

WORKING PAPER

ATMOSPHERIC DEPOSITION OF SULFUR AND BASE CATIONS TO EUROPEAN FORESTS

Wilfried Ivens

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Preface

Forests have a tendency to filter out air pollutants, thereby absorbing larger amounts of dry deposition than an equal area of open ground. Furthermore, although atmospheric deposition everywhere in the industrial world is on the average acidic, it is well known that some precipitation events are in fact alkaline. Since 1983 IIASA's Acid Rain Project has included work on the forest filtering effect and alkaline deposition in connection with the development of the Regional Acidification Information and Simulation (RAINS) model.

Wilfried Ivens from the University of Utrecht, The Netherlands, has prepared this overview which analyzes measurements from forested sites in different parts of Europe. This working paper represents the most detailed examination that has so far been carried out at IIASA of the forest filtering effect and alkaline deposition.

Roderick W. Shaw
Leader
Acid Rain Project

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Abstract

To simulate acidification processes in forests (soils), it is important to know as well as possible the atmospheric input. Large scale models have recently been improved to take better into account the differences in deposition between forests and other surfaces.

In this report measurements of sulfur-fluxes onto the forest floor (54 case studies) are compared with deposition fluxes as calculated by the EMEP-model and by the RAINS modifications on this model. The value of the filtering parameter used in RAINS at this moment is discussed. A new quantitative basis for the filtering effect of different tree species is given.

Fluxes of base cations are compared to sulfur fluxes to quantify the neutralizing effects of base cations. There appears to be no direct proportional relationship between base cation and sulfur fluxes onto the forest floor. It is proposed to study the possibility of linking, within the RAINS model, basic cation deposition with the amount and magnitude of several sources of basic cations.

Table of Contents

1. Introduction	1
2. The problem of estimating forest filtering of S-compounds	1
3. Measurements of fluxes onto the forest floor	4
4. Calculation of sulfur deposition	4
5. Comparison of sulfur fluxes	6
5.1 Difference between the fluxes onto the forest floor and bulk deposition	6
5.2 "Observed" versus "calculated" deposition	7
5.2.1 Observed fluxes versus EMEP model results	7
5.2.2 Observed fluxes versus RAINS model results	7
6. Conclusions on the forest filtering of sulfur regarding RAINS	8
7. Base cation deposition	8
8. Conclusions on base cation calculations of RAINS	10
References	12

Atmospheric Deposition of Sulfur and Base Cations to European Forests

Wilfried Ivens

1. Introduction

IIASA's Acid Rain Project has developed an acidification model, RAINS (Regional Acidification and INformation Simulation), which links atmospheric transport and deposition of air pollutants with the ecological impacts of air pollutants, notably sulfur, on a European scale (Alcamo *et al.*, 1987).

The transport and deposition of S-compounds in the RAINS model are based on the output of the EMEP long range transport model for sulfur compounds (Eliassen and Saltbones, 1983; Lehmhaus *et al.*, 1986). The EMEP-model computes mean overall S-deposition to large areas, including both forested and open areas. The atmospheric sulfur input in forested ecosystems is very important to the impact models incorporated in RAINS. Because the deposition to forests has been assumed to be greater than other areas (the forest filtering effect), some modifications of the EMEP-model results have been made to estimate the specific forest deposition within RAINS (e.g., Kämäri, 1986). Besides this, the deposition of alkaline compounds has to be taken into account within RAINS because of their neutralizing effect on sulfur-driven acidifying processes (e.g., Kauppi *et al.*, 1986). The objectives of this study are to find a new basis for:

1. The quantification of the forest filtering effect with relation to sulfur, and
2. The estimation of deposition of basic cations.

In this study the observed atmospheric deposition in different forest stands around Europe is compared to the calculated deposition. Both the original EMEP estimates and the estimates modified in RAINS are used to obtain the "calculated" deposition.

2. The Problem in Estimating Forest Filtering of S-Compounds

The EMEP-model computes the annual wet and dry S-deposition throughout Europe on grid squares of 150 × 150 km. Estimates of the EMEP-model for the annual mean S-concentrations in air and in precipitation have been compared with measurements (Lehmhaus *et al.*, 1986). The correlation between calculated and observed values was 0.87 for sulfur dioxide. The particulate sulfate concentrations in air and in precipitation were less well predicted, the correlation between predicted and calculated observations being 0.59 and 0.65, respectively. The calculated overall annual mean air concentrations of SO_2 and sulfate were very close to the observed values. However, the model appeared to underestimate the overall annual mean sulfate concentrations in precipitation by about 15%. Similar model valida-

tions have not been done for the deposition estimates. The deposition of S-compounds will depend not only on air and precipitation concentrations, but also on the aerodynamic conditions, and on the physical, chemical and physiological characteristics of the receptor surface (cf. Fowler, 1980). Observations confirm that the deposition velocity of S-compounds depends on the receptor surface (land use), although the range is wide and just a few estimates are available for forests (Table I).

Table I. Deposition velocities of SO_2 and SO_4^{2-} above different categories of land use, $cm \cdot s^{-1}$ (from Voldner *et al.*, 1986).

Land use	SO_4^{2-}	SO_2
Water	0.4 (0.0 - 1.2) n = 7	0.5 (0.16 - 4.0) n = 5
Snow	0.13 (0.04 - <.20) n = 4	0.05 (0.005 - .17) n = 5
Soil + urban	—	0.9 (0.04 - 2.5) n = 9
Grass + crops	0.20 (0.0 - 2.3) n = 39	0.72 (0.2 - 2.6) n = 34
Forest	0.93 (0.50 - 1.45) n = 6	0.4 (0.01 - 1.5) n = 6

The most recent EMEP-model (Lehmhaus *et al.*, 1986) calculates the dry deposition velocity of SO_2 as a function of windspeed and surface roughness (Table II). A "mean" surface roughness is assigned to every grid square. For particulate sulfate, the dry deposition velocity is set to the constant value of 0.1 ms^{-1} for all receptor surfaces, which is lower than the mean values indicated in Table I. Because of the coarse resolution of the model, it is not possible to calculate the deposition at a specific site within the grid. As a result, the model will probably underestimate deposition at sites with higher surface roughness, such as forests.

Table III indicates that forests tend to "filter" sulfur compounds, i.e., increase the dry deposition flux from the air to the soil surface. Between the different kinds of forest, there can be large differences. However, "filtering" will not only depend on tree species. The structure of the stand is probably also of great importance.

A method has been developed in RAINS to modify the output of the EMEP-model in order to obtain realistic estimates for forest deposition (Kauppi *et al.*, 1986). The method is based on the rationale that the sum of forest deposition, α_f , and open land deposition, α_o , equals the total deposition as estimated by the EMEP-model, α_{tot} , in grid element i that is

$$f_i \alpha_{f,i} + (1-f_i) \alpha_{o,i} = \alpha_{tot,i} \quad (1)$$

where f_i is the fraction of forest land in grid element i . Data to describe f for each grid square were collected from the World Forestry Atlas (1975). A coefficient, ϕ , was defined to denote the factor by which open land deposition is multiplied to obtain an estimate for the deposition on a nearby forest stand that is

$$\alpha_f = \phi \alpha_o \quad (2)$$

Table II. Surface roughness assumed in EMEP-model.

<i>Surface type</i>	<i>Surface roughness (m)</i>
Sea	10^{-5}
Desert, snow	10^{-3}
Grass	3×10^{-2}
Countryside	0.25
Suburbia, cities	0.8
Woods	1.0

Source: Kllassen and Saltbones, 1983.

Table III. Total sulfur deposition in forest compared to deposition in adjacent terrain with low vegetation.

