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Temporal changes and fluxes of sulphur and calcium in wet and dry deposition, internal circulation as well as in run-off and soil in a forest at Gårdsjön, Sweden

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Abstract. Sulphur dioxide in air as well as dry deposition of sulphur dioxide to a forest has decreased by a factor of 20 during the last two decades. It was earlier found that the internal circulation of calcium in Norway spruce follows the dry deposition of sulphur dioxide. The sulphur and calcium fluxes from 1992 were calculated from wet deposition, throughfall and a surrogate surface. Earlier fluxes from 1981 to 1991 were calculated using assumptions of the dry deposition of non-marine sulphate and calcium. The new estimates confirm the earlier studies that the internal circulation of calcium in a coniferous forest is directly related to the dry deposition of sulphur dioxide to the canopies and that the internal circulation of calcium decreases at the same rate as the deposition of sulphur decreased during the last two decades. The deposition fluxes were also compared to run-off and uptake of calcium in the forest as well as on modelled weathering rates from severely acidified forested catchments near the coast and inland and the soil pool. A reconstruction of changes in the soil pool of calcium over the last 100 years indicate that the soil pool has decreased by ca. 70% in catchments with complete harvest of the forest and ca. 40% if branches and needles are left on the ground. In a natural forest without acid deposition the soil pool of calcium would have increased by 6%. Estimates for the 21st century show that harvesting of stems + branches and needles may almost empty the soil pool of calcium in the next 100 years. Increased nitrogen deposition has increased forest growth, which indirectly increased uptake of calcium by the trees and indirectly caused a further decrease of the soil pool of calcium.

Introduction

Forested catchments are excellent tools to study the effects of atmospheric deposition and forest management practices on base cations in biomass, wet and dry deposition, soils and run-off water. Biogeochemical studies at Hubbard Brook Experimental Forest (Likens et al. 1998) showed that calcium had declined in atmospheric input, stream output and soils between 1963 and 1992. Falkengren-Grerup and Tyler (1991) found that the exchangeable pools of calcium in Swedish forest topsoils have declined between 1949 and 1989. The soil depletion is the result of leaching due to acid deposition during the past 50 years, to decreased atmospheric input of calcium and to changes of net storage of calcium in biomass. Johnsson (1992), Ragsdale et al. (1992) and Currie et al. (1996) showed that biotic flux of base cations are far greater than geologic and atmospheric input as well as output by run-off. The accumulation of base cations in biomass corresponded to a production of acidity which together with acid deposition may result in a depletion of base on the soil exchange complex (Currie et al.

1994). Also harvesting of forests has resulted in soil depletion of calcium (Reynolds and Stevens 1988; Hornbeck et al. 1990).

At the Lake Gårdsjön watershed, a coniferous forested catchment on the Swedish West Coast, wet deposition, throughfall and run-off of acidifying, neutralising and eutrophying species have been measured continuously since 1980/81 (Hultberg et al. 1983; Hultberg 1985; Hultberg and Grennfelt 1992). These studies aim to quantify chemical effects of acid air pollutants to forested ecosystems to allow comparisons with other studies elsewhere in the world (Hultberg and Likens 1992; Hultberg et al. 1994). Dry deposition and internal circulation of Norway spruce was estimated using a surrogate surface since May 1992 (Ferm and Hultberg 1995; Hultberg and Ferm 1995; Ferm and Hultberg 1999). This earlier work has been the basis for the presented results.

Experimental

The measurements were carried out on a forested catchment at the Gårdjön Experimental Area ca. 15 km from the open Swedish West Coast (58.07°N, 12.02°E, and altitude 120 m). The forest consists of ca. 13 m tall and 100-year-old Norway spruce. The catchment (F1) is 3.7 ha and has 653 stems per hectare.

