biomass expansion functions (Marklund, 1988), which are based on empirical relationships
220 between the diameter at breast height and empirical biomass data for a large number of trees
221 in Sweden. The amount of stems, branches and needles was multiplied with concentrations of
222 Ca, Mg, K and Na in the different tree compartments compiled in Akselsson (2005). Stem
223 harvesting was defined as removal of the stem only, whereas whole-tree harvesting was
224 defined as the harvest of the stem together with 60% of the branches in thinnings and in final
225 felling (Swedish Forest Agency, 2008). Furthermore, it was assumed that 75% of the needles
226 on 60 % of the branches, i.e. in total 45% of the needles, were removed. The methodology is
227 described in more detail in Akselsson et al. (2007).

228

229 3. Results

230 The comparison of the total base cation weathering rates in the upper 50 cm of the mineral
231 soil showed significantly ($p=0.006$, t-test) higher rates for PROFILE (with an interval of 12-
232 $135 \text{ meq m}^{-2} \text{ yr}^{-1}$ and a median of $39 \text{ meq m}^{-2} \text{ yr}^{-1}$) than for the depletion method (with a
233 corresponding interval of 8-41 $\text{meq m}^{-2} \text{ yr}^{-1}$ and a median of $19 \text{ meq m}^{-2} \text{ yr}^{-1}$). There were
234 substantial deviations in estimated weathering rates for three sites. The rate estimated by the
235 PROFILE model was extremely high in Nunasvaara for Ca ($35 \text{ meq m}^{-2} \text{ yr}^{-1}$), Mg (40 meq m^{-2}
236 yr^{-1}) and Na ($51 \text{ meq m}^{-2} \text{ yr}^{-1}$); 5 times higher than the rates derived from the depletion
237 method. In Målaskog the PROFILE-modeled Ca weathering rate was also high (31 meq m^{-2}
238 yr^{-1}) compared to the depletion method ($6.6 \text{ meq m}^{-2} \text{ yr}^{-1}$). In Vålberget it was the other way
239 around, with high rates of Ca ($21 \text{ meq m}^{-2} \text{ yr}^{-1}$) and Mg ($13 \text{ meq m}^{-2} \text{ yr}^{-1}$) weathering from the
240 depletion method compared to PROFILE (4.8 and $2.3 \text{ meq m}^{-2} \text{ yr}^{-1}$ for Ca and Mg
241 respectively).

242 For Ca and Mg there was correlation between the results from the two methods (Fig. 2). The
243 weathering of Mg was generally higher according to the depletion method, whereas it was the
244 other way around for Ca. For K and Na there was weak or no correlation between the two
245 methods. PROFILE gave generally higher weathering rates for Na, whereas there was no
246 such difference for K. The Spearman rank correlation between the depletion method and
247 PROFILE (Table 4) indicated significant correlation for Ca ($\rho=0.44$, $p=0.04$), while it was
248 non-significant for Mg ($\rho=0.51$, $p=0.09$), Na ($\rho=0.25$, $p=0.34$) and K ($\rho=0.07$, $p=0.80$). For
249 the sum of the base cations the correlation was non-significant ($\rho=0.11$, $p=0.67$). The
250 correlation coefficients for Ca and Mg were strongly influenced by the three extreme sites
251 where either method gave highly deviating results. Disregarding these sites resulted in highly
252 significant correlations for Ca ($\rho=0.71$, $p=0.007$) and Mg ($\rho=0.82$, $p=0.001$). For Ca and Na
253 weathering estimated with the same method there was a highly significant correlation; $\rho=0.80$
254 ($p<0.001$) for PROFILE and $\rho=0.73$ ($p=0.001$) for the depletion method.

255 The average mass loss estimated by the depletion method (Fig. 3) increased towards the
256 surface in the investigated profiles, but the gradients varied for the different base cations. The
257 largest average mass loss in the uppermost mineral soil was found for Mg (66%), followed by
258 Ca (43%), Na (38%) and K (29%) as compared to the reference soil layers. The gradient in
259 weathering rate with depth was the opposite for the two methods (Fig. 4). The depletion
260 method, reflecting the average rates since the last glaciation, showed high rates in the upper
261 layer, and decreasing rates with depth, reaching ca 12 meq m⁻³ yr⁻¹ at the 50 cm level. The
262 PROFILE results showed, on the other hand, an increasing weathering rate with depth,
263 reaching ca 110 meq m⁻³ yr⁻¹ at the 50 cm level.

264 The calculation of harvest losses of base cations showed that harvesting of branches and
265 needles almost doubled the losses compared with stem harvest only (Fig. 5). The comparison
266 between harvest losses and weathering rates showed that the losses at whole-tree harvesting
267 were of the same order of magnitude as PROFILE modeled weathering rates for the three
268 northern sites, whereas the weathering rates from the depletion method were about half the
269 size. For the southern sites, the harvest losses at whole-tree harvesting were substantially
270 higher than the weathering rates from either approach.

271

272 **4. Discussion**

273 *4.1. Comparison between results and concepts*

274 The weathering rates estimated by the two methods agreed in some respects, but not in others
275 and the observed differences may be due to a number of factors. The depletion method gives
276 the average weathering rate since the deglaciation, but the historical development of the
277 weathering in the soil is not fully known. Steady state weathering, on the other hand, is a
278 theoretical concept used in PROFILE, which is based on steady state calculations where the
279 input data are kept constant at today's levels, and the steady state soil chemistry and
280 weathering are estimated under the assumption that the present minerals are not depleted. The
281 difference in time perspective for the two methods makes it crucial to consider the historical
282 development of the weathering rate in the interpretation of the results. The influence of soil
283 age on the weathering rate has been investigated in chronosequence studies (Bain et al., 1993;
284 Taylor & Blum, 1995; White et al., 1996; Hodson & Langan, 1999) and by modelling
285 (Warfvinge et al., 1995). The results show high initial weathering rates followed by a decline,
286 which is most pronounced in the first centuries following soil exposure, but the results varies
287 greatly between sites. The decline is caused by the depletion of easily weathered minerals and
288 decrease in reactive mineral surface area (Taylor & Blum, 1995). Attempts have been made to
289 correct the long-term weathering rate to today's levels (Taylor & Blum, 1995) using a power-
290 law function fitted to depletion weathering rates from chronosequence data. These results
291 together with simulations of historical weathering rates (Warfvinge et al., 1995) indicate that

292 the ratio between today's weathering rate and the long-term historical weathering rate is
293 approximately 0.3-0.5.