<i>Vegetation</i>	<i>Forest</i>			<i>Low Veg.</i>
	<i>Quercus robur</i>	<i>Betula pendula</i>	<i>Pinus sylvestris</i>	<i>Pteridium aquilium</i>
Deposition ($g / m^2 / year$)	4.45	4.21	10.12	1.60
The ratio: $\frac{\text{forest deposition}}{\text{low vegetation deposition}}$	2.8	2.6	6.3	---

Source: Skeffington, 1983.

<i>Vegetation</i>	<i>Forest</i>				<i>Low Veg.</i>
	<i>Quercus robur</i>	<i>Fagus sylvatica</i>	<i>Pinus sylvestris</i>	<i>Picea abies</i>	<i>Calluna vulgaris</i>
Deposition ($g / m^2 / year$)	3.31	5.17	3.53	8.76	1.88
The ratio: $\frac{\text{forest deposition}}{\text{low vegetation deposition}}$	1.8	2.8	1.9	4.7	---

Source: Matzner, 1983.

On this basis it is straightforward to calculate $d_{f,t}$ [= forest deposition per unit of land area in grid square i ($gm^{-2}yr^{-1}$)] as a function of $d_{tot,t}$, f , and φ :

$$d_{f,t} = d_{tot,t} \varphi / [1 + (\varphi - 1)f_t]. \quad (3)$$

Posch *et al.* (1985) demonstrated that d_f is estimated to be very similar to d_{tot} if $1 < \varphi \leq 1.2$ or if $0.7 \leq f < 1.0$ that is, when forest deposition does not differ substantially from the deposition to open land or when forests cover 70 to 100% of the area of the grid square. As an example, RAINS would estimate forest deposition 50% higher than the corresponding EMEP grid average deposition in conditions where φ is 2.0 and the fraction of forest land is 30% (Figure 1).

3. Measurements of Fluxes onto the Forest Floor

The focus of this study was on element fluxes to the forest floor by throughfall and stemflow. Throughfall is the water dripping through the canopy during rainfall and stemflow is the water running along the trunk. These fluxes include both wet and dry deposition onto the tree surface.

A total of 22 publications of experimental studies were screened describing the S-flux at 54 sites, the Ca-flux at 47 sites and the Mg-flux at 38 sites. All measurements were done between 1967 and 1986. The duration of measurements varied from a few months to more than 10 years. If the measurement period was shorter than one year, the fluxes were interpolated to annual flux by multiplying by 365/measurement period (days/days).

Most of the stands (38 cases) were formed by conifers (*Table IV*). Stand age was reported in 29 cases and stand height in 8 cases. For the location of the sites as well as references see *Table V* and *Figure 2*.

Table IV. Tree species in case studies (total = 54).

Sitka spruce	6	Beech sp.	4	Mixed	
Douglas fir	1	Oak sp.	3	Conif./Decid.	2
Norway spruce	22	Birch sp.	3		
Scots pine	9	Maple sp.	1		
		Mixed deciduous	3		
Conifers	38	Deciduous	14		

The aim was to include both throughfall and stemflow fluxes into the "observed" deposition. In 31 cases only the throughfall was given. The stemflow flux was estimated in these cases based on the ratios between stemflow and throughfall fluxes reported elsewhere (Verstraten *et al.*, 1983; van Breemen *et al.*, 1982; Miller *et al.*, 1980; Nihlgård, 1970; Johnson *et al.*, 1986). Stemflow contributions between 0 and 20% were assigned depending on tree species and stand age. Bulk deposition, being the precipitation collected in the open land by means of continuously opened funnels, was available on all sites. The fluxes were corrected for the contribution of sea-salt particles, using sodium and chloride as sea-salt tracers (Asman *et al.*, 1981).

4. Calculation of Sulfur Deposition

To obtain model deposition estimates, sulfur emissions of each European country were computed by means of RAINS-model (Alcamo *et al.*, 1987) during the years of the throughfall and stemflow measurements.

The impact of these emissions on the deposition to the forest sites was computed by means of two methods. First, the deposition estimates were calculated from the average results of runs for the years 1979, 1980, 1983 and 1984 of the most recent EMEP-model (cf. Lehmhaus *et al.*, 1986). A second set of "calculated" deposition estimates was prepared using the RAINS modification of EMEP output given in Eq. (3).

Table V. Description of study sites.

<i>NAME</i>	<i>REF</i>	<i>TIME</i>	<i>LNG</i>	<i>LTT</i>	<i>FC</i>	<i>SP</i>
Königstein	13	83-85	8.28	50.11	45	3
Grebenau	13	83-85	9.29	50.45	49	3
Witzenhausen	13	83-85	9.51	51.20	40	3
Wintersw. 1	34	81-82	6.44	51.58	20	5,6
Wintersw. 2	34	81-82	6.44	51.58	20	1,6
Hackfort	7	81	6.14	52.05	18	6,7
Campina	7	81	5.15	51.35	14	4
Grabbtorp 1	31	74*	18.20	59.40	44	4
Grabbtorp 2	31	75*	18.20	59.40	44	4
Tillingb. 1	33	81	-0.20	51.10	13	6
Tillingb. 2	33	81	-0.20	51.10	13	7
Tillingb. 3	33	81	-0.20	51.10	13	4
Kilmichael	26	75-77	-5.28	56.05	3	3
Leanachan	26	75-77	-4.50	56.50	8	3
Strathyre	26	75-77	-4.19	56.19	5	3
Kershope	26	75-77	-2.50	55.10	5	3
Elibank	26	75-77	-2.50	55.40	5	3
Fetteresso	26	75-77	-2.20	56.55	6	3
Birkenes 1	17	77*	7.10	58.20	0	7
Birkenes 2	17	77*	7.10	58.20	0	3
Birkenes 3	17	77*	7.10	58.20	0	4
Dividal 1	17	77*	19.40	68.45	26	7
Dividal 2	17	77*	19.40	68.45	26	3
Dividal 3	17	77*	19.40	68.45	26	4
Solling 1	5	69-83	9.25	51.45	20	5
Solling 2	5	69-83	9.25	51.45	20	3
Lüneb H. 1	5	80-84	10.00	53.00	12	6
Lüneb H. 2	5	80-84	10.00	53.00	12	4
Gårdsjön	14	80-81	11.30	58.00	9	3
Kongalund 1	27	67-68*	13.10	55.50	19	5
Kongalund 2	27	67-68*	13.10	55.50	19	3
Alptal	3	86*	8.45	47.10	31	3
Lägern	3	86*	8.22	47.29	31	3
Davos	3	86*	9.50	46.48	27	3
Schönbuch 1	8	79-83	9.10	48.30	49	3
Schönbuch 2	8	79-83	9.10	48.30	49	5
Feldberg 1	8	86	8.02	47.51	41	3
Feldberg 2	8	86	8.02	47.51	41	8
Gribskov	11	84*	12.19	55.56	11	3
Jädrås	2	ca. 77	16.23	60.48	68	4
Delamere	32	77-78*	-2.40	53.13	5	4
Waroneu	9	ca. 82	6.00	50.35	24	3,5
RobINETTE	9	ca. 82	6.00	50.35	24	3
Kootwijk	18	85-86	5.46	52.11	5	2
Ispina	21	73-74*	20.13	50.02	30	6,9
Edinburgh	28	79	-3.30	55.50	5	4
Wingst	6	83	9.02	53.43	10	3
Harz	6	83	10.25	51.45	32	3
Hils	6	84-85	9.40	52.00	19	3
Harste	6	82-85	9.50	51.35	20	5
Spanbeck	6	82-85	10.50	51.35	20	5

continued on next page ...

