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Sulfur deposition onto European forests: throughfall data and model estimates

By WILFRIED IVENS, *Department of Physical Geography, University of Utrecht, Heidelberglaan 2, 3508 TC Utrecht, The Netherlands,* PEKKA KAUPPI, JOSEPH ALCAMO and MAXIMILIAN POSCH, *Transboundary Air Pollution Project, International Institute for Applied Systems Analysis (IIASA), A-2361 Laxenburg, Austria*

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ABSTRACT

The assessment of atmospheric sulfur deposition to forest is difficult because of its complex aerodynamic structure. Therefore, atmospheric deposition of sulfur to forest is often estimated by means of measuring throughfall fluxes onto the forest floor. In this paper, reported measurements of throughfall fluxes in European forests are analyzed. These fluxes are compared to deposition to bulk collectors located in nearby open land, to get an idea of the filtering efficiency of forests. In addition, fluxes are compared with deposition estimates from a long-range transport model of air pollutants, linked to an emission generation model. According to reported measurements from 52 European conifer stands, we found that the sulfur flux was 3.8 ± 2.3 times greater onto the forest floor than onto precipitation collectors. In a similar data set of 13 deciduous stands this ratio was 2.3 ± 0.9. The ratio of throughfall flux to model estimate was 1.8 ± 0.9 in coniferous stands and 0.9 ± 0.3 in deciduous stands. For sites that are located in moderately to highly sulfur polluted areas, it is assumed that throughfall fluxes give a good estimation of the atmospheric sulfur deposition. We conclude that (1) sulfur deposition to forests is 1.5 to 6 times higher than deposition to smooth receptor surfaces due to an efficient filtering by the forest canopy, (2) average annual sulfur deposition at a given location is 50 to 100% greater on conifers than on deciduous trees, (3) the existing European scale model that links sulfur deposition to the pollution generation processes is quite accurate as far as deciduous forests are concerned, and (4) the model underestimates deposition to coniferous forests.

1. Introduction

Atmospheric pollutants are carried into forests by precipitation (wet deposition), by cloud droplet and fog interception (droplet deposition), and by the deposition of gases and particles onto the canopy surface (dry deposition). Wet deposition is usually assumed to be independent of the receiving land surface, and we believe that this assumption is valid in terrestrial European environments. Droplet deposition however tends to be higher onto forest vegetation than onto other receptor surfaces (Miller and Miller, 1980; Dollard and Unsworth, 1983). Dry deposition of particles, like sulfate aerosols, is enhanced in forests compared to more smooth surfaces, due to the relative high aerodynamic roughness of forests. Dry deposition of gases, like SO_2, to forests might be hampered by the relative high stomatal resistance of trees (Fowler, 1984). In contrast, SO_2 absorption by wetted surfaces can be very high especially in presence of ammonia (Adema et al., 1986). Hultberg (1985) found an overall enhanced atmospheric sulfur deposition to forests compared to clearcut and bog areas. The difference between deposition onto forest and deposition onto low vegetation surrounding the forest has been referred to as "forest filtering" (Kämäri, 1986). Deposition models generally do not account for the forest filtering effect. The aim

of this investigation is to analyze deposition measurements and model results in order to estimate sulfur deposition onto forests in Europe.

To quantify the excess filtering by forests compared to the filtering by low vegetation, total deposition to both the forest and the surrounding area should be measured. Wet deposition can be measured using wet-only samplers situated above the canopy or in a clearing next to the forest. Dry deposition on the forest can be measured by atmospheric flux methods such as eddy correlation or profile methods (Hicks et al., 1986). These methods are resource demanding, and therefore, sulfur deposition to forests is often estimated from the sulfur flux in canopy drip and stemflow. Canopy drip is the water dripping from the canopy during rainfall and stemflow is the water descending along the trunk. Henceforth, the sum of these two fluxes will be referred to as throughfall flux.

Sulfur in throughfall originates from four sources: wet deposition, dry deposition, droplet deposition, and the metabolic extracts of the canopy tissues. Throughfall deposition can be less than atmospheric deposition when SO_2 is taken up by stomata, incorporated in the organic parts of the plant, and is not leached out as sulfate by rain (irreversible stomatal uptake). Throughfall deposition can be an overestimation of atmospheric deposition if sulfur that is taken up by the tree from the soil is leached out of the tree foliage as sulfate (internal cycling). Throughfall fluxes only can be used to estimate atmospheric deposition when irreversible stomatal uptake and internal cycling are small compared to the atmospheric flux.