294 Based on this, the depletion method was expected to give higher values than the PROFILE
295 model. For Mg our results agreed with the theory; the depletion method gave higher
296 weathering rates than PROFILE. However, for Ca, Na and K the PROFILE model gave
297 higher values than the depletion method, although for K there was large non-systematic
298 variation between the two approaches for many sites. Thus, the results for Ca, Na and K
299 contradict the theory about decreasing weathering rates over time. A contributing factor to our
300 observed differences between PROFILE and the depletion method is that the parent material
301 for our profiles is relatively poor in easily weathered minerals that may give rise to such
302 extreme initial weathering rates reported elsewhere (Bain et al., 1993; Taylor & Blum, 1995).
303 Furthermore, the soil profiles were developed in glacial till that partly consists of soil material
304 that was newly formed during the Weichselian glaciation and partly includes redistributed old
305 till deposits from previous glaciations. Consequently, the parent material in the investigated
306 profiles was not completely unweathered at the time of deglaciation. The relatively elevated
307 acid deposition at present could also contribute to increased weathering (April et al. 1986), as
308 well as the increased forest growth and harvest levels during the last century, leading to
309 increased acidification and removal of weathering products. It is, however, difficult to design
310 experiments aiming at quantifying these effects. A sensitivity analysis of PROFILE (Hodson
311 et al., 1996) showed that increased sulphur deposition led to increased or unchanged
312 weathering rates for most minerals, but the effect was generally relatively small.

313 The correlation analysis for Ca and Mg showed that the coefficients were negatively
314 influenced by three sites where one of the two methods yielded highly deviating results. As
315 discussed below, there are good reasons to believe that these deviating results were caused by
316 conditions that caused either model to fail. Hence, the result indicates significant correlations
317 for Ca and Mg weathering when conditions are suitable for both methods. For K and Na the
318 correlation was weak and it could be hypothesized that K and Na weathering in PROFILE
319 was more sensitive to variation in other input data than mineralogy than for Ca and Mg. This
320 means that uncertainties in other input data would have a greater impact on the results. For K,
321 this is to some extent supported by Hodson et al. (1996), who identified K-feldspar, one of the
322 main sources of K, as a mineral particularly sensitive to input variations. To be able to fully
323 explain the difference, even more in-depth studies are required, including an accurate
324 determination of the actual minerals present. The Ca and Na weathering rates estimated with
325 the same method were significantly correlated for both methods. This indicates a proportional
326 release of Ca and Na that could be explained by the occurrence of both elements in the same
327 minerals e.g. the plagioclases, which are rather weatherable and very common in the
328 investigated area. The correlation study showed very different results for the different base
329 cations, which highlights the importance to compare individual base cations rather than
330 merely the sum of them.

331 Uncertainties related to the specific surface area are generally one of the main sources of
332 uncertainties to the mineral weathering rates estimated with PROFILE. This can be due to
333 lack of detailed data on grain size distribution, and due to the generalized equations
334 transforming grain size distribution to specific surface area. Hodson et al. (1998) questioned
335 the equation for estimating specific surface area from grain size distributions, based on a
336 study in Scotland. However, since the equation was developed based on data from Sweden,
337 the applicability can be expected to be higher in Sweden. Hodson and Langan (1999)
338 highlighted the fact that changes over time of specific surface area and mineral reactivity are
339 not taken into account in the PROFILE model, leading to uncertainties in the estimated
340 weathering rates. The uncertainties introduced by the data on specific surface area can be
341 reduced by using detailed input data on grain size distribution and densities for several layers,
342 as was done in this study. However, although detailed data is used, the uncertainties will be
343 substantial, due to the problems discussed above.

344 The depletion method showed very large mass loss of base cations in the upper part of the soil
345 profile considering the relatively young age of the investigated soils (ca 10 000 – 16 000
346 years). Particularly the loss of Mg was considerable. It has been recognized that Mg is one of
347 the most mobile elements in similar soils (Bain et al., 1993; Lång, 1995; Land et al., 1999;
348 Olsson & Melkerud, 2000). In Land et al. (1999) they found a depletion level of 70% for Mg
349 in the E horizon of Podzol soils in northern Sweden. The Mg loss is probably associated with
350 depletion of easily weathered minerals such as chlorite and hornblende, which are important
351 Mg-bearing minerals in the soils of the Svecofennian granitic bedrock and important for forest
352 soil productivity (Stendahl et al., 2002). The high depletion levels for all base cations indicate
353 a general loss of easily weathered minerals.

354 Important drivers for the weathering include processes at the soil surface such as biological
355 production and deposition of acid compounds, which explain the pattern with higher historical
356 weathering intensity towards the surface. However, there is an on-going depletion of easily
357 weathered minerals over time in the uppermost part of the soil, which limits the weathering
358 rate towards the surface. The weathering intensity is gradually shifted towards lower depths,
359 where there are more easily weathered minerals present. Since less weathering agents (i.e.
360 organic acids, HCO_3^-) are being consumed in the uppermost part of the soil, they are
361 transported down in the profile and contribute to the weathering at lower depths. In our
362 results, the steady-state weathering rate estimated with PROFILE exceeds the historical rates
363 estimated by the depletion method in deeper soil layers (Fig. 4).