Rouquet	30	68-70	3.40	43.50	6
Oberwarm. 1	15	84-86	11.47	49.59	3
Oberwarm. 2	15	85-86	11.47	49.59	3
Weulfersreuth	15	84-86	11.46	50.04	3

REF = reference, see literature list.

TIME = year(s) of measurement. * = part of the year.

LNG = longitude.

LTT = latitude.

FC = forest coverage in KMEP grid (%).

SP = tree species: 2 = Douglas fir (*Pseudotsuga* spp.); 3 = Spruce (*Picea* spp.);

4 = Pine (*Pinus* spp.); 5 = Beech (*Fagus* spp.); 6 = Oak (*Quercus* spp.);

7 = Birch (*Betula* spp.); 8 = Maple (*Acer* spp.); 9 = Linden (*Tilia* spp.).

5. Comparison of Sulfur Fluxes

5.1. Difference between the fluxes onto the forest floor and bulk deposition

Kämäri (1986) calculated an approximation for the φ -parameter by comparing the measured total stemflow and throughfall flux of sulfur to the forest soil (TD) with the deposition measured on bulk collectors in the open field (bulk deposition = BD):

$$\varphi = TD / BD \quad (5)$$

He proposed an average value of $\varphi = 2$ for whole Europe, which is used at the moment in RAINS. All the 14 case study sites forming the data investigated by Kämäri (1986) were included also in this study. An additional number of 40 sites were found in literature so that the data base of this investigation is somewhat larger and more suitable for a statistical analysis.

The deposition to the forest floor appeared to be significantly related to the bulk deposition (*Figure 3*). The mean φ of all sites was 2.9 ± 1.5 . The φ -value was not equal for all forest types, coniferous forest having the highest φ and deciduous forest the lowest (*Table VI*). There are two reasons why it is somewhat uncertain to approximate the true filtering effect (atmospheric deposition to forest vs. that to a nearby crop or grass field) by Eq. (5). First, part of the flux of sulfur measured in stemflow and throughfall could be due to the internal nutrient cycle of the ecosystem. Lindberg *et al.* (1986) and Bredemeier (1987) argue, however, that in case of S the internal flux is insignificant (< 5%) in conditions found in Central Europe where the total deposition to the canopy is high. Secondly, bulk deposition probably underestimates the deposition to grass and crops. Low vegetation filters dry deposition to some extent and thereby tends to absorb SO_2 and SO_4^{2-} more efficiently than the bulk collector. Deposition on bulk collectors is predominantly comprised of gravitational deposition. A small amount is contributed by capture from the atmosphere through turbulent transfer, impact and diffusion. Little is known about the ratio of bulk deposition to deposition on low vegetation. Skeffington (1983) found about 10% higher S-deposition on grass (*Pteridium aquilinum*) compared to bulk deposition. Heil *et al.* (1988) showed that sulfur deposition to grass is closely related to the leaf area index of the grass. Deposition to unmown grassland appeared to be 3 times bulk deposition in summer and about equal to bulk deposition during the other seasons. Both of the sources of uncertainty act in the

way that Eq. (5) tends to overestimate the true φ , i.e. the filtering of forest as compared with that of grasslands and crops.

Table VI. Ratio between the flux of S onto the forest floor (TD) and bulk deposition (BD).

<i>Forest type</i>	<i>All forests</i>	<i>Coniferous</i>	<i>Deciduous</i>
n	54	38	13
TD/BD	2.89 ± 1.50	3.15 ± 1.61	2.05 ± 0.73

5.2. "Observed" versus "calculated" deposition

5.2.1. Observed fluxes versus EMEP model results

The correlation between the measured fluxes to the forest floor and the deposition calculations done by the EMEP-model was 0.70. This correlation is less than the correlation reported between calculated and measured SO_2 -air concentrations but higher than the correlation between calculated and measured concentrations of sulfate in air and precipitation (Lehmhaus, 1986).

There appeared, however, to be an obvious bias. Measured fluxes generally were higher than model estimates. This bias was not the same for all forest types (Table VII and Figures 4a,b). Coniferous forests had the highest ratio between observed and calculated deposition. Only in coniferous forests was the difference between the flux to the forest floor and the EMEP-model estimate statistically significant (paired t-test, $\alpha = 0.05$).

Table VII. Ratio of observed and calculated total S-deposition.

<i>Forest type</i>	<i>All forests</i>	<i>Coniferous</i>	<i>Deciduous</i>
n	54	38	14
Obs./calc.	1.40 ± 0.72	1.57 ± 0.77	0.98 ± 0.36

5.2.2. Observed fluxes versus RAINS model results

Figure 5 shows the comparison of observed data with RAINS predictions. The value 2.0 was used for φ as usual. The correlation between the observed fluxes and the RAINS estimates was 0.70. The RAINS-estimates were significantly higher than the observed fluxes (paired t-test, $\alpha = 0.05$), indicating that the overall value of the forest filtering parameter of 2.0 is too high.

6. Conclusions on the Forest Filtering of Sulfur Regarding RAINS

It can be concluded that forest filtering is a significant factor determining the fate of atmospheric sulfur deposition. In particular, spruce forests absorb high deposition loads. The total deposition estimates of the EMEP-model agree very well with the deposition measured in deciduous forests, but it underestimates deposition in coniferous forests on the average by 30–40%. Therefore, it is proposed to estimate forest deposition in RAINS applying a value of 1.0 for the forest filtering parameter φ for deciduous forests and a value of 1.6 for coniferous forests. Such a transformation appears to result in an overall mean deposition estimate which equals the overall mean observed throughfall and stemflow flux (*Figure 6*).

The difference between coniferous forests and other forests might be caused by the fact that conifers are green throughout the year and their canopies provide a large receptor surface continuously. Also the specific (micro) structural characteristics of the conifer canopy may play a role, involving a higher aerodynamic surface roughness than other forests. In the next phase, it would be interesting to collect more material of this kind, to subtract wet deposition from total deposition estimates, and to investigate forest filtering specifically related to dry deposition.

In addition, research is needed on the physical and meteorological mechanisms of forest deposition.

7. Base Cation Deposition

As discussed by Kämäri (1986) two rather different approaches have been considered in RAINS on how to take into account the neutralizing effect of base cation deposition. One assumes that base cation deposition is proportional to sulfur deposition. If sulfur emissions are reduced and sulfur deposition decreases, base cation deposition according to this assumption will also decrease in a proportional way. The reduction of sulfur emissions is thus assumed to reduce base cation emissions as well.

The other approach would be to assume that the base cation deposition is independent of the sulfur deposition. This would be correct, if the sources of emissions are not the same for base cations and for sulfur. The estimated neutralizing effect of base cations could then be subtracted from the estimated acidifying effect of sulfur deposition. In *Figures 7a* and *b* the fluxes of Ca and Mg onto the forest floor at several European sites are compared to the flux of sulfur onto the forest floor.

Calcium flux in these data tended to be low at sites where sulfur flux is also low (*Figure 7a*). The relationship seems to be curvilinear, although the wide scatter especially connected to high values of sulfur flux does not allow firm conclusions. Magnesium flux (*Figure 7b*) was rather constant at 40–50 $\text{meqm}^2\text{-yr}$ and thus independent of sulfur flux.

The relationship between calcium and sulfur fluxes is most probably coincidental and does not indicate that calcium and sulfur would originate from the same sources. Power plants, for example, emit considerable amounts of sulfur dioxide but very little calcium compounds. Wind erosion, road dust and agricultural liming practices are sources of calcium emission into the air but are insignificant sources of atmospheric sulfur. The relationship may reflect the simple fact that power plants and other sources of sulfur are located in the same regions as calcium sources (agricultural fields and roads). Both sources are concentrated in

central Europe where population density, industrial development and agricultural production have a stronger effect on the environment than, for example, in Scandinavia.