Net throughfall (NTF), dry deposition (DD) and internal circulation (IC) of calcium and sulphur from May 1992 to December 2000 was calculated using throughfall, wet deposition and a surrogate surface (Ferm and Hultberg 1995, Hultberg and Ferm 1995; Ferm and Hultberg 1999). During this period the non-marine (DD Ca^{2+} nm) and marine (DD Ca^{2+} m) dry depositions of calcium were 2.8 and 3.0 mmol m^{-2} $year^{-1}$, respectively. Between January 1981 and April 1992 the dry deposition of non-marine calcium was set equal to the dry deposition of marine calcium (average 3.1 mmol m^{-2} $year^{-1}$ during this period). The internal circulation of calcium was then estimated from IC Ca^{2+} = NTF Ca^{2+} − DD Ca^{2+} m − DD Ca^{2+} nm. The assumption that the dry deposition of marine and non-marine calcium were equal has a small effect on the calculated internal circulation of calcium since the estimated non-marine calcium was only 13% of the net throughfall of calcium during this period (1981–1992). April 1992 and earlier, the non-marine sulphate deposition was estimated from the measured particulate sulphate concentrations in air using a deposition velocity of 7.3 mm/s.

This deposition velocity was equal to the velocity obtained between 1992 and 2000 using the surrogate surface (for DD SO_4^{2-} nm) and the air concentration. The dry deposition of SO_2 (uptake) was subsequently obtained from NTF SO_4^{2-} minus the marine and non-marine particulate sulphate. The calculated non-marine fraction of sulphate was 24% of the net throughfall of sulphate.

Results and discussion

The estimated dry deposition of SO_2 is plotted together with the measured SO_2 concentration in Figure 1. The SO_2 concentration have decreased from ca. 180 to

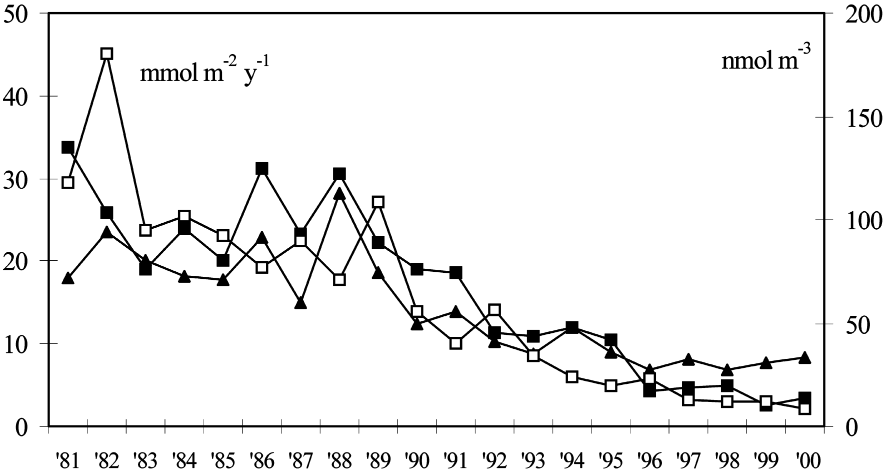

Figure 1. Dry deposition of SO_2 (filled squares), SO_2 concentration in air (open squares) and internal circulation of calcium (triangles) as a function of time.

9 nmol m^{-3}, that is, a factor of 20 during the last two decades or 16% per year. The estimated annual dry deposition of SO_2 was well correlated with the SO_2 concentration in air ($r^2 = 0.74$ with one outlier removed).

The average SO_2 deposition velocity was calculated to 10.0 mm/s. The particulate sulphate concentrations only decreased by 4.8% per year or a factor of 2.6 (from 46 to 18 nmol m^{-3}) during the same period. The internal circulation of calcium is plotted in the same Figure.

The new calculated internal circulation of calcium for the years 1981–1991 is also plotted against the dry deposition of SO_2 in a scatter plot in Figure 2 together with 1992 to 2000 fluxes based on results using a surrogate surface (Ferm and Hultberg 1995; Ferm and Hultberg 1999). The slope of the regression line is 0.58 and the intercept 4.6 mmol m^{-2} year^{-1}. This confirms the earlier observation that the internal circulation of calcium co-variates with the SO_2 deposition and that the two fluxes also are similar in magnitude (Hultberg and Ferm 1995).