364

365 *4.2. Applicability of the two approaches for different conditions*

366 The three sites that were outliers in the comparison can be useful to analyze critical conditions
367 for the two approaches. In Nunasvaara the PROFILE weathering rates of Ca, Mg and Na were
368 much higher than for any of the other sites, and also several times higher than the weathering

369 rates estimated with the depletion method. The depletion method indicated moderate losses
370 despite the seemingly good mineralogy indicated by the base cation content. The high
371 modeled weathering rates by PROFILE can be explained by a combination of high contents of
372 minerals rich in Ca, Mg and Na in the soil, e.g. plagioclase, hornblende, biotite and augite
373 (Table 3), high soil density and a high fraction of fine-grained material (Table 1). A possible
374 explanation to the deviation between the methods could be that the simplified handling of
375 hydrology in PROFILE does not take into account that the high soil density may lead to
376 saturation of the soil and thus substantially increased product inhibition, slowing down
377 weathering rates. In Vålberget the situation was the other way around; the depletion method
378 gave many times higher weathering rates for Ca and Mg than PROFILE. The Ca, Na and K
379 weathering rates in Vålberget were the lowest of the 16 sites according to PROFILE and the
380 Mg weathering rate was among the lowest. The depletion method gave by far the highest
381 weathering rates of the 16 sites for Ca and Mg, whereas the K weathering was one of the
382 lowest and the Na weathering was intermediate compared to the other sites. The fractional
383 mass loss of this site was also very large; on average 55% of the Ca and 44% of the Mg has
384 been lost down to 50 cm depth, while in the uppermost layer the result showed that as much
385 as 81% of the Ca and 87% of the Mg has been lost by weathering. An indication that the Ca
386 weathering rate in Vålberget could be problematic was that the site deviated completely from
387 the otherwise strong relationship between Ca and Na weathering estimated by the depletion
388 method. This indicate that the systematic Ca gradient in the profile was caused by
389 heterogeneities rather than soil development and that the weathering rate thus was
390 overestimated. In Målaskog the PROFILE model resulted in much higher weathering rates
391 than the depletion method for Ca and Na. Although the soil in Målaskog was relatively
392 coarse, its high Ca content led to the second highest Ca weathering rate according to
393 PROFILE. The soil profile in Målaskog showed opposite gradients both for mobile and
394 immobile ions indicating vertical heterogeneity in parent material composition, which means
395 that the method assumptions for the depletion method were not fulfilled.

396 *4.3. The results in a sustainability perspective*

397 For detailed sustainability assessments the complete mass balance has to be taken into
398 account, i.e. base cation deposition, weathering, harvest losses and leaching (Sverdrup et al.,
399 2006; Akselsson et al., 2007). Base cation deposition in Sweden is in the same order of
400 magnitude as weathering rates (Akselsson et al., 2007), but the input of base cations through
401 weathering and deposition needs to be substantially higher than the harvest losses, to avoid
402 soil acidification. However, simplified mass balance including only weathering and harvest
403 losses is often used as an indication of the sustainability (Olsson et al., 1993; Klaminder et al.,
404 2011). If weathering rates are substantially higher than base cation removal at whole-tree
405 harvesting, the harvesting effects on the nutrient budget can be expected to be small.
406 However, if the weathering rates are in the same order of magnitude or lower than the base
407 cation removal, there is an obvious risk of negative effects on the soil nutrient status. Our
408 results show that the removal at whole-tree harvesting were in the same order of magnitude or

409 higher level than the weathering rate, no matter which of the approaches that was used. For
410 the sites in southern Sweden, the losses at harvesting were substantially larger than the
411 weathering rates. This area also contains the most acidic soils, due to high historical levels of
412 acid deposition (Pihl Karlsson et al., 2011). Thus, the risk of negative effects associated with
413 base cation losses at harvesting is largest in these regions. The northern sites have better soil
414 status to start with from an acidification point of view, and the losses at whole tree harvesting
415 are smaller. These areas are associated with lower risks, but since the losses at harvesting are
416 as big as the weathering rates, and since the base cation deposition is relatively low in
417 northern Sweden, there is not much left for the runoff water, which may lead to negative
418 effects for surface waters on a long term.

419 **5. Conclusions**

420 Where conditions are suitable for both methods there is a correspondence in weathering rates
421 between the depletion method and PROFILE. For Mg, the depletion method yield higher rates
422 than PROFILE, probably due to strong depletion of main Mg-bearing minerals, such as
423 chlorite and hornblende. For Ca, K and Na the weathering rates are generally higher
424 according to PROFILE than according to the depletion method. This indicates that initial
425 weathering rates of these elements at the time of soil formation were less extreme in the
426 investigated sites compared to other regions. The weathering rates in the upper 50 cm
427 decrease with depth according to the depletion method, but increase with depth according to
428 PROFILE. The pattern can be explained by the methods difference in time perspective;
429 historically most of the weathering has taken place in the upper soil, which is now depleted of
430 easily weathered minerals leading to a lower current rate as represented by PROFILE. The
431 application of multiple methods on a large selection of sites can help identify deviating results
432 for the methods included. The losses from whole-tree harvesting exceed weathering for both
433 the depletion method and PROFILE, especially for sites in southern Sweden.

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575 **Figure captions**

576 Fig. 1. Location of the 16 sampling sites.

577 Fig. 2. Weathering rate ($\text{meq m}^{-2} \text{ yr}^{-1}$) for Ca, Mg, K and Na estimated by the PROFILE and
578 the depletion method for 16 soil profiles.

579 Fig. 3. The fractional mass loss of Ca, Mg, K and Na at different soil depths (cm) estimated
580 by the depletion method. The error bars represent the 95% confidence limits.

581 Fig. 4. Weathering intensity ($\text{meq m}^{-3} \text{ yr}^{-1}$) of the sum of base cations vs. soil depth (cm)
582 estimated by the PROFILE and the depletion method. The error bars represent the 95%
583 confidence limits.