The magnesium flux data indicated hardly any gradient between industrialized regions and remote areas. This suggests that most of the magnesium falling onto the forest floor has its origin either within the forest stand or in other non-anthropogenic processes.

Possible sources of Ca and Mg are:

1. Soil dust (mainly from agricultural land);
2. Agricultural fertilizers (liming);
3. Road dust (mainly from unpaved roads);
4. Limestone quarries;
5. Burning of fuels containing Ca and Mg;
6. Sea-spray.

To assess the influence of each of these sources on the deposition of basic cations in European forests, these sources should be parameterized. Some possibilities are:

- ad1. Area of agricultural land on calcareous soils (km^2);
- ad2. Consumption of limestone fertilizers (kg);
- ad3. Length of unpaved roads on calcareous soils (km);
- ad4. Number of quarries;
- ad5. Ca + Mg emission (kg), calculated from fuel use;
- ad6. Distance to sea (km).

To estimate the relative importance of these variables, a multiple regression according to the following model could be done:

$$BC = a + b \cdot VAR1 + c \cdot VAR2 + d \cdot VAR3 + e \cdot VAR4 + f \cdot VAR5 + g \cdot VAR6 \quad (6)$$

where BC = Ca + Mg deposition to European forests estimated from throughfall and stemflow fluxes and bulk precipitation.

A more demanding scientific task would be to describe the actual magnitude of Ca and Mg emissions and to develop atmospheric long range transport models for these elements. Even if the transport of Ca and Mg occurs over shorter distances than sulfur compounds, a description of the physics that connect sources and receptors would be very useful for ecological assessment purposes.

Further work is needed before these ideas can be introduced into RAINS. A careful literature study should be carried out to examine how to take into account the internal cycle of Ca and Mg ions within forest ecosystems. Unlike with sulfur, it is not clear whether how significant is the leaching of the internally circulating base cations. The contribution of all the different sources to the basic cation deposition in forests should be studied. Also, it would need to be studied what is the filtering effect of forests regarding base cations.

8. Conclusions on Base Cation Deposition in RAINS

Finally, we may examine what conclusions can be drawn from this material with respect to the way base cation deposition is taken into account in the RAINS model. Other methods have been considered, but presently RAINS uses just one specific way of taking into account the deposition of base cations. Sulfur deposition, obtained from energy-emissions and atmospheric submodels, is transformed into an estimate of acid load by assuming that each mole of sulfur produces two moles of protons. Base cation deposition is assumed to neutralize one-third of this acid load.

Did these data support the above method? Calcium plus magnesium flux, measured under the forest canopy, is presented as a function of the corresponding sulfur deposition in *Figure 8*. A non-linear curve is fitted into the data (solid line $y=100 + 60 * \tanh(S/100-17)$; $n=43$; $r^2=0.66$). The shape of the curve is not derived from any theory. In fact, we believe that there are very few causal relationships between sulfur and base cation deposition and, therefore, the relationship may not follow any simple theory.

The dashed straight line is the RAINS assumption that one-third of the acidifying potential of S is neutralized by base cations. The empirical relationship between base cations and S indicates higher values of base cation deposition than the RAINS estimate over most of the range (light shading). Only with very high sulfur deposition values RAINS seems to overestimate the neutralizing effect due to base cations (dark shading). However, the scatter of the data in high deposition values is quite substantial. The only obvious conclusion is that in remote areas (where S deposition is low) the base cation flux that is *measured under forest canopy* is higher than that assumed by the RAINS model.

These results, however, need not be interpreted in the way that the RAINS model would underestimate the neutralizing effect of base cation deposition falling *from the atmosphere onto the forest canopy*. A large fraction of base cations in stemflow and throughfall samples can have their origin in the tree metabolism and ultimately in the base cation reserves of the soil. Calcium and magnesium are effectively cycled within the ecosystem. Therefore basic cation deposition, measured by means of collecting throughfall and stemflow, generally will overestimate atmospheric base cation deposition to some extent. Sulfur, in turn, has long been known as a "mobile anion" that effectively flows from the atmosphere through the terrestrial environment into aquatic ecosystems.

Estimates of the relative importance of internal cycling to the total base cation deposition onto the forest floor could possibly be gained by comparing bulk precipitation and throughfall and stemflow deposition both for base cations and other ions like sodium and chloride. This should be studied in future.

At the present time there are no European-wide quantitative estimates on the internal cycle of base cations. *Figure 8*, given the considerations above, encourages to keep the current method within RAINS as it is, as far as the time period 1978's and 1980's is concerned.

Although the method is in a reasonable agreement with conditions of the 1970's and 1980's, we must examine the question, *will the relationship between sulfur and base cation deposition remain unchanged in the future?* No, is the current best answer. According to the current emission reduction plans, sulfur emissions will be 30 to 40% smaller in 1995 than they were in 1980. Calcium and magnesium emissions are likely to remain at their current level; at least there are no major international plans to reduce their emissions.

If sulfur emissions decline and base cation emissions remain constant over time, base cation deposition will neutralize a larger fraction of sulfur deposition in the future than today. The stronger the sulfur emission reductions, the faster we will approach the situation that most or all of the acidity due to sulfur deposition will be neutralized by base cation deposition. This is an important finding as regards the RAINS model. The treatment of Ca and Mg deposition should be changed as far as future acid deposition scenarios are concerned.

The default method for computing the neutralization effect of base cation deposition in RAINS future projections should be the following. The model should be changed in such a way that base cation deposition is allowed to vary over space but is kept constant over the time between 1980-2040. The spatial variation could be described in a number of alternative ways. The ideal way would be to have inventories of the atmospheric emissions of base cations and a long range transport model to describe the source-receptor relationships. An alternative way is to develop a regression model (see Eq. 6) with explanatory variables such that can be described over all Europe. In the short term, however, the only option is to draw on the relationships of *Figure 8* that is, to use the coincidental relationship of base cation deposition to S deposition.

Sulfur deposition, after taking into account the forest filtering effect (Eq. 3) is described for the year 1975 into the forest land of each grid square of the RAINS model (the impact model grid). The year 1975 is selected because the data of *Figure 8* represent approximately that period of time. Acid load is then computed, and the neutralizing effect of base cation deposition is estimated as usual as one-third of that load. This spatial distribution of the neutralizing effect is then stored into RAINS and kept constant over time in all RAINS scenarios.

The above procedure seems to be the most justified default method for RAINS calculations for the time being. The main impact of this change will be that the ecological models (soil model and lake model) will respond more strongly to a decrease of sulfur emissions than they do in their present form. Soil acidification and lake acidification according to new calculations will be estimated to cease when sulfur emissions are reduced by 65-70% from the sulfur emission levels in 1975-1980. However, given the additional acid load due to nitrogen compounds, overcoming the soil and lake acidification problem in the most sensitive areas may require additional reductions in both sulfur and nitrogen emissions.