The loss of exchangeable calcium from the forest soil can be calculated from: calcium in run-off − (wet + dry deposition) + growth − weathering. Table 1 shows the mean annual fluxes during 1981–2000. The fluxes of sulphur and sodium are also shown. Sodium fluxes were used to estimate the marine fractions and to check the accuracy of the measurements. The net loss of exchangeable calcium was on the average 10 mmol m^{-2} year^{-1} during 1981–2000.

The output of calcium from the catchment by run-off is strongly linked to the sulphur deposition. Hultberg et al. (1990) found that addition of acidic sulphate to a catchment in the Lake Gårdsjön research area caused an increased loss of calcium and aluminium (inorganic) as well as small amounts of magnesium and hydrogen ions in run-off.

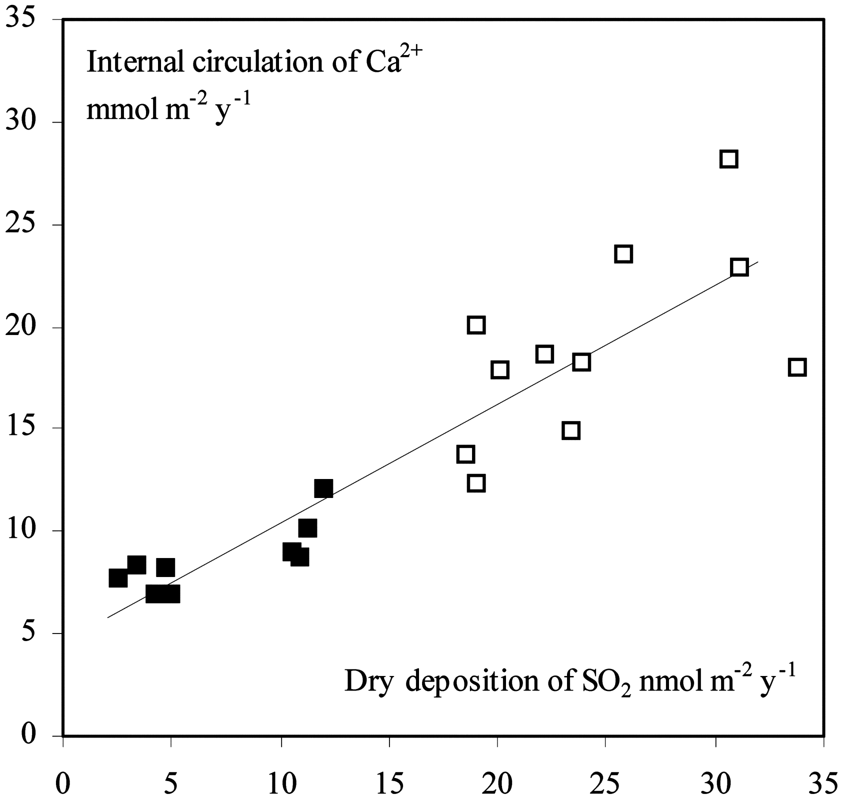

Figure 2. Internal circulation of calcium as a function of dry deposition of sulphur dioxide. Solid squares represent data calculated using the surrogate surface.

Table 1. Fluxes in mmol m^{-2} $year^{-1}$ during 1981–2000 and storage in mmol m^{-2} (around 1990). The storage of S only represents the adsorbed sulphate. The total sulphur content in the soil pool is 8300 mmol m^{-2}.

	Ca	S	Na	References
Wet deposition (m)	1.5	4.2	70	
Wet deposition (nm)	3.3	27.6		
Dry deposition (m)	3.1	8.4	140	
Dry deposition (nm)	2.8	23.7		
Weathering	10.5	–	15.8	Sverdrup et al. (1998); Hultberg (1985)
Σ Supply	21.1	63.9	226	
Run-off	17	71	227	
Growth	14.2	2	0.9	Rosén (1988); Hultberg (1985)
Σ Removed	31	73	227	
Gain/loss	−10	−9	−2	
Storage*	1030	620	460	Andersson et al. (1998)