584 Fig. 5. Weathering rates of base cations estimated by the two approaches compared with
585 harvest losses of base cations at eight spruce sites. Harvest losses are calculated for a whole-
586 tree harvesting scenarios where all tree stems, 60% of the branches and 75% of the needles on
587 the branches are harvested. The dashed lines in the harvesting bars indicate the level when
588 only tree stems are harvested.

Tables

Table 1. Site characteristics and soil properties at 50 cm depth in the mineral soil for the 16 investigated sites.

Site	Lat.	Long.	MAT (°C)	MAP (mm)	Clay (% _{wt})	Silt (% _{wt})	Sand (% _{wt})	Density ^b (g dm ⁻³)	Soil depth (cm)	Soil age (calendar years)	Soil moisture class ^c	Tree species	Stem volume production (m ³ ha ⁻¹ yr ⁻¹)
Bodafors	57.6	14.7	5.4	729	2	38	60	1400	192	14 000	3	Norway spruce	10.7
Gårdsjön	58.0	12.0	6.6	952	12	33 ^a	55 ^a	1055	72	14 340	3	Norway spruce	9.5
Hjärtasjö	59.0	15.3	5.2	650	5	31 ^a	64 ^a	992	113	11 280	3	Norway spruce	9.5
Hässlen	60.3	16.3	4.3	629	9	41 ^a	50 ^a	1442	130	10 830	3	Other broadleaves	8.4
Kloten	59.9	15.3	4.2	807	2	48	50	1590	125	10 910	4	Scots pine	6.1
Kullarna	62.0	16.6	3.8	552	4	12 ^a	84 ^a	1390	102	10 400	3	Norway spruce	5.2
Lammhult	57.2	14.8	5.5	688	2	35	63	1376	178	14 260	3	Norway spruce	9.7
Målaskog	56.8	14.3	6.4	707	0	23 ^a	77 ^a	1072	175	14 360	3	Other broadleaves	11.3
Nunasvaara	67.8	21.4	-2.0	456	4	43 ^a	53 ^a	1852	227	9 880	3	Scots pine	1.7
Risfallet	60.3	16.2	4.3	629	7	35 ^a	58 ^a	1560	136	10 810	3	Scots pine	7.0
Skånes Värsjö	56.3	13.5	6.4	765	3	25 ^a	72 ^a	1291	112	14 700	3	Norway spruce	11.6
Stöde	62.3	16.4	2.5	707	2	33 ^a	65 ^a	1271	263	10 330	3	Norway spruce	5.5
Svartberget	64.3	19.7	1.0	573	2	26 ^a	72 ^a	1534	217	10 120	4	Norway spruce	3.6
Söderåsen	56.1	13.0	7.2	795	6	24 ^a	70 ^a	1350	126	15 900	3	Beech	12.6
Vindeln	64.2	19.6	1.2	523	4	37	59	1407	207	10 130	3	Scots pine	3.6
Vålberget	60.8	13.8	2.6	701	3	17 ^a	80 ^a	1243	176	10 580	3	Clearcut (spruce)	6.2

^a The fraction 0.02-0.06 mm, which according to common definitions is a part of the silt fraction, was included in sand fraction in the original data for many sites. In this study, the sand and silt fractions were recalculated according to common definitions to be compatible with the PROFILE model. The calculations were based on relationships from the other sites, where complete grain size distributions were available.

^b Volume weight of < 2 mm fraction including coarser dead organic material (volume of live roots, gravel and stones deducted from sampled volume).

^c Drainage class according to the NORDSOIL database: 1. Excessively, 2. Somewhat excessively, 3. Well, 4. Moderately, 5. Imperfectly, 6. Poorly 7. Very poorly.

Table 2. Element concentrations for the < 2 mm soil fraction at the reference layer depth used in the depletion method estimates.

Site	Ref. depth (cm)	Ca (%)	Mg (%)	K (%)	Na (%)	Zr (ppm)
Bodafors	62	1.47	0.65	2.31	2.03	298
Gårdsjön	57	1.60	0.68	2.54	2.29	299
Hjärtasjö	57	1.13	0.88	1.99	1.62	189
Hässlen	82.5	1.19	0.79	3.10	1.72	250
Kloten	48.5	0.94	0.28	2.47	1.76	247
Kullarna	62	1.14	0.58	2.99	1.84	343
Lammhult	98	1.54	0.59	2.34	2.31	232
Målaskog	60.0	2.39	1.07	2.47	2.66	261
Nunasvaara	54.5	2.95	2.04	1.88	3.29	192
Risfallet	76	1.01	0.61	2.55	2.05	181
Skånes Värsjö	62	1.00	0.24	3.10	2.33	466
Stöde	55.5	1.47	0.86	2.32	1.87	229
Svartberget	154	1.37	0.41	2.41	2.35	241
Söderåsen	101	0.75	0.36	2.84	1.8	341
Vindeln	129	1.00	0.92	2.47	1.67	279
Vålberget	117	1.57	0.77	1.36	0.79	186

Table 3. Normative mineralogy at 50 cm depth in the mineral soil based on the A2M model (Qu=Quartz, Or=Orthoclase, Pl=Plagioclase, Ho=Hornblende, Ep=Epidote, Bi=Biotite, Au=Augite, Ap=Apatite, Mu=Muscovite, Ch=Chlorite, Il=Illite, Ve=Vermiculite, An/Ab=ratio between Anorthite and Albite in Plagioclase).