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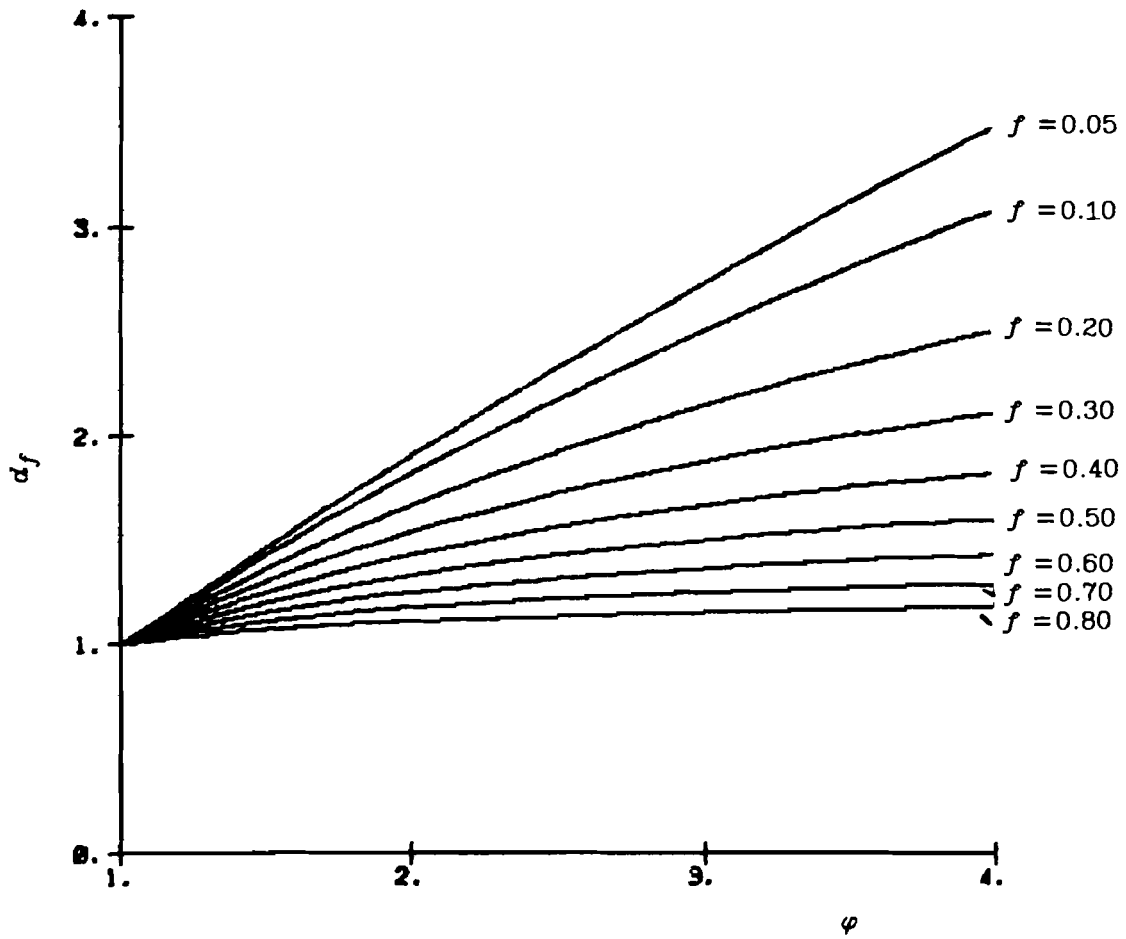


Figure 1. Deposition on forests as a function of the filtering factor φ for various values of the forest coverage f .

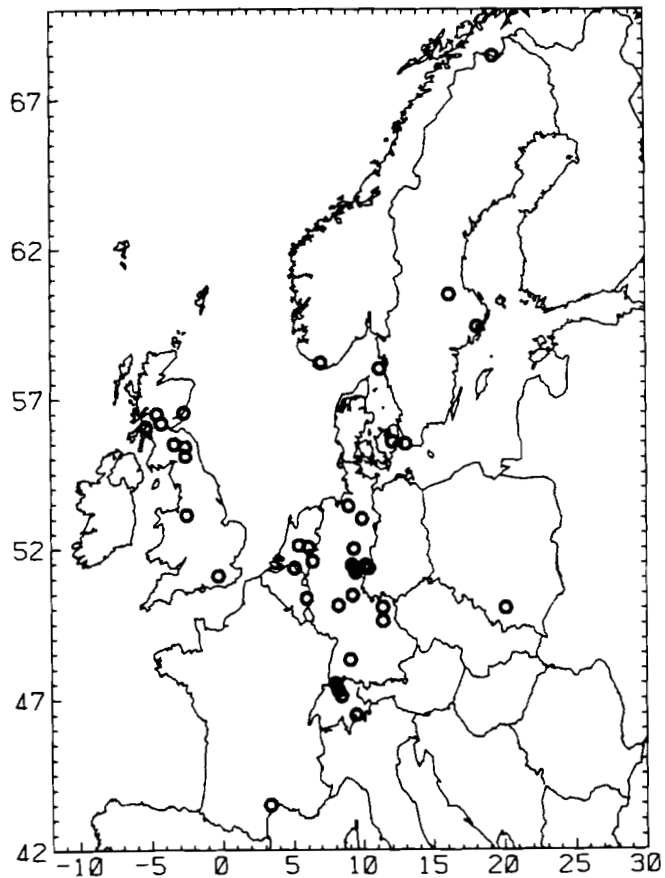


Figure 2. Location of the measurement sites.