Garten et al. (1988) conducted radioactive ^{35}S studies in red maple and yellow poplar trees, to evaluate the applicability of the throughfall method to estimate the atmospheric deposition. During a 104 days period in the growing season, internal cycling was 0.05 and 0.04 g S m^{-2} for maple and polar respectively, being about 7% of the total sulfur deposition in this period. The poplar stand showed relative high internal cycling amounts during the leaf-fall period (0.15 g S m^{-2}), while the maple stand did not show increased internal cycling in this period. By comparing these experiments with deposition estimates from air concentration measurements, it was concluded that the throughfall method measures both gaseous and aerosol sulfur deposition and that internal cycling is a minor contributor to the sulfur flux to the forest floor in heavily sulfur polluted areas (Garten et al., 1988). Meiwes and Khanna (1983) estimated leaching from leaf tissues in a sulfur polluted area in Central Europe, by comparing the sulfur concentration of freshly fallen leaf litter with fresh tissue. Their calculations suggest that 0.22 g S m^{-2} could be leached out from senescent leaves of beech and 0.18 g S m^{-2} from senescent spruce leaves. Lindberg et al. (1986) simultaneously sampled dry deposition on leaves and plates, incident precipitation, throughfall plus stemflow and the concentration of airborne particles and vapors. From these measurements, the annual internal sulfur cycling of a mixed hardwood forest in a moderately polluted area in the USA was estimated to be 0.09 g S m^{-2} yr^{-1}. Lindberg et al. (1986) concluded that sulfur was not retained by the foliage and that SO_2 absorbed by the leaves will be leached by subsequent rain events.

Combining these findings, we conclude that atmospheric sulfur deposition, which is on the order of 1.0 to 10.0 g S m^{-2} yr^{-1} (Lehmhaus et al., 1986) over most of Europe, largely determines the amount of sulfur-falling from the canopy layer onto the forest floor.

Bulk precipitation measurements are often used to assess the total deposition to low vegetation. Bulk precipitation is sampled using continuously open funnels. All wet deposition is thus included, but a fraction of dry deposition may be excluded because the collector geometry is, for example, different from that of low vegetation. Grass acts as an effective pollutant filter during the growing season when the leaf area index is as high or higher than in forests (Heil et al., 1988). Bulk precipitation will provide good estimates of atmospheric deposition to smooth receptor surfaces like short grass, but will underestimate deposition to other surfaces.

2. Methods

2.1. Throughfall and bulk precipitation data

We compiled throughfall and bulk precipitation measurements taken between 1967 and 1988 from 65 forest stands in 10 European

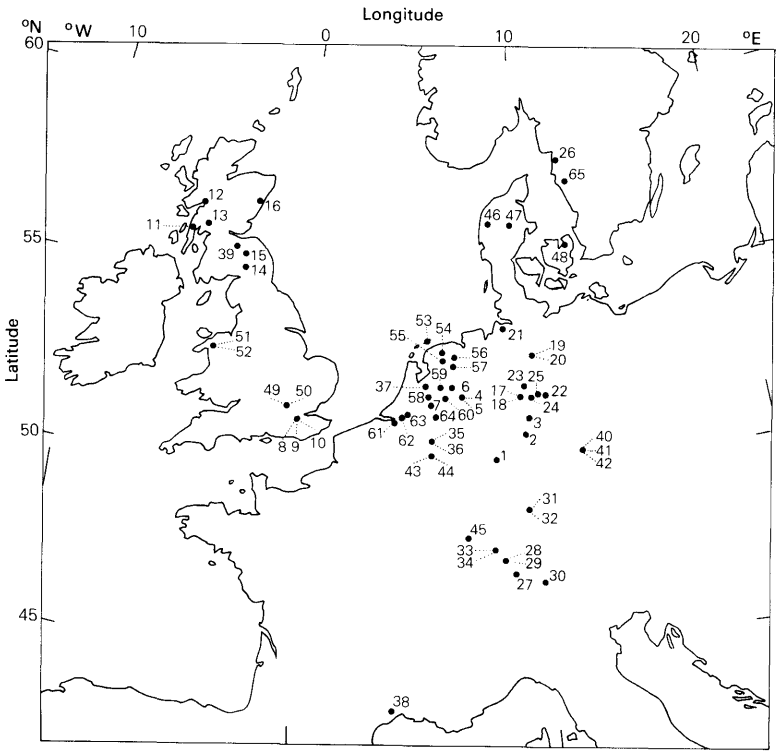

Fig. 1. Location of the case study sites. For references see Table 1.