	Mineral contents (%)												
	Qu	Or	Pl	Ho	Ep	Bi	Au	Ap	Mu	Ch	Il	Ve	An/Ab
Bodafors	41.0	13.2	25.8	1.2	3.8	0.0	0.0	0.5	3.6	2.5	1.6	5.5	0.12
Gårdsjön	36.5	4.7	26.4	1.0	3.6	0.0	0.0	0.2	8.4	1.4	12.2	3.5	0.14
Hjärtasjö	49.3	3.0	20.4	0.8	1.9	0.0	0.0	0.2	9.4	1.9	9.1	4.1	0.12
Hässlen	40.2	15.1	21.8	1.0	2.5	0.0	0.0	0.2	6.3	2.1	6.6	3.6	0.13
Kloten	51.5	13.6	21.8	0.5	3.0	0.0	0.0	0.2	3.4	1.0	3.0	1.3	0.10
Kullarna	39.1	15.3	25.6	1.0	2.9	0.0	0.0	0.2	5.5	1.8	4.8	2.5	0.15
Lammhult	37.9	14.6	29.0	1.5	4.2	0.0	0.0	0.4	2.4	2.0	1.2	4.9	0.12
Målaskog	28.5	15.7	31.8	2.6	7.5	0.0	0.0	0.6	2.6	2.7	1.3	5.2	0.11
Nunasvaara	16.7	10.2	36.0	11.1	2.1	2.7	8.1	0.5	0.0	1.3	4.5	4.7	0.05
Risfallet	45.4	13.5	26.2	0.9	2.6	0.0	0.0	0.2	3.8	2.1	3.4	3.2	0.10
Skånes Värsjö	38.8	16.5	29.7	0.7	2.4	0.0	0.0	0.3	4.3	1.0	2.2	1.4	0.10
Stöde	40.5	4.7	23.4	1.9	3.7	0.0	0.0	0.4	9.9	3.6	9.3	2.5	0.13
Svartberget	42.4	15.1	29.1	1.2	2.9	1.6	0.5	0.2	0.0	0.8	4.5	1.9	0.15
Söderåsen	48.9	10.2	21.3	0.5	1.4	0.0	0.0	0.3	9.9	0.7	4.9	0.9	0.08
Vindeln	39.1	9.8	24.3	1.0	2.3	1.2	0.5	0.3	0.0	0.7	17.1	5.4	0.09
Vålberget	66.3	6.4	8.9	2.6	2.5	0.0	0.0	0.3	3.6	1.6	0.8	5.2	0.22

Table 4. Spearman rank correlation coefficients between weathering estimates by PROFILE and the depletion method where Σ indicates the sum of base cations (n=16, p-values in italic).

		Depletion method				
		Ca	Mg	K	Na	Σ
PROFILE	Ca	0.44	0.20	0.31	0.59	0.36
		<i>0.09</i>	<i>0.46</i>	<i>0.24</i>	<i>0.02</i>	<i>0.16</i>
	Mg	0.12	0.51	0.23	0.22	0.24
		<i>0.65</i>	<i>0.04</i>	<i>0.40</i>	<i>0.42</i>	<i>0.36</i>
	K	-0.13	-0.01	0.07	0.18	-0.07
		<i>0.63</i>	<i>0.97</i>	<i>0.80</i>	<i>0.50</i>	<i>0.80</i>
	Na	0.05	0.07	0.15	0.25	0.04
		<i>0.85</i>	<i>0.80</i>	<i>0.57</i>	<i>0.34</i>	<i>0.88</i>
	Σ	0.19	0.11	0.14	0.34	0.11
		<i>0.49</i>	<i>0.69</i>	<i>0.62</i>	<i>0.19</i>	<i>0.67</i>