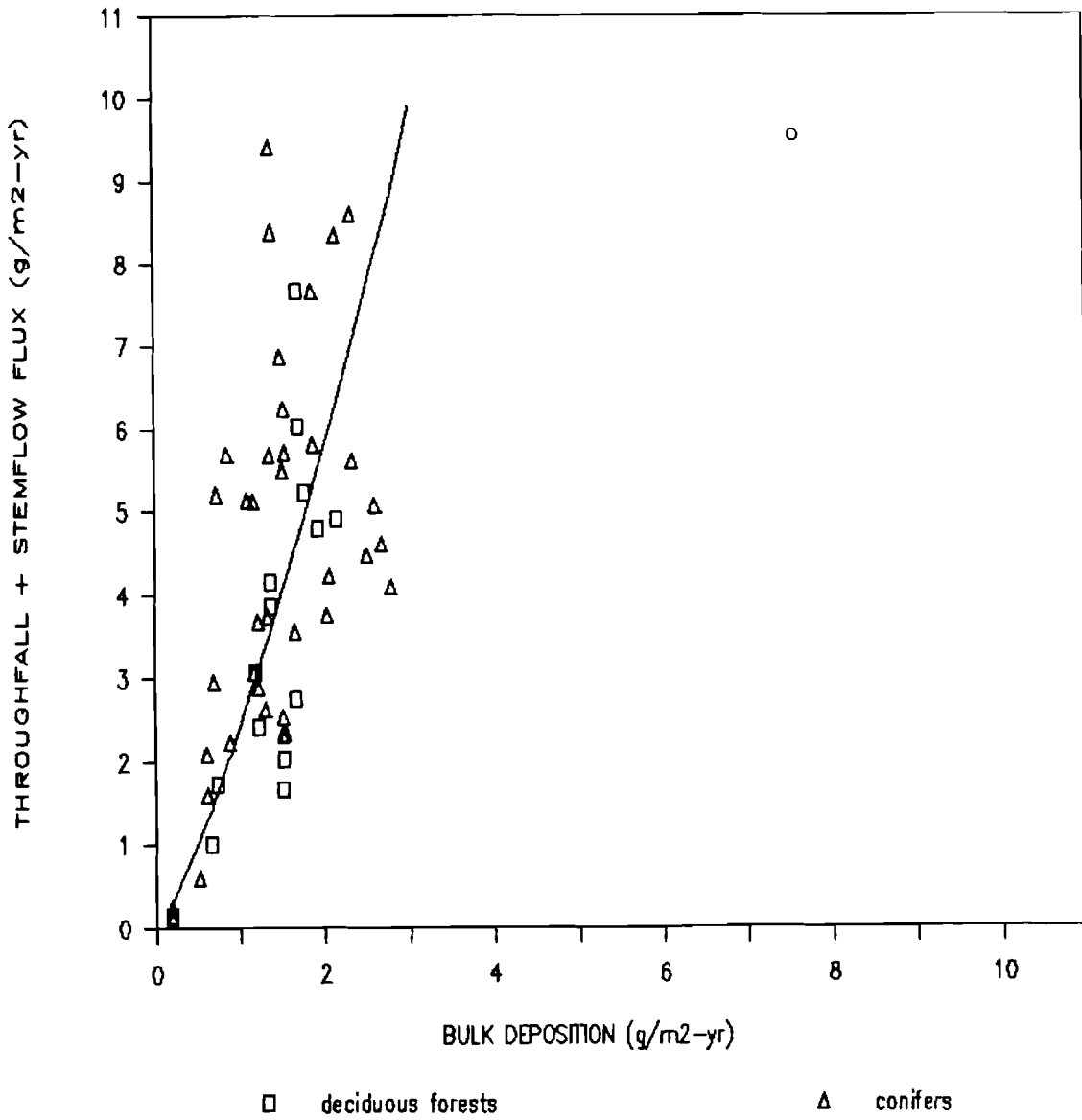


Figure 3. Comparison of the S-flux onto the forest floor (TD) and the S-flux in bulk deposition (BD). The data point indicated with "o" refers to measurements taken in southern Poland (Karkanis, 1976). It may or may not represent east European conditions more broadly; the other observations are from western Europe (Figure 1). Rejecting this data point the following regression is obtained: $TD = 2.42 \cdot BD^{1.26}$ ($n = 53, r^2 = 0.70$)

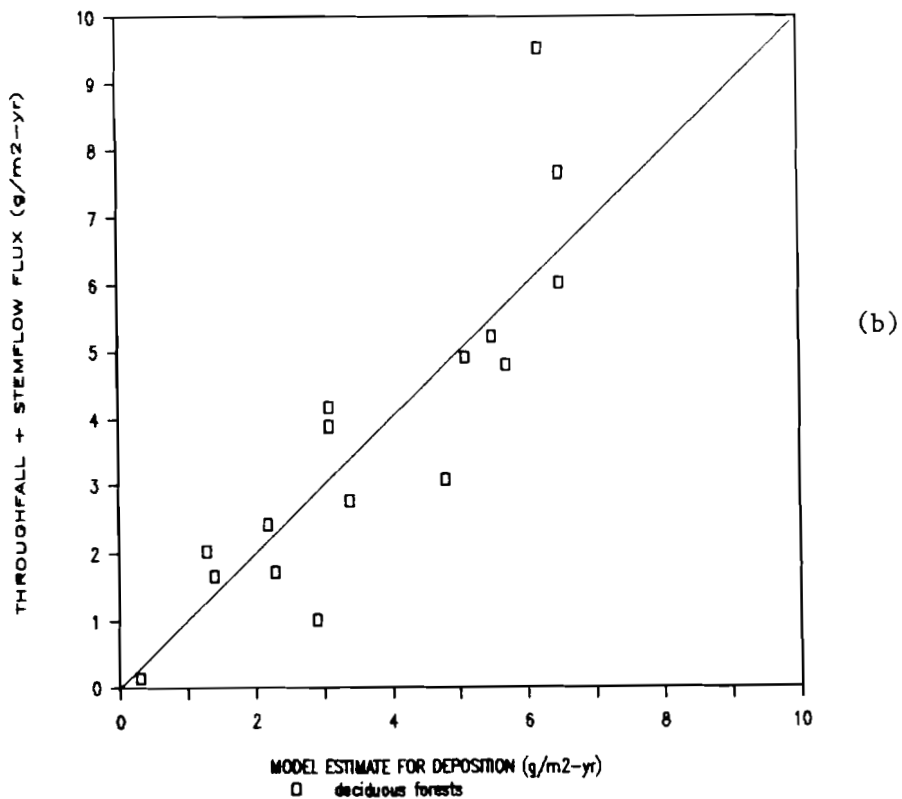
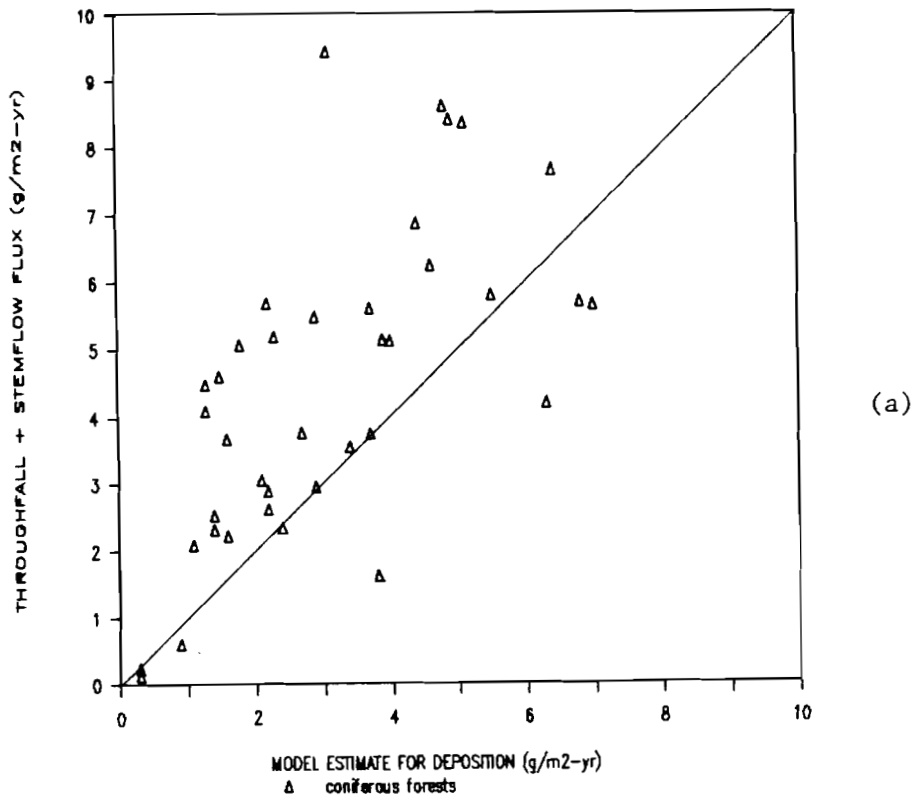


Figure 4. Throughfall and stemflow flux of sulfur onto the forest floor versus S-deposition calculated by the EMEP-model, (a) in coniferous forests, and (b) in deciduous forests.

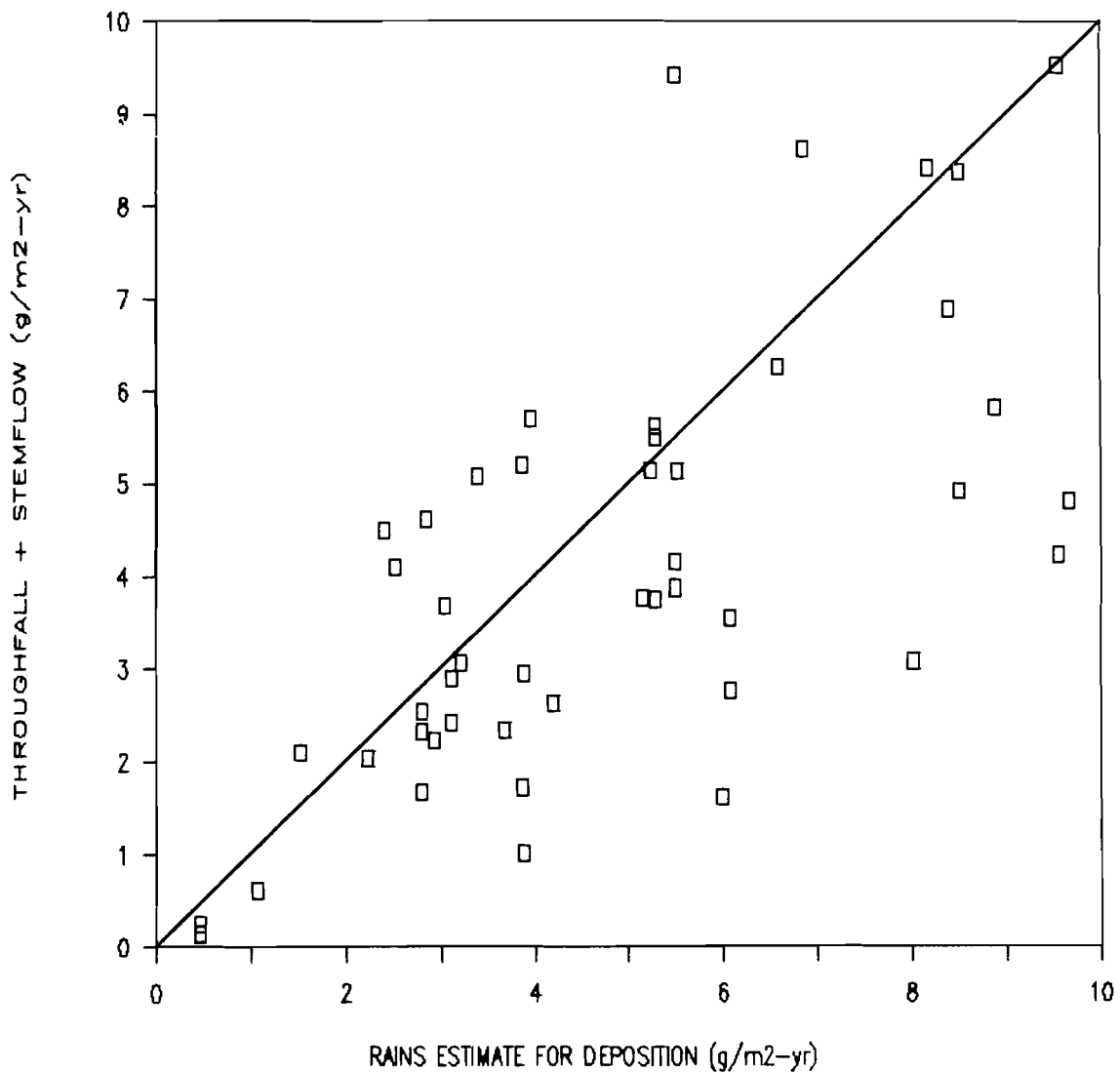


Figure 5. Throughfall and stemflow flux of sulfur onto the forest floor versus S-deposition calculated by the RAINS-model applying a forest filtering parameter (φ) value of 2.0.

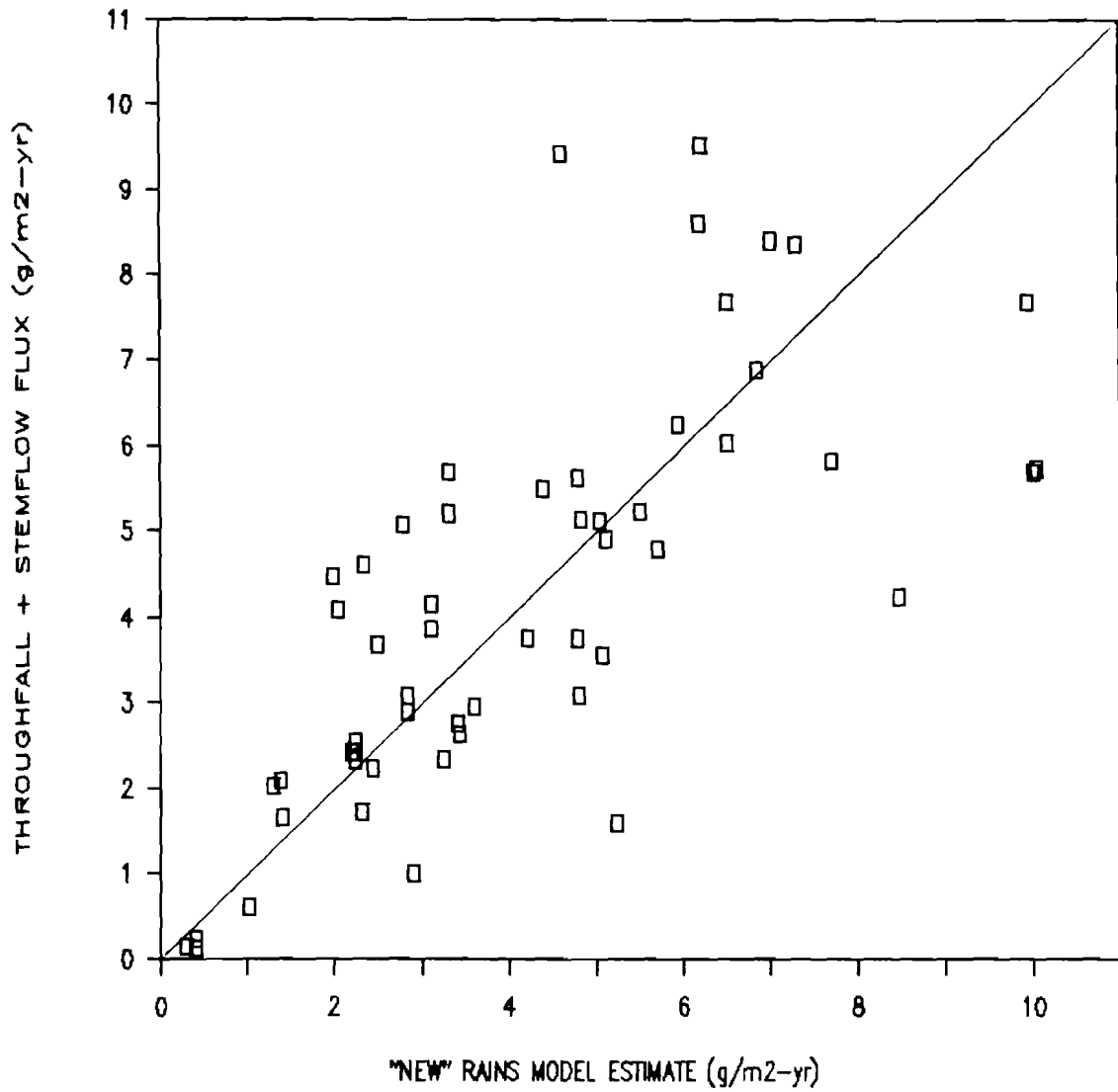


Figure 6. Throughfall and stemflow flux of sulfur onto the forest floor versus S-deposition calculated by the RAINS-model applying $\varphi = 1.6$ for coniferous forests and $\varphi = 1.0$ for deciduous forests.