countries (Table 1). The duration of the measurements varied from one year to more than 10 years. The material consisted of 52 coniferous stands and 13 deciduous stands. The coniferous group consisted of 24 stands of Norway spruce, 8 stands of Sitka spruce, 6 stands of Scots pine, 1 stand of Douglas fir, 9 stands of a mixture of coniferous species and 4 stands of coniferous species mixed with some deciduous species. The deciduous stands consisted of 5 stands of beech, 3 stands of oak, 2 stands of birch, 1 stand of maple and 2 stands of a mixture of deciduous species. Our data do not represent European forest ecosystems strictly in the statistical sense, because the sites were not located systematically or randomly. No data were available for the eastern and southern European countries and for remote areas in northern Scandinavia (Fig. 1).

For 40 cases, the stemflow flux was not given. In these cases, the stemflow flux has been estimated, assuming ratios between stemflow flux and canopy drip flux, depending on tree species and stand age. These ratios were taken from literature (Alcock et al., 1985; Ivens et al., 1988; Johnson et al., 1986; Leitat et al., 1987; Mayer, 1987; Miller and Miller, 1980; Nicholson et al., 1987; Nihlgard, 1970; Rapp, 1973; Van Breemen et al., 1989; Verstraten et al., 1984).

The sulfur fluxes were corrected for the contribution of sea salt particles, using sodium and chloride (depending on which was available) as sea salt tracers (Asman et al., 1981). Average sea-salt contribution was 6% of the total sulphur in throughfall. Sea-salt contributions up to 46% of the total sulfur in throughfall were assigned at coastal sites.

2.2. *Model estimates*

To obtain model deposition estimates, the sulfur emissions of each European country were computed with the Regional Acidification INformation and Simulation model (RAINS)