Figures

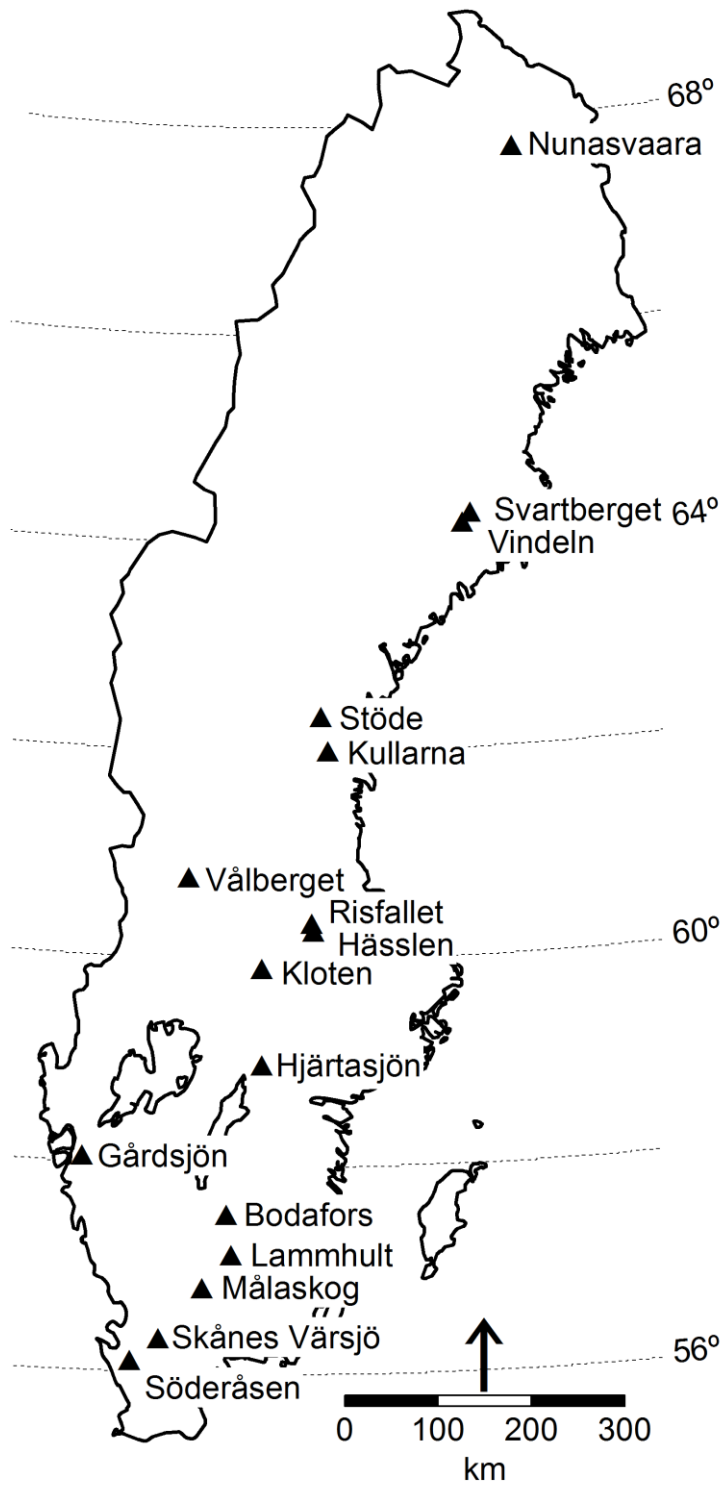


Fig.1

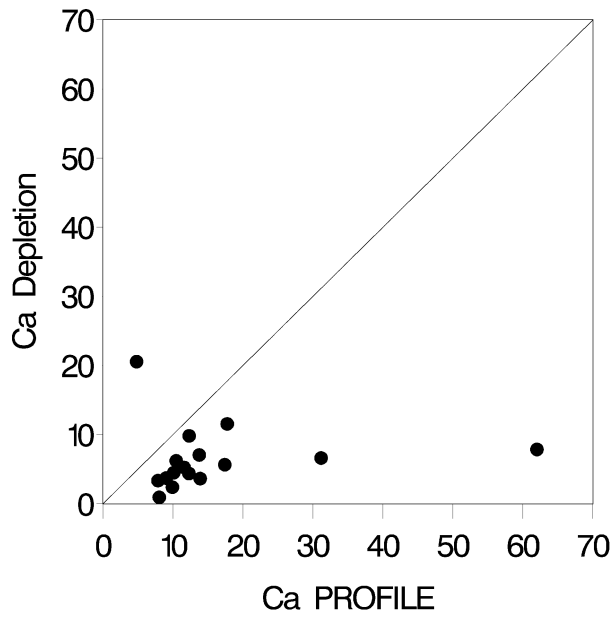


Fig. 2a

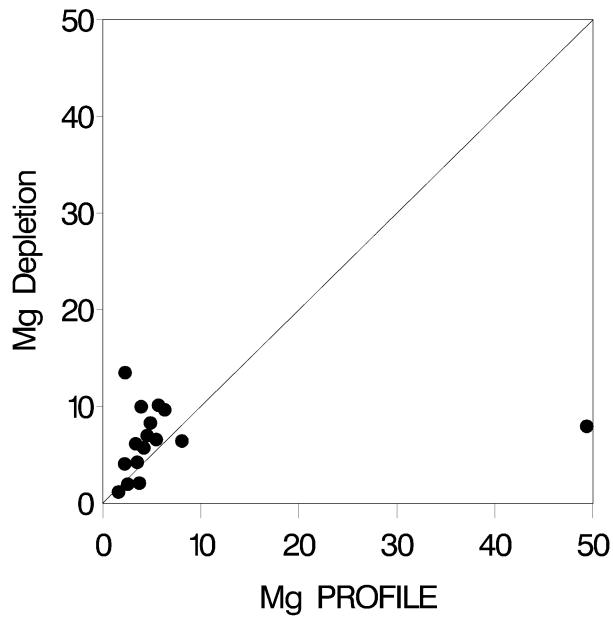


Fig. 2b

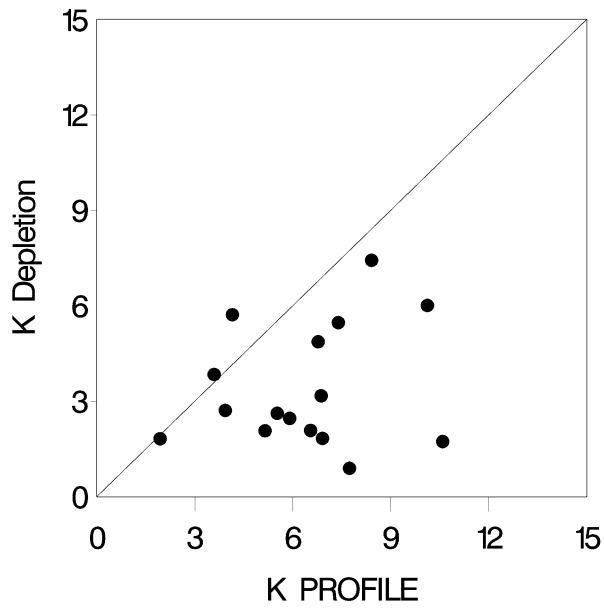