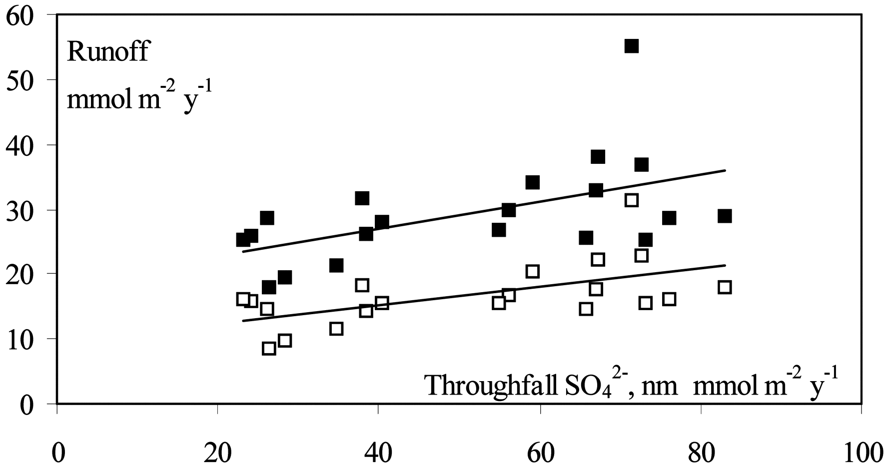

Figure 3. Run-off of calcium (open squares, used for calculations between 1970–1980) and calcium + inorganic aluminium (filled squares, used for calculations between 1900–1970) as a function of atmospheric sulphur deposition (throughfall). Fluxes are yearly averages from 1981 to 2000.

In another experiment dolomite was applied to a forest soil and soil chemistry and run-off was measured. The output of sulphate was measured, but not mentioned in the article (Hultberg et al. 1995) because no effect of liming on the sulphate run-off could be observed. A correlation between the atmospheric non-marine deposition of sulphate and the calcium in run-off is shown in Figure 3 by a scatter plot of annual output fluxes of calcium and calcium + aluminum in catchment F1 versus the throughfall of non-marine sulphate during 1981–2000.

If the two fluxes are plotted as a function of time instead, no time delay between the decreasing sulphate deposition and the decreased calcium in run-off can be observed.

The exchangeable calcium store has decreased by 50–60% in forests in the southern parts of Sweden during the 20th century (Gunnar Borg, personal communication). Falkengren-Grerup and Tyler (1991) analysed the exchangeable pools of calcium in Swedish forest topsoils between 1949 and 1989 and found a 50% reduction. To see if such a large decrease of the exchangeable calcium store can be explained by fluxes derived from this investigation, the following calculation was made.

In an earlier study the sulphur deposition to the EMEP grid containing the Gårdsjön research area was estimated between 1900 and 1994 (Ferm and Hultberg 1998). The estimation was made from published numbers on coal and oil consumption in different countries in Europe and its sulphur content. The average ratio between a country's sulphur emission and its contribution to this grid (according to the EMEP model) during 1979–87 was then used to estimate the sulphur deposition to this grid. The estimated deposition was very close to the measured wet deposition at Gårdsjön. The throughfall deposition of sulphur to this forest was a factor 2.1 higher than the modelled average deposition to the grid. There are many uncertainties in this calculation and the grid contains areas with lower dry deposition than a forest, such as lakes and meadows. The factor 2.1 was therefore used to estimate the sulphur deposition and the equation for the regression line in Figure 3 was used to integrate the run-off of calcium between 1970 and 1980. Between 1900 and 1970 the regression line for calcium + aluminium in Figure 3 was used, because the soil was less acid and there was a negligible loss of aluminium. A total loss from the soil by run-off of 2.2 mol m^{-2} of calcium was obtained for these 81 years.