Table 1. *Characteristics and data for the measurement sites*

Nr	Reference	Long	Lat	Year	Kf	Model	Tf	Bp
1	Georgii et al., 1986	8.47	50.18	83–85	NS	3.80	5.08	1.22
2	Georgii et al., 1986	9.48	50.75	83–85	NS	3.85	5.10	1.15
3	Georgii et al., 1986	9.85	51.33	83–85	NS	5.18	6.19	1.61
4	Verstraten et al., 1984	6.73	51.97	81–82	Dm	5.72	6.02	1.76
5	Verstraten et al., 1984	6.73	51.97	81–82	Cdm	5.72	7.40	1.76
6	Van Breemen et al., 1988	6.23	52.08	81–82	Dm	5.55	5.09	1.91
7	Van Breemen et al., 1982	5.25	51.58	81–82	SP	5.97	5.83	1.62
8	Skeffington, 1983	−0.33	51.17	81	OA	3.20	3.92	1.29
9	Skeffington, 1983	−0.33	51.17	81	BI	3.20	3.74	1.29
10	Skeffington, 1983	−0.33	51.17	81	SP	3.20	10.67	1.29
11	Miller & Miller, 1980	−5.47	56.08	75–77	SS	1.06	4.09	2.66
12	Miller & Miller, 1980	−4.83	56.83	75–77	SS	1.01	4.48	2.42
13	Miller & Miller, 1980	−4.32	56.32	75–77	SS	1.29	4.61	2.69
14	Miller & Miller, 1980	−2.83	55.17	75–77	SS	1.93	3.76	2.02
15	Miller & Miller, 1980	−2.83	55.67	75–77	SS	1.76	2.63	1.22
16	Miller & Miller, 1980	−2.33	56.92	75–77	SS	1.28	5.07	2.51
17	Bredermeier, 1988	9.42	51.75	69–85	BE	5.11	4.92	2.31
18	Bredermeier, 1988	9.42	51.75	69–85	NS	5.11	8.30	2.31
19	Bredermeier, 1988	10.00	53.00	80–85	OA	2.52	2.70	1.57
20	Bredermeier, 1988	10.00	53.00	80–85	SP	2.52	3.38	1.57
21	Bredermeier, 1988	9.03	53.72	83	NS	2.43	5.49	1.34
22	Bredermeier, 1988	10.42	51.75	83	NS	6.27	4.18	2.15
23	Bredermeier, 1988	9.67	52.00	84–85	NS	4.43	8.40	1.41
24	Bredermeier, 1988	9.83	51.58	84–85	BE	5.06	3.07	1.26
25	Bredermeier, 1988	10.33	51.58	84–85	BE	7.76	5.66	1.43
26	Grennfelt et al., 1985	11.50	58.00	80–81	NS	1.44	2.27	0.84
27	Klöti, 1988	8.75	47.17	86–87	NS	2.31	1.95	1.13
28	Klöti, 1988	8.37	47.48	86–87	NS	1.95	4.01	0.98
29	Klöti, 1988	8.37	47.48	86–87	BE	1.95	1.69	0.98
30	Klöti, 1988	9.83	46.80	86–87	NS	2.85	1.12	0.28
31	Bücking & Steinle, 1988	9.17	48.50	79–83	NS	2.87	2.94	0.73
32	Bücking & Steinle, 1988	9.17	48.50	79–83	BE	2.87	0.97	0.69
33	Bücking & Steinle, 1988	8.03	47.85	86	NS	2.26	2.72	1.28
34	Bücking & Steinle, 1988	8.03	47.85	86	MA	2.26	2.34	1.28
35	Buldgen & Remacle, 1984	6.00	50.58	82	Cdm	5.78	4.99	1.80
36	Buldgen & Remacle, 1984	6.00	50.58	82	NS	5.78	5.83	1.91
37	Ivens et al., 1988	5.77	52.18	85–87	DF	4.72	8.06	1.55
38	Rapp, 1973	3.67	43.83	66–70	OA	1.49	1.84	1.42
39	Nicholson et al., 1980	−3.50	55.83	79	SP	1.43	3.68	1.23
40	Hantschel, 1987	11.78	49.98	84–86	NS	5.61	6.55	2.41
41	Hantschel, 1987	11.78	49.98	85–86	NS	5.61	7.74	2.41
42	Hantschel, 1987	11.77	50.07	84–86	NS	6.07	4.00	1.37
43	Laitat & Fagot, 1987	5.92	50.28	85–86	NS	4.25	3.20	1.61
44	Laitat & Fagot, 1987	5.92	50.28	85–86	NS	4.25	4.55	1.61
45	Probst & Dambrine, 1988	7.00	48.03	86–87	NS	2.48	1.94	0.92
46	Rasmussen, 1987	8.42	56.47	85–86	NS	1.52	2.08	1.43
47	Rasmussen, 1987	9.42	56.35	85–86	NS	1.84	2.17	0.91
48	Rasmussen, 1987	12.32	55.93	85–86	NS	2.04	2.32	1.10
49	Alcock & Morton, 1985	−0.68	51.40	76–77	BI	3.50	3.13	3.65
50	Alcock & Morton, 1985	−0.68	51.40	76–77	SP	3.50	2.96	3.65
51	Stevens, 1987	−4.12	53.08	84–85	NS	2.01	3.43	1.57
52	Stevens, 1987	−4.12	53.08	84–85	SS	2.01	3.27	1.57
53	Houdijk & Roelofs, 1988	5.20	53.38	86–87	SP	1.90	5.86	0.94
54	Houdijk & Roelofs, 1988	6.27	53.08	86–87	Cm	2.70	7.09	1.81

Table 1 *continued*

Nr	Reference	Long	Lat	Year	Kf	Model	Tf	Bp
55	Houdijk & Roelofs, 1988	6.32	52.87	86–87	Cm	3.04	5.60	1.83
56	Houdijk & Roelofs, 1988	6.67	52.85	86–87	Cm	3.24	7.22	1.45
57	Houdijk & Roelofs, 1988	6.62	52.58	86–87	Cdm	3.70	7.31	1.45
58	Houdijk & Roelofs, 1988	5.62	52.08	86–87	Cdm	4.43	9.42	1.63
59	Houdijk & Roelofs, 1988	6.02	52.12	86–87	Cm	4.54	8.59	1.43
60	Houdijk & Roelofs, 1988	5.90	51.78	86–87	Cm	5.35	8.65	2.17
61	Houdijk & Roelofs, 1988	4.05	51.28	86–87	Cm	4.04	12.77	1.75
62	Houdijk & Roelofs, 1988	4.33	51.38	86–87	Cm	4.30	7.72	1.63
63	Houdijk & Roelofs, 1988	4.77	51.58	86–87	Cm	4.78	16.88	1.23
64	Houdijk & Roelofs, 1988	5.62	51.30	86–87	Cm	6.30	10.08	1.24
65	Alenaš & Skärby, 1988	12.37	57.26	84–85	NS	1.49	3.63	1.47

Long, Lat = longitude and latitude in decimal units.
Kf = kind of forest: NS = Norway Spruce; SP = Scotch Pine; SS = Sitka Spruce; DF = Douglas Fir; BE = Beech; OA = Oak; BI = Birch; MA = Maple; Cm = mixed coniferous; Cdm = coniferous mixed with some deciduous trees; Dm = mixed deciduous.
Year = measurement period.
Model = model sulfur deposition estimate (g S m^{-2} yr^{-1}).
Tf = throughfall sulfur deposition (g S m^{-2} yr^{-1}).
Bp = bulk precipitation sulfur deposition (g S m^{-2} yr^{-1}).

developed at IIASA (Alcamo et al., 1987). The emission estimates were computed to correspond to the years of the forest deposition measurements. The impact of these emissions to wet plus dry sulfur deposition on the forest sites was estimated based on the average annual transport pattern provided by the runs of the EMEP II long-range transport model for the years 1979, 1980, 1983 and 1984 (Eliassen and Saltbones, 1983). In this way, we did account for the variations in emission generation such as those due to energy combustion between 1967 and 1988 in different countries, but omitted interannual variability in meteorology. However, Alcamo and Posch (1986) estimated the typical variability of total annual S deposition at any location in Europe to be only 5 to 20% due to year-to-year variation in precipitation, winds and other meteorological factors. EMEP model results have been extensively evaluated with measurements (see, e.g., Eliassen and Saltbones, 1983 and Lehmhaus et al., 1986). In an independent evaluation, a consultant to the World Health Organization declared that the model calculated seasonal trends of gaseous SO_2 and sulfate in precipitation "quite well" (WMO, 1988). Eliassen and Saltbones (1983) showed that model computations agree with annual averages within a factor of two.

The EMEP model accounts for wet deposition, and dry deposition due to turbulent transfer in the surface layer. This aspect of dry deposition is calculated with a simple parameterization which depends among other factors, on surface roughness length. This roughness length is estimated from land use type (forest, cropland, etc.). The higher the roughness length, the higher the deposition velocity; hence, under identical meteorological conditions, the computed dry deposition velocity to forests exceeds that to cropland. However, because of the model's coarse spatial resolution, surface roughness is averaged over large grid elements (150 × 150 km). Hence, if a grid element includes both forested and open areas (a typical situation), we expect the model to overestimate open land deposition and underestimate forest deposition within that grid element.

The model does not incorporate surface resistance to dry deposition which depends on stomata openings, wetness of the receptor surface, etc. The EMEP model also neglects droplet deposition which may be more pronounced in coniferous forests than in croplands,

because coniferous forests are often located at hillsides where the frequency of fog is high and the needles contribute to the total receptor area throughout the year. We conclude, therefore, that the RAINS-EMEP model output can be interpreted as the average deposition over large spatial areas of rural, non-forested lands.

3. Results and discussion

The amount of sulfur found in throughfall was significantly greater than that in bulk precipitation (Fig. 2). The ratio of throughfall sulfur to bulk precipitation sulfur was 3.8 ± 2.3 (median $= 3.1$) for coniferous stands and 2.3 ± 0.9 (median $= 2.1$) for deciduous stands. The difference between the coniferous and deciduous forests was statistically significant (t-test; $\alpha = 0.05$).

Throughfall measurements from deciduous stands were not significantly different from the deposition calculations of the EMEP-RAINS model (paired t-test, $\alpha = 0.05$) (Fig. 3a). The average ratio between the measurements and the calculations was 0.9 ± 0.3 (median $= 1.0$). The model estimate clearly exceeds the throughfall at three sites, but at the other 10 sites, the model estimates are very close to the throughfall measurements.

Throughfall measurements from coniferous stands were significantly greater than deposition calculated with the model (paired t-test, $\alpha = 0.05$) (Fig. 3b). The average ratio between the measurements and the calculations was 1.8 ± 0.9 (median $= 1.6$). The throughfall deposition showed to be poorly, but significantly, correlated with model estimates. The regression line between the observed (y) and calculated (x) fluxes was $y = 0.96x + 2.23$ ($r^2 = 0.26$). Some interesting conclusions can be drawn from this regression. Because the regression coefficient is close to one, the intercept indicates that throughfall deposition exceeds model deposition with 2.2 g S m^{-2} yr^{-1} in both polluted and less polluted regions. This can be interpreted in two ways: (1) throughfall deposition overestimates actual deposition to coniferous forests; (2) model deposition underestimates actual deposition to coniferous forests. The only source of overestimation of the atmospheric deposition by

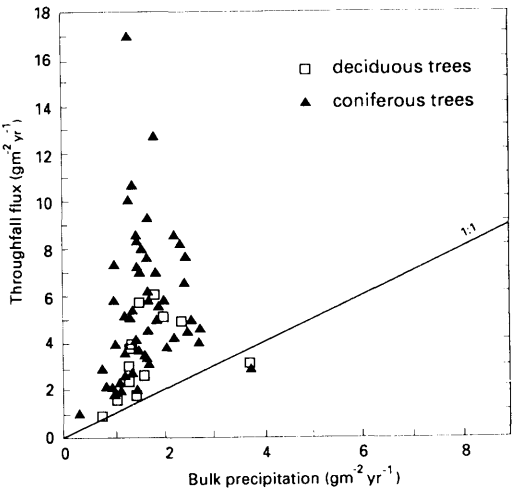

Fig. 2. Sulfur flux onto the forest floor as a function of bulk precipitation sulfur measured in open land nearby the forest stand.

throughfall measurements could be due to the contribution of internal cycling. Internal sulfur cycling in coniferous forests probably does not exceed 0.3 g S m^{-2} yr^{-1}, as has been pointed out before. Thus, we conclude that a great part of the observed constant difference (≈ 1.9 g S m^{-2} yr^{-1}) must be caused by an underestimation of the atmospheric deposition by the model.

The scatter observed in Fig. 3 originates both from model uncertainty and measurement errors. The model does not distinguish between different emission sources within the 150×150 km grid elements, which can cause large errors for sites close to SO_2 sources. In ammonia polluted areas, such as the Netherlands, sulfur deposition is enhanced by the presence of ammonia (Adema et al., 1986; Ivens et al., 1988; Van Breemen et al., 1982). In our data, we found a good correlation between sulfur (S) and ammonium–nitrogen (NH_4–N) in throughfall plus stemflow:

$$S = 2.84 + 0.79 * (NH_4\text{–}N)$$

(g S m^{-2} yr^{-1}; $n = 44$; $r^2 = 0.68$).

Therefore, the scatter may partly be caused by the omission in the model of the chemical interaction of sulfur with ammonia on the receptor surfaces.

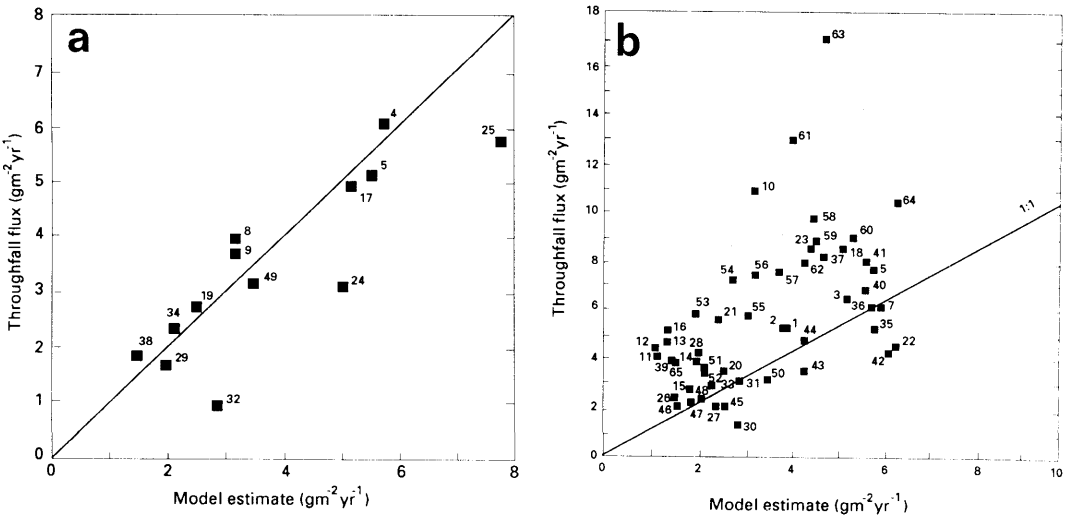

Fig. 3. (a) Sulfur flux onto the floor in 13 deciduous stands as a function of the total sulfur deposition calculated by the RAINS-EMEP model. (b) Sulfur flux onto the forest floor in 52 conifer stands as a function of total sulfur deposition calculated by the RAINS-EMEP model. For references see Table 1.

Even within a uniformly sulfur polluted area, throughfall measurements can differ from site to site because of different forest characteristics, such as canopy structure, stand density and tree height (Ivens et al., 1988; Lovett and Reiners, 1986). The exact location of the samplers in the forest stand is of importance. The closer the sampler is to the edge of the forest stand, the more it will receive sulfur. Throughfall fluxes of sulfur in forest edges can be about 1.5 times greater than the sulfur fluxes in the interior of the forest (Draaijers et al., 1988; Hasselrot and Grennfelt, 1987).

The average deposition to the deciduous sites and the coniferous sites was respectively 3.9 ± 1.8 (median = 3.2) and 3.4 ± 1.6 (median = 3.2) g S m^{-2} yr^{-1}; the difference is not statistically significant (t-test, $\alpha = 0.05$). Therefore average sulfur air-pollution was expected to be comparable for both groups. The average throughfall deposition was 3.5 ± 1.6 (median = 3.1) at the deciduous forests and 5.5 ± 3.0 (median = 5.0) in the coniferous forests, the difference being statistically significant (t-test, $\alpha = 0.05$). Thus throughfall deposition at coniferous forests is 1.6 ± 1.1 times greater than at coniferous forests.

A better comparison of the throughfall deposition in these two kinds of forests can be done for sites where throughfall deposition is measured in both coniferous and deciduous forests. In our data set, such a comparison is only possible for 8 sites. At 7 sites throughfall deposition is greater in coniferous forest than in deciduous forest (Fig. 4). The average ratio of throughfall deposition in coniferous stands forest stands and in deciduous stands at these 7 sites is 1.8 ± 0.8, which agrees with the estimate made before.

We think that throughfall measurements can be used for the validation of the model in moderately to highly polluted areas. The model can not provide exact deposition estimates for any given forest stand, but is most useful to describe the general deposition pattern on a continental scale. The prediction of the deposition onto deciduous forests was rather accurate given all the potential sources of uncertainty in the method of this study. The model is likely to underestimate deposition onto areas dominated by conifers. High altitude forests and forested headwater lakes, which have been identified to be sensitive to the effects of sulfur deposition, are mostly dominated by conifers.

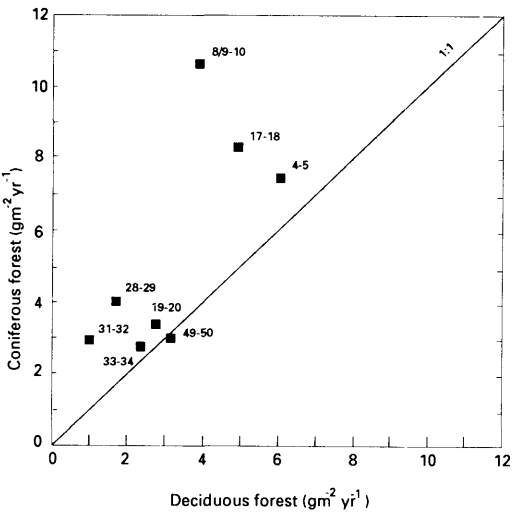

Fig. 4. Sulfur flux onto the forest floor in coniferous stands versus sulfur flux in deciduous stands. For references see Table 1.

Therefore, correction coefficients may be needed to assess the deposition onto a landscape where conifers are the main tree species. In general the EMEP model could be improved by: (a) taking into account droplet deposition; (b) taking into account the interaction of sulfur and ammonia deposition; (c) partitioning deposition estimates into forest and non-forest components.

If indeed the deposition to deciduous forests equals the model deposition estimate, and the deposition to coniferous forests is greater than this estimate, then the deposition to non-forest land must be less than this estimate. Otherwise the average deposition to the grid element would be biased. Assuming that it is not biased, taking into account that conifers and deciduous trees cover respectively 7 and 15% of the considered land area (OECD, 1987), we may speculate that the deposition onto non-forest land is less than the model deposition estimate by a factor of ≈ 0.85.

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