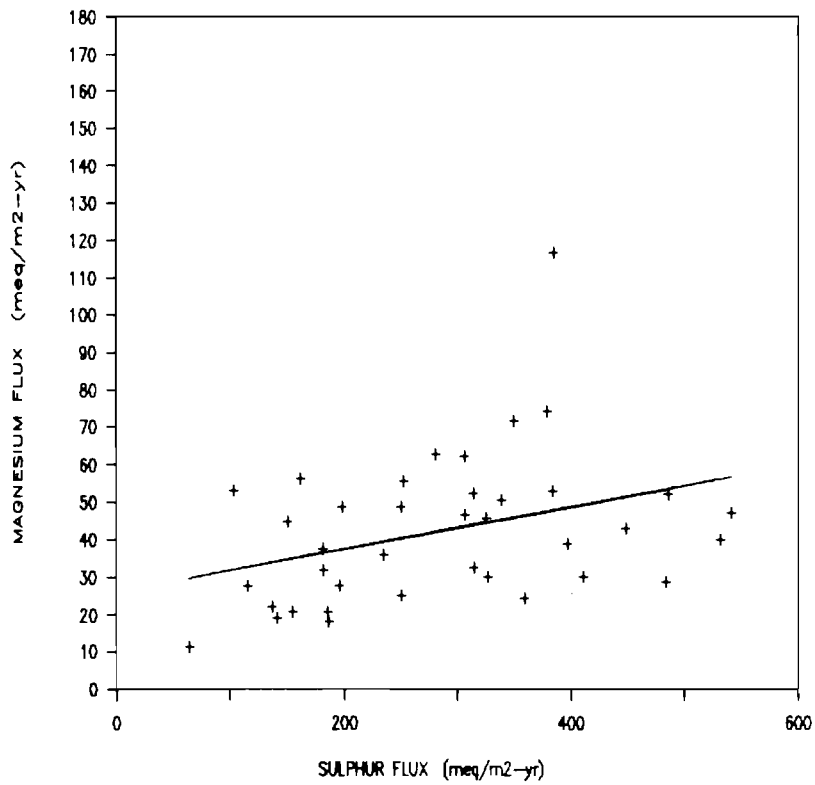
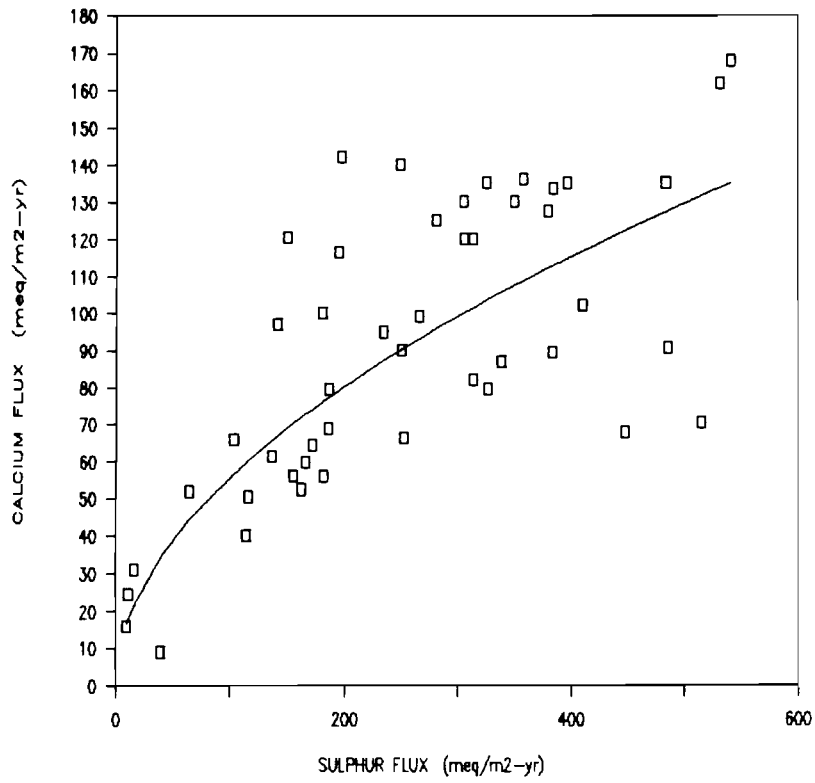


Figure 7. Calcium (Ca) and magnesium (Mg) versus sulfur (S) in the flux onto the forest floor. $Ca = 4.99 * S^{0.52}$ meq/m²-yr ($n = 47$, $r^2 = 0.66$). $Mg = 26.20 + 0.057 * S$ meq/m²-yr ($n = 47$, $r^2 = 0.13$).

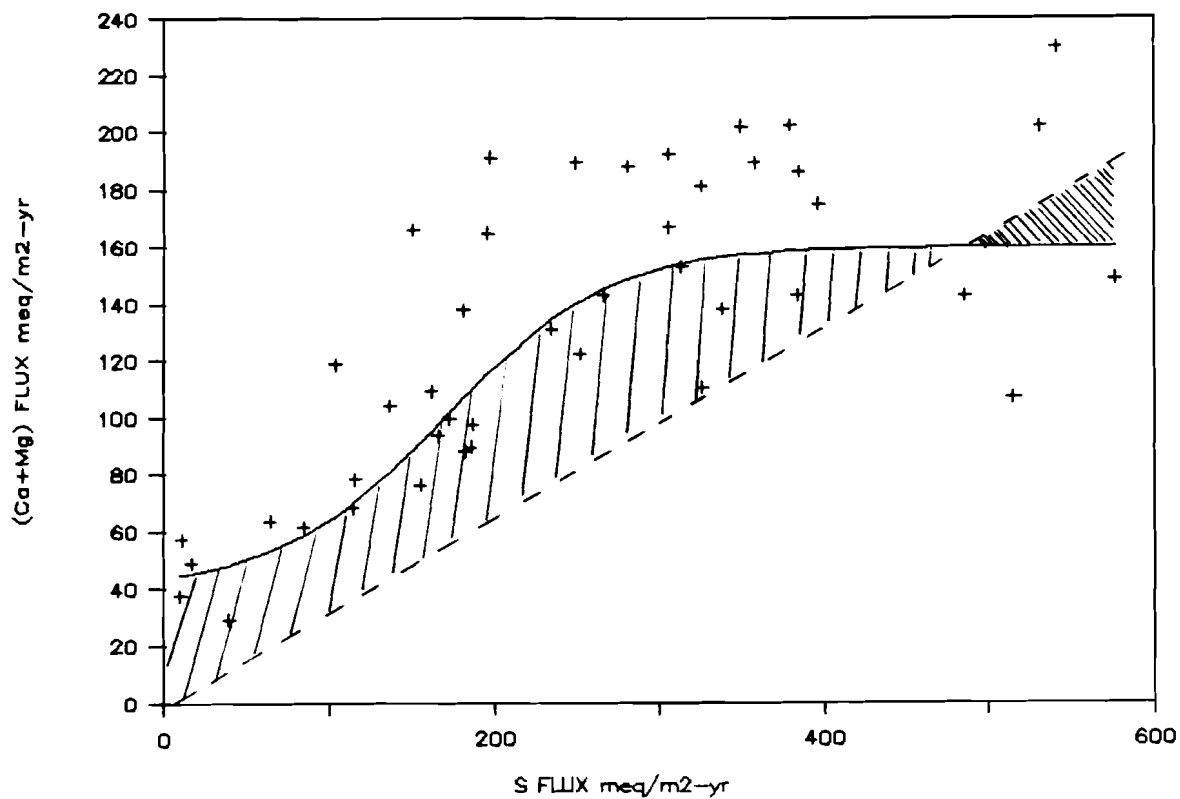


Figure 8. The relationship between base cation deposition and S deposition. Solid line ($y=100+60 * \tanh(S/100-17)$; $n=43$; $r^2=0.66$) has been fitted into the data. Dashed line ($y=0.33 S$) is the current assumption on this relationship within RAINS.