If the non-marine calcium deposition during 1900–1980 was half of that between 1981–2000 (because the trees were smaller), the net loss of exchangeable calcium from the soil would be 1260 mmol m^{-2} during that period. The loss of exchangeable calcium during the 20th century would then be 67%. If the trees are not harvested, but allowed to die the calcium loss would be 23%. Further from the coast, where the deposition of marine calcium is negligible, the calcium loss would be larger (72%). Changes in run-off losses of calcium after a clear-cut has not been considered here, because such measurements have been carried out at Gårdsjön Experimental Area in 1983, but no important changes in run-off of calcium was observed. There are many uncertainties in the calculations and it is hard to estimate the accuracy in the exchangeable amount of calcium in 1900. The calculations, however, give the same magnitude of loss of exchangeable calcium as the earlier mentioned soil studies.

Conclusions

The long term monitoring of wet and dry deposition at the coniferous forested catchment in the Lake Gårdsjön watershed shows that the trees respond to sulphur dioxide deposition to the tree canopies by neutralising the formed acidic sulphate by releasing calcium at the same magnitude. Over the last two decades decreased deposition of sulphur dioxide resulting from decreased emissions from all European countries has resulted in a proportional reduction in dry deposition of sulphur dioxide as well as in the internal circulation of calcium by the trees.

The total deposition of anthropogenic sulphur has resulted in severe acidification of lakes, running waters, soil water and groundwater as well as of the soils in acid sensitive regions of Europe. Our calculations of the effects on the soil storage of exchangeable calcium are based on monitoring of fluxes and estimates of fluxes and taking into account atmospheric deposition of nonmarine and marine sulphur and calcium input as well as tree growth, catchment output and weathering.

Figure 4 shows that ca. 70% of the total soil pool of exchangeable calcium has been lost during the last 100 years of acid sulphur deposition in forests with tree harvesting of stems. Twenty three per cent of the soil pool of calcium has been lost due to acid deposition in forests with no forestry. Forestry and acid rain are thus of equal importance for the loss of exchangeable calcium in the soil.

Figure 4 also shows that in a natural forest without acid deposition and harvesting of the forest the soil pool of calcium would have increased by 6%.

Estimates of inputs and outputs of calcium for a forest during the 21st century are shown in Figures 5 and 6.

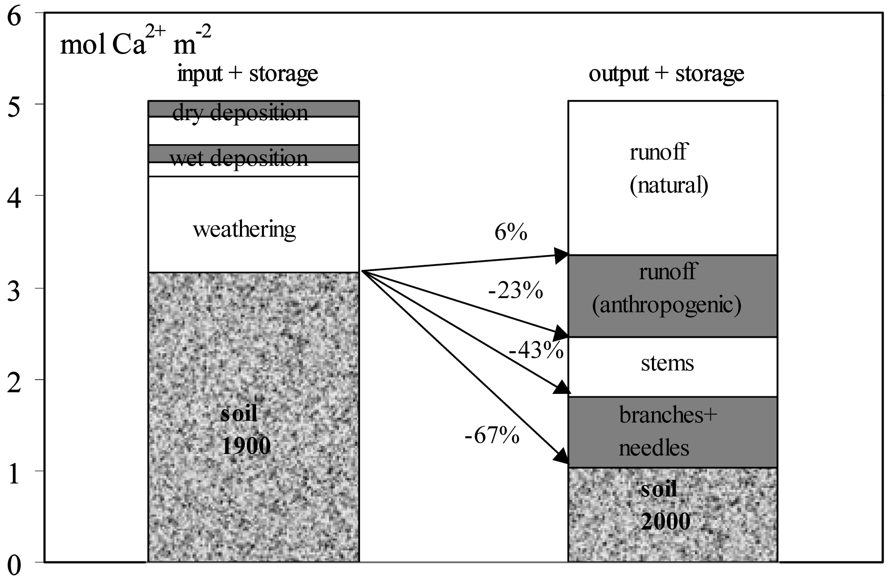

Figure 4. Calcium budget for the 20th century. The white areas of the deposition represent the marine fraction and the dark areas the non-marine fraction.

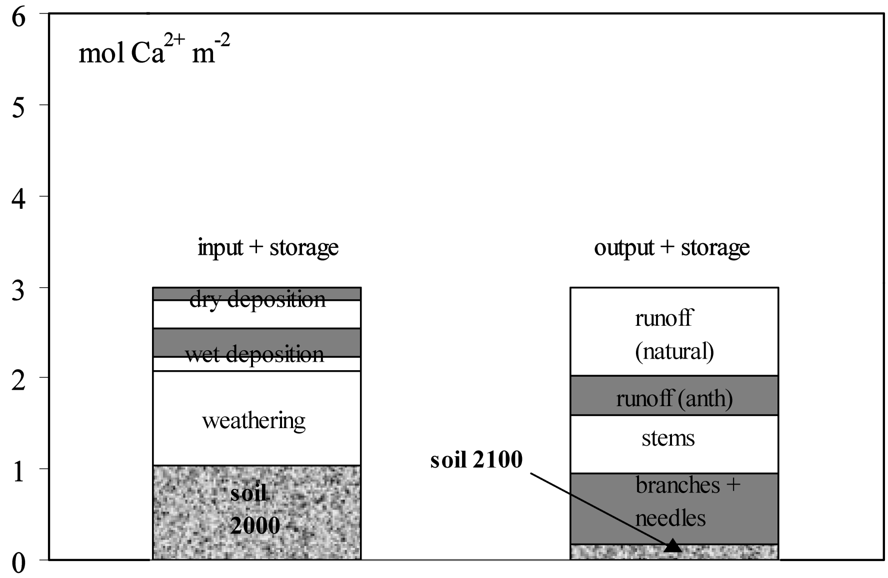

Figure 5. Calcium budget for the 21st century if needles and branches are harvested. The white areas of the deposition represent the marine fraction and the dark areas the non-marine fraction.

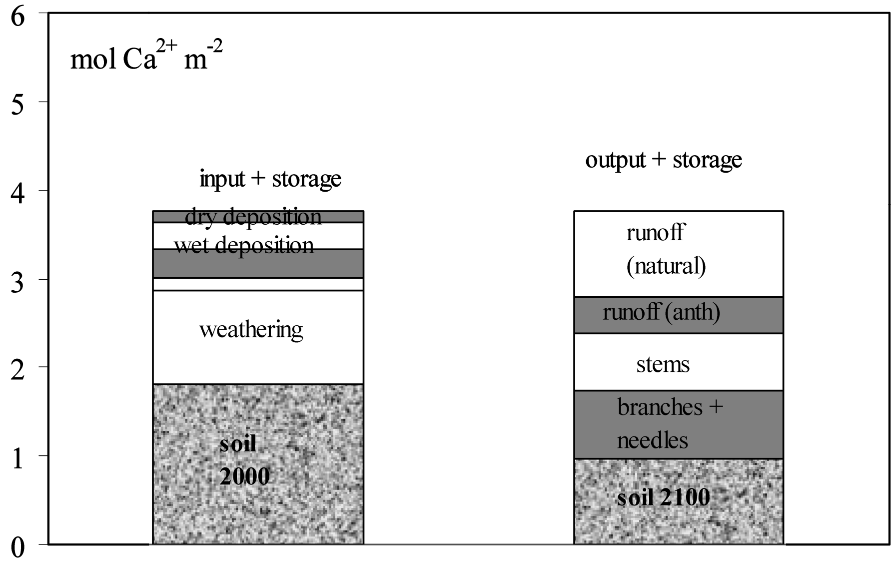

Figure 6. Calcium budget for the 21st century if needles and branches are left in the forest. The white areas of the deposition represent the marine fraction and the dark areas the non-marine fraction.

Figure 5 shows that harvest of stems + branches and needles may almost empty the soil pool of calcium in the next 100 years. Harvest of only the stems as shown in Figure 6 will still decrease the soil pool of calcium but at a slower rate.

Increased nitrogen deposition has increased forest growth during the last 100 years. The increased forest growth has indirectly increased uptake of calcium by the trees, which indirectly has caused a further decrease of the soil pool of calcium. These effects of nitrogen deposition may be an increasing problem in the future unless emissions of nitrogen oxides and ammonia are reduced.

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