Fig. 2c

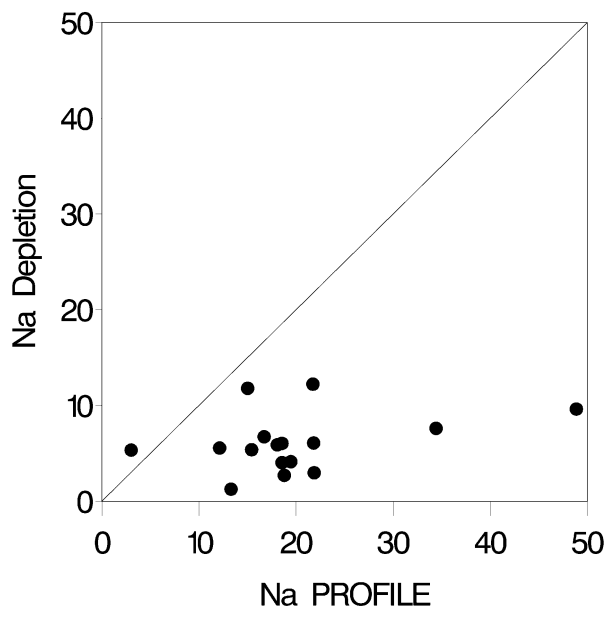


Fig. 2d

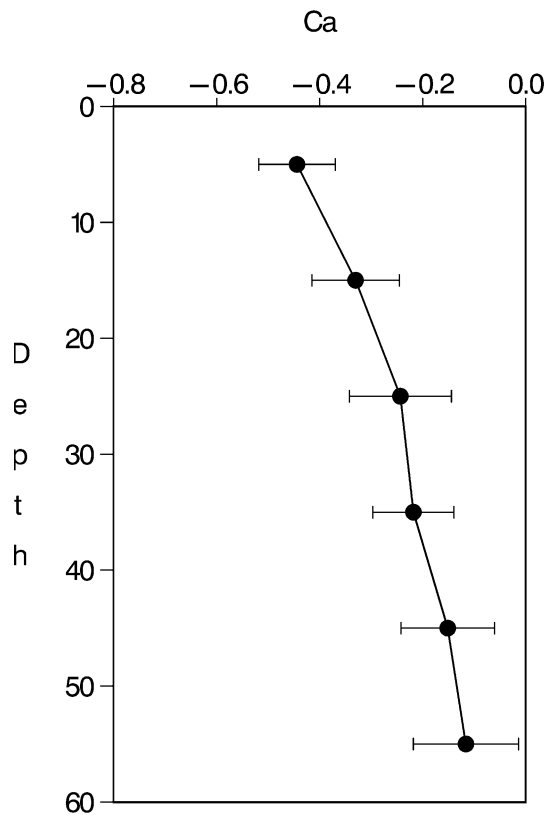


Fig. 3a

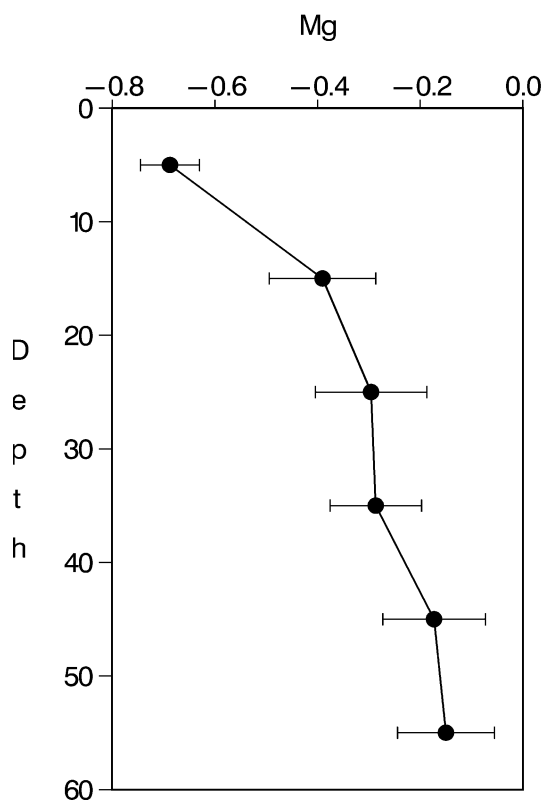


Fig. 3b

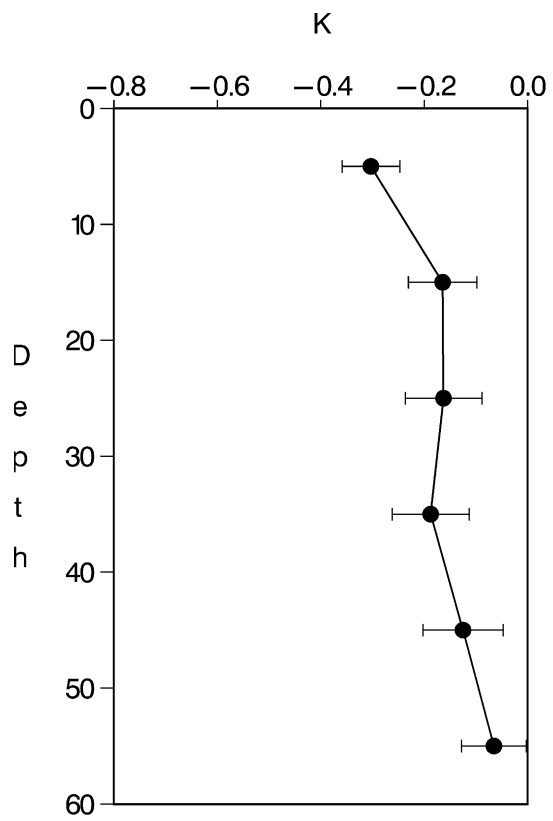


Fig. 3c

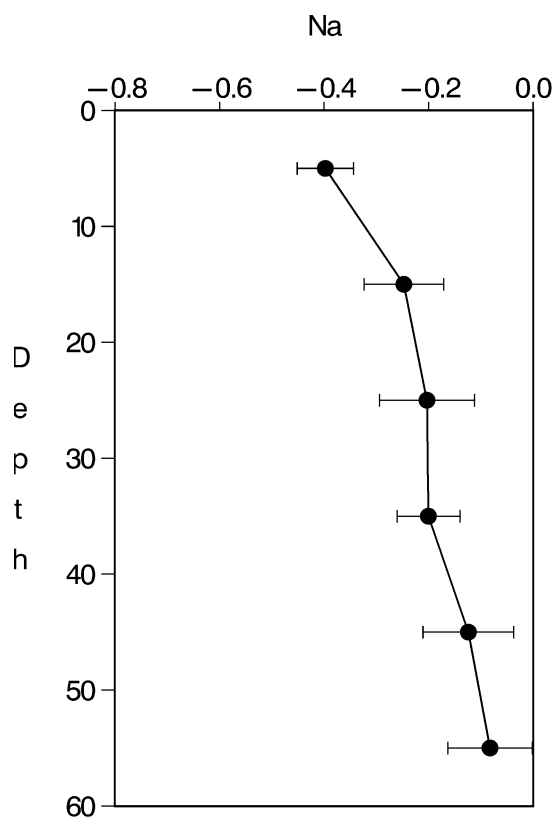


Fig. 3d

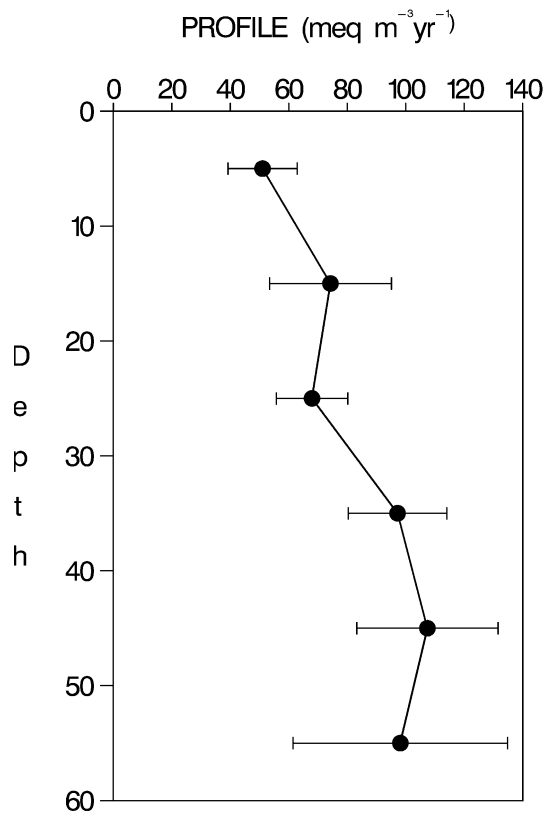


Fig. 4a

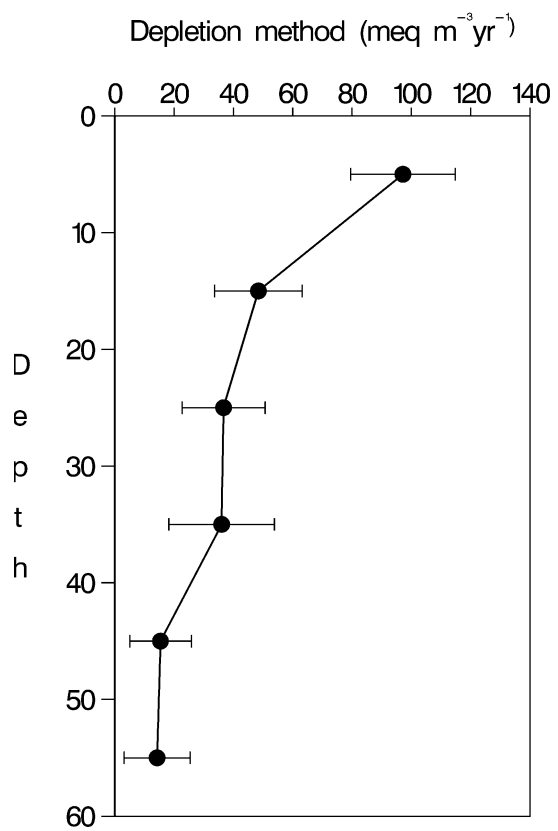


Fig. 4b

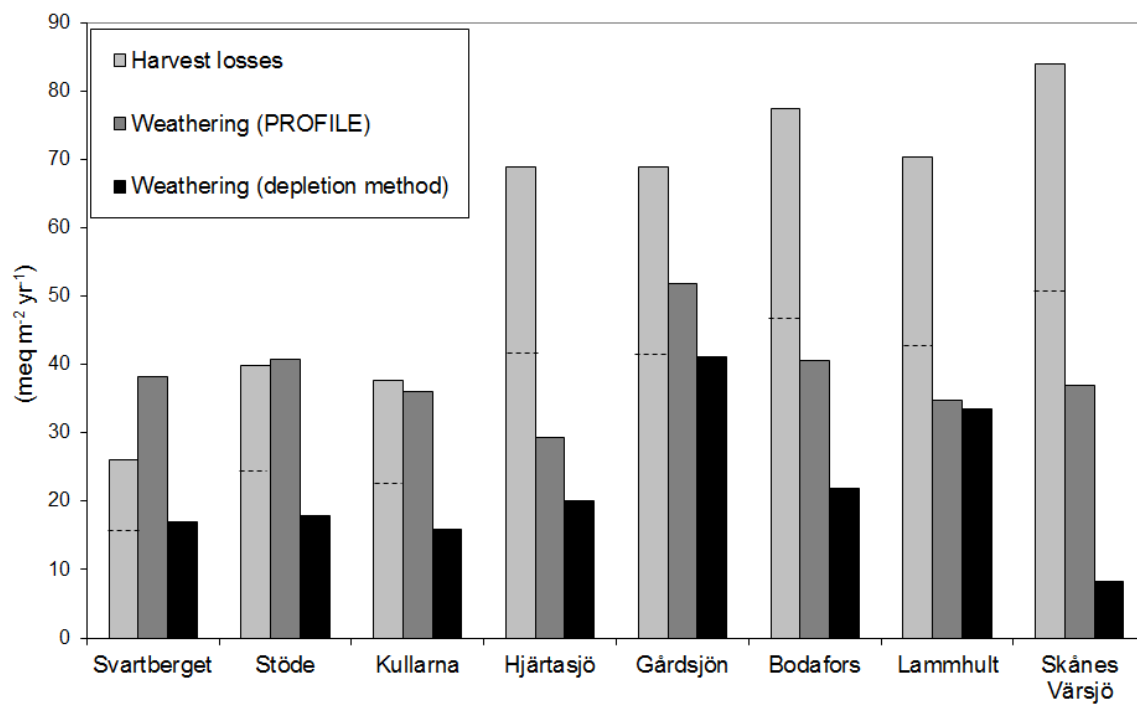


Fig. 5