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Review of deposition monitoring methods

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ABSTRACT

Deposition monitoring may generally serve at least one of the following purposes: (a) determining ecosystems at risk, (b) evaluating temporal and spatial trends, (c) developing numerical models, or (d) estimating deposition effects. In this paper, an overview of available knowledge about monitoring methods is given. Furthermore, it is tried to illustrate the (dis)agreement between results obtained using different methods. It might be concluded that throughfall, micro meteorological methods (supplemented/supported by inference) and watershed balance methods (S saturated systems) yield similar estimates of the annual mean total deposition of sulphur, within generally acceptable uncertainly limits ($\sim 30\%$). A larger uncertainty exists to estimate reduced or oxidised nitrogen and base cation fluxes. It is clear that for individual ecosystems deposition in general, and dry deposition in particular, can still not be quantified with sufficient accuracy. The various methods have different advantages and drawbacks and the choice of a certain method for estimation of the flux of a specific pollutant to a specific ecosystem may in many cases depend on the purpose of the study and on requirements on accuracy and costs.

1. Introduction

Quantification of the input of airborne substances to aquatic and terrestrial ecosystems has been of concern for decades, but despite many years of research and development, methods to measure deposition of air pollutants still face major problems. Several techniques have been developed for measuring different types of deposition: wet, dry, cloud and fog and total deposition. Wet deposition can be measured with reasonable accuracy, whereas direct measurement of and especially routine monitoring for dry deposition and for cloud and fog deposition are still very difficult.

This paper aims at presenting the "state of the art" of deposition monitoring. How far have we progressed, which substances can be measured by what methods, and what future research is required? The paper contains a short summary of measurement methods, mainly based on existing excellent reviews by Hicks et al. (1986, 1989) and Davidson and Wu (1989). Furthermore, an overview and assessment is presented of possibilities for measuring deposition of specific substances, mainly focused on dry deposition methods. A separate section is devoted to monitoring deposition in complex terrain, such as hill slopes, mountainous regions and roughness transition zones (e.g., forest edges). It is investigated how deposition monitoring can be used to develop numerical models in order to derive area average deposition maps. Deposition maps may serve as tools in the process of definition and evaluation of emission reduction plans and as tools in deposition effect studies for those areas where deposition monitoring is difficult or impossible. Gaps in knowledge are identified and summarised.

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2. Monitoring methods

2.1. *Wet deposition*

Wet deposition is defined as the process by which atmospheric pollutants become associated with cloud and precipitation droplets (or particles) and subsequently delivered to the earth's surface during precipitation. The amount of compounds thus received per unit of area of the surface is defined as wet deposition. Wet deposition is measured by collecting precipitation in samplers placed in the open field. Precipitation is collected in bottles for time periods typically ranging from days to months, and the sample is analysed for its chemical composition. Wet deposition equals to the amount of precipitation per unit area of the funnel of the sampler multiplied by the measured concentration of the pollutant in the sample. Wet deposition is considered to be independent of receptor surface in all but mountainous environments.

Although measurement of wet deposition seems relatively simple, there are some serious error sources (see, e.g., Eriksson, 1952; Allerup and Madson, 1980; Slanina et al., 1982; Fowler and Cape, 1984; Buijsman and Erisman, 1988; Beier and Rasmussen, 1989). In earlier years, the most commonly used sampler was the bulk or open sampler, which has no provision to exclude dry deposition during dry periods. Dry deposition of gases and particulate matter may therefore influence the chemical composition of precipitation passing the contaminated funnel (Slanina et al., 1982; Ridder et al., 1984). The use of wet-only samplers, in which the funnel is open to the atmosphere only during precipitation events, is widely recommended. Also, short exposure periods are recommended when sample deterioration is expected due to bacterial or chemical action. Nowadays, several national and international precipitation monitoring networks exist (EMEP, CaPMoN, NADP).

2.2. *Cloud and fog deposition and dew*

Vegetation may intercept cloud and fog droplets or directly collect water vapour that forms dew. Cloud droplets typically are in the diameter range 10–50 μm and can be efficiently captured by vegetation (Fowler, 1984; Fowler et al., 1989; Hicks et al., 1989). Impaction to, and interception by foliage and other obstacles in their path may be very effective. Cloud droplets usually contain higher concentrations of pollutants than are found in rain, and cloud water deposition may exceed annual precipitation, especially at high elevation sites (Lovett, 1988). In Europe, at altitudes of 400 m above sea level and above, low clouds are present between 500 and 2000 h per year (Fowler et al., 1991). Cloud water deposition might be a significant input mechanism for such regions in Europe.

Recently, much research on cloud and fog deposition has been conducted (Weathers et al., 1986; Schemenauer, 1986; Mohnen, 1988; Lovett and Kinsman, 1990; Mitchell et al., 1990; Fowler et al., 1991; Johnson and Lindberg, 1992). Several techniques have been applied by which droplet deposition could be separated from dry deposition and deposition in precipitation. Cloud water can be collected by droplet impaction on a collection surface usually consisting of Teflon strings of thickness comparable to that of conifer needles (Weathers et al., 1986; Waldman et al., 1982; Mallant and Kos, 1990). Also, estimates of cloud water deposition have been made using throughfall measurements (see Section 2.3), together with sensors for cloud occurrence and measurements of cloud water amount (Joslin and Wolfe, 1992). Dollard et al. (1983) and Gallagher et al. (1988) reported micro meteorological measurements of cloud water deposition to short vegetation. From these studies, it was found that cloud water deposition rates, averaged over the droplet size spectrum in these measurements, are close to the reciprocal of the aerodynamic resistance for momentum.

Dew is water vapour condensed on relatively cool bodies (vegetation). Therefore, fresh dew usually contains very low concentrations of pollutants (van Aalst and Erisman, 1991; Römer et al., 1991). Dew may play an important role in the process of dry deposition by enhancing surface wetness; in this way, surface resistance to soluble gases will be lowered and dry deposition increased. This process is therefore important for dry deposition estimates.

To our knowledge, no routine methods for cloud, fog or dew deposition are available, although some methods have been used for extended periods at selected sites (e.g., Mohnen, 1988).

2.3. Dry deposition

Dry deposition is the process whereby gases and aerosols are deposited directly from the atmosphere to surfaces. Dry deposition is governed by the concentration in air, by turbulent transport processes in the atmospheric boundary layer, by gravitational settling (for sufficiently large particles), by molecular diffusion or other transport processes near the surface, by the chemical and physical nature of the depositing species, and by the capability of the surface to capture or absorb gases and particles.

Micro meteorological methods. Among the methods for measuring dry deposition, micro meteorological methods are most suitable for determining the dry deposition of many gases. With these methods, the flux to the total system may be determined, and the relationship between air concentrations, meteorology and the flux is directly established. In flat homogeneous terrain, the flux measured at a sampling point above the surface within the constant flux layer represents the average vertical flux over the upwind fetch. With the most direct of the micro meteorological methods, the eddy correlation method, the flux is derived from measurements of the vertical component of the wind velocity and the gas concentration. With another method, the gradient technique, the flux is derived from measurements of air concentrations at several heights above the receptor surface and meteorological variables. Other techniques comprise Bowen ratio approaches, variance and a variety of so called conditional sampling methods.

Micro meteorological methods require a considerable monitoring effort. Continuous measurements using a dry deposition monitoring system for low vegetation have been made for SO_2 (Davies and Mitchell, 1985; Erisman et al., 1993A), NH_3 (Erisman and Wyers, 1993; Wyers et al., 1992) and NO_2 (Erisman et al., 1993B). Monitoring systems based on micro meteorological techniques for forests are now under development (Vermetten et al., 1992; Enders et al., 1992; Erisman, 1992; Erisman et al., 1993C). No routine micro meteorological methods for measuring dry deposition of particles are available.

An advantage of micro meteorological techniques over other methods is that measurements are made above the surface, measuring fluxes to the total sink surface; i.e., including vegetation and soil. Another advantage is that these techniques allow continuous measurements and thus the opportunity to study dry deposition processes in relation to meteorological and receptor conditions.

Time-average measurements at a certain level provide an area integrated average of the exchange rates between the surface and the atmosphere. The measuring height must be well within the constant flux layer, and near to the earth surface (5–10 m above vegetation). This means that usually only local fluxes can be measured (low measuring heights to fulfil these demands). Extrapolating these fluxes or derived deposition parameters to larger areas is still a great problem, because of varying surface properties and roughness characteristics and accordingly non-homogeneous turbulent behaviour. Fluxes cannot be estimated with these methods in complex terrain or near to sources, where the constant flux layer is not fully developed. It is required that measurements are made over terrain with an upwind fetch over a homogeneous surface, large enough to establish a fully developed constant flux layer. In addition to fetch requirements, the development of the constant flux layer requires that no sources or sinks exist in the atmosphere above the surface, and that the concentration of the constituent does not vary significantly with time (Baldocchi et al., 1988; Hicks et al., 1987; Businger, 1986; Erisman et al., 1993A). Sources or sinks in the atmosphere may be the result of rapid chemical reactions in the layer between the height of flux measurement and the surface. The influence of reactions has been suggested for NH_3 and acidic gases, such as HNO_3 and HCl (Huebert and Robert, 1985; Erisman et al., 1988; Brost et al., 1988; Allen et al., 1989; Erisman and Wyers, 1993) and has been demonstrated for the photo stationary equilibrium between NO, NO_2 and O_3 (Lenschow, 1982; Duyzer, 1991; Wesely et al., 1989; Kramm, 1989). In general, all these limitations make the micro meteorological methods less suitable for routine application. However, they are very useful for studying deposition processes and for validating models and measuring methods more suitable for monitoring.

Surface accumulation methods. Measurements of dry deposition can also be made by measuring the deposition at the surface itself directly or indirectly (surface accumulation methods). Direct methods comprise the measurement of deposition to natural surfaces, using the throughfall method (see below), or to surrogate surfaces, such as dustfall buckets, flat plates, petri dishes or other devices intended to approximate natural surfaces. All these methods may be useful for measuring deposition of large particles. Other direct methods include measurements of accumulated material by electron microscope counting, laboratory studies (on, e.g., stomatal conductance), wind-tunnel studies, and chamber studies. Here the discussion is limited to field experiments. While inert surfaces have provided useful data in process level studies (e.g., Davidson et al., 1985), they do not directly simulate vegetation. The direct method with most potential in this regard is the throughfall method.

Throughfall method. Throughfall has typically been used for quantification of soil loads with nutrients and not for atmospheric deposition estimates. Throughfall refers to the water dripping from canopies and stemflow to that running down tree trunks. The difference between throughfall flux and open field wet deposition is "net throughfall". To the extent that deposited material is washed from the canopy by rain, the net throughfall flux of an ion or insoluble element below the canopy, provides information on the dry deposition. However, the quality of this information is strongly dependent on the extent of the processes influencing the content of solutes in throughfall or the knowledge about these processes. Ions in throughfall have many sources such as incoming rain, dry deposition wash off, and foliar leaching. The term foliar leaching refers to the process where nutrients which enter the plant by root uptake, are incorporated in foliage, and "leach" into throughfall during a rain event (hence, not representing deposition wash off). If this is significant, it will clearly bias throughfall results. Several studies reviewed in Lindberg et al. (1992) support the assumption that wash off of deposited sulphur occurs, and that foliar leaching occurs, but the latter is a relatively small contributor of S to throughfall. If the results of these studies are universal, throughfall may provide as accurate an estimate of the long-term total atmospheric deposition of S as is possible with other techniques (20–30%, Hicks et al., 1986). A short qualitative description of factors influencing throughfall (and stemflow) is presented in Parker (1983) and Lindberg et al. (1992).

Throughfall is measured by placing precipitation samplers beneath the canopy and collecting the water dripping from the canopy. Throughfall measurements are often performed with open samplers on a time scale similar to precipitation. Several collectors should be used, or throughfall collectors should be designed to adjust to throughfall patterns to the forest floor (Rasmussen et al., 1986; Beier and Rasmussen, 1989; Ivens, 1990). Stemflow is measured occasionally; it usually represents only 1–10% of the total ion flux (van Breemen et al., 1988). Throughfall measurements in grassland and in heath land have been reported by Bobbink et al. (1992) and Heil et al. (1989).

Analysis of throughfall in forests has become an increasingly attractive tool for studies of atmospheric fluxes. This method holds some advantages over other techniques: it provides long-term mean fluxes, and it is much cheaper and requires less effort than micro meteorological methods. Furthermore, throughfall measurements may be useful to estimate input fluxes in those areas where micro meteorological methods cannot be used, such as near to sources or in complex terrain. It can provide useful information about processes in the canopy and on the variation of fluxes within a site (Lindberg et al., 1988). Finally, total throughfall fluxes may be used in some instances as estimates of total wet + dry + cloud deposition of an ion.

However, a limitation of this approach is that often important information is lacking to distinguish between in-canopy or atmospheric sources of chemical compounds (Ivens, 1990; Lindberg et al., 1986). Furthermore, throughfall measurements do not provide information about the origin of the input in terms of gas or particle deposition except for base cations and measurements of throughfall are not possible in areas without vegetation. Finally, dry deposition directly to the forest floor is not measured.

Watershed mass balance method. The outflow from catchments is equal to the sum of wet + dry deposition, weathering release and net change in storage in biomass and soil. By measuring the outflow flux, an estimate for the deposition to the catchment is obtained when the latter two are negligible or can be quantified. Up to now, this approach has been applied successfully for the Lake Gårdsjön area in Sweden (Hultberg & Grennfelt, 1992) and the Hubbard Brook forest ecosystem in the USA (Likens et al., 1990) and has given reliable deposition estimates for S, Na and Cl to whole catchments. The possibility of using this method is limited to those areas where the necessary assumptions are valid. Where the facilities (gauged watersheds with routine stream chemistry monitoring) already exist, it is a simple and cheap method for estimating deposition.

Inferential technique. As an alternative to the direct measuring methods, knowledge on dry deposition processes can be used to infer dry deposition fluxes from basic information on routinely measured air concentrations and meteorological parameters. The flux is inferred as the product of ambient concentration of the chemical of interest and its dry deposition velocity. The dry deposition velocity is derived using a multiple-resistance transfer model (Hicks et al., 1987). The resistance model provides a framework for coupling individual processes, some being surface dependent or pollutant dependent. The three major resistance's are the aerodynamic resistance R_a, a near surface boundary resistance R_b, and the surface resistance R_c. R_c is estimated from dry deposition measurements, from published data, or from use of a more detailed canopy model.

Flux estimations on a routine basis using the inferential technique are made in two networks in the USA (Hicks et al., 1991; Clarke et al., 1992). Weekly mean measurements of HNO_3, SO_2, NO_3, SO_4 and NH_4 concentrations in these networks are combined with meteorological parameters to obtain weekly average fluxes (Hicks et al., 1991). This technique has also been applied in studies done in the UK (UK Review Group on Acid Rain, 1990), in Sweden (Lövblad et al., 1991) and in the Netherlands (Erisman et al., 1989; Erisman, 1992; 1993A).

The quality of results obtained by this method depends on the availability and quality of data, and on the description of the resistance's. R_a and R_b can be estimated from measurements of wind speed and radiation and information on surface roughness, together with parametrizations given by Garland (1977) and Wesely and Hicks (1977). R_c values can be obtained from micro meteorological studies and from chamber methods. The most important parameters determining R_c should be included on a relevant time and spatial scale.

Eder and Dennis (1990) developed an inference technique to allow estimation of the annual and monthly dry deposition of Ca, Mg, Na and K. The technique is based on the strong correlation between concentrations within precipitation and the surface-level air at 23 stations in Ontario, Canada.

3. Evaluation and comparison of different methods

3.1. Wet deposition

Comparisons between wet-only and bulk precipitation fluxes are made by Grennfelt et al. (1985), Georgii et al. (1986), Clark & Lambert (1987), Mosello et al. (1988), Ridder et al. (1984), Ruijgrok and Römer (1993), Slanina et al. (1990); Stedman et al. (1990) and Richter & Lindberg (1988). In these studies, bulk precipitation of SO_4, NO_3 and NH_4 is found to be between 4% and 34% higher than corresponding wet-only precipitation fluxes. Even larger differences are reported for base cations like Na, Ca, Mg and K (7–75%). Especially coarse particles seem to deposit onto bulk precipitation funnels during dry periods (through sedimentation). The amount of dry deposition onto bulk precipitation funnels is influenced by the distance to local sources, ambient concentrations, wind-shading effects around the funnel, design of sampler, sampling frequency and cleaning protocols.

Inter-comparison of wet-only samplers have shown that the results obtained are very sensitive to the type of sensor used for the registration of onset/offset of the precipitation events (Graham et al., 1988).

Bulk samplers might be useful in specific conditions (high elevation, remote areas). However, wet-only samplers are generally recommended. Sampling procedures need to be designed with great care and sampling locations should be selected to be representative for the area under study (not near to sources or to obstacles such as fences, trees and buildings). In addition, measures should be taken to preserve the samples if biologically or chemically active species are to be studied; i.e., using light-protected bottles, addition of preservatives, reducing storage temperature, or by minimising the sampling and storage period.

3.2. Dry deposition

Measurement of dry deposition of different pollutants exhibit different difficulties and no single method can probably be regarded as the "best" for monitoring. In the following we will summarise the results of comparison studies between different measuring methods applied for sulphur, nitrogen and particulate matter.

Sulphur. Detailed long-term comparisons between dry deposition estimates from inferential plus deposition plate methods on the one hand, and dry deposition estimates from the throughfall method on the other were made by Lindberg and Lovett (1992) during the integrated forest study (IFS). Sulphur was found to behave more or less conservatively in the canopy. For a large number of sites scattered over the USA, Canada and Europe, minor SO_2 uptake balanced to some extent foliar leaching of soil derived sulphate. The estimated total annual deposition of sulphur compounds was found to be within 15% of the measured sulphate fluxes in throughfall plus stemflow in each case ($R^2 = 0.97$).

Experimental evidence for canopy uptake of SO_2 was found by Gay & Murphy (1985), but Schaefer & Reiners (1990) and Granat & Hällgren (1992) concluded that essentially all of the dry deposited sulphur dioxide retained by the canopy (taken up via the stomata) is eventually extracted out of the apoplast pools (i.e., aqueous layer on the outside of cell membranes) by rain and appears in throughfall. Furthermore, several radioactive $^{35}SO_4$ studies have shown that, in general, less than 4% of the total throughfall flux of sulphate is caused by foliar leaching of soil-derived sulphate. (Garten et al., 1988; Lindberg & Garten, 1989; Cape et al., 1992).

Comparisons between inferential dry deposition estimates on the one hand, and dry deposition estimates from micro meteorological measurements and/or throughfall data on the other were made by Ivens (1990), Draaijers & Erisman (1993), Lovett et al. (1992) and Erisman (1993B). Ivens (1990) compared SO_4, NO_3 and NH_4 throughfall fluxes from all over Europe with deposition estimates from the long-range transport model EMEP used in RAINS (Eliassen & Saltbones, 1983; Alcamo et al., 1987). For coniferous tree species, sulphate throughfall fluxes were found to be significantly higher (on average, 80%) compared to sulphur deposition estimates from the RAINS model. For deciduous tree species, both estimates were not significantly different.

Erisman (1993B) compared SO_4, NO_3 and NH_4 dry deposition estimates using the inferential technique with throughfall measurements in 24 forest locations in the Netherlands. The two estimates showed large differences, throughfall fluxes being 45% higher than inference estimates. However, variations in time and space were similar. Draaijers & Erisman (1993) made a detailed comparison between net throughfall measurements of sulphate in thirty different forest stands in a small area in the Netherlands (Van Ek & Draaijers, 1992), and sulphur dry deposition estimates from the same receptor oriented inferential model (Erisman, 1992; 1993A). Similar results were obtained as those by Erisman (1993B). However, by extending the surface resistance parametrization for SO_2 in the inferential model with results obtained by a field study on co-deposition of NH_3 and SO_2 (Erisman and Wyers, 1993), the relationship between the two estimates was found to be good. No systematic differences were found. Erisman et al. (1993B) and Erisman (1992) compared SO_4 throughfall measurements under heather (Bobbink et al., 1992) with results of the annual average flux of SO_2 measured at the heath land using the dry deposition monitoring system for SO_2 (sulphate particle deposition was estimated by inference). They found good agreement between the two estimates. For the Hubbard Brook forest ecosystem, Lovett et al. (1992) reported higher (65%) sulphur dry deposition estimates from the throughfall method compared to estimates from an inferential model (Hicks et al., 1987). However, the inferential model is known to underestimate dry deposition in complex terrain (Hicks and Meyers, 1988).

Inference estimates of sulphur dry deposition in the Netherlands compared well with estimates using the LRTAP model TREND (van Jaarsveld and Onderdelinden, 1992) on a 5 × 5 km scale. Furthermore, inference results compared well with dry deposition measurements using micro meteorological methods (Erisman, 1992; 1993A). Wesely and Lesht (1989) compared RADM SO_2 dry deposition results with site specific estimates using inference. Their results showed some systematic differences between the two techniques, due to differences in the algorithms for computing resistance's. Weekly averages of the deposition velocities were within approximately 30% of each other.

Comparisons between dry deposition estimates obtained from the throughfall method and dry deposition estimates from the watershed mass balance method are reported by Hultberg & Grennfelt (1992) for the Lake Gårdsjön area in Sweden and by Likens et al. (1990) for the Hubbard Brook Forest Ecosystem in the USA. Runoff and throughfall fluxes of sulphate are found very similar suggesting the change in storage in biomass and soil, and weathering release being of minor importance. Sulphate fertilisation in several catchments did not enhance sulphate throughfall fluxes significantly, supporting the hypothesis that foliar leaching is insignificant, and that throughfall provides a reasonably good measure for sulphur ($SO_2 + SO_4$ aerosol) deposition.

Nitrogen. There is considerable experimental evidence for significant uptake of inorganic nitrogen by canopy foliage, stems, epiphytic lichens or other micro flora. Canopy foliage has been demonstrated to be capable of absorbing and incorporating gaseous NO_2 and HNO_3, as well as NO_3^- and NH_4^+ in solution (Reiners & Olson, 1984; Bowden et al., 1989). In laboratory experiments, NH_4^+ in solution is found to be exchanged with base cations present in leaf tissues (Roelofs et al., 1985). Epiphytic lichens have also been shown to be active absorbers of NO_3^- and NH_4^+ in solution (Lang et al., 1976; Reiners & Olson, 1984).

The absorption or removal of nitrogen in the canopy will generally cause throughfall measurements to underestimate the total deposition. Johnson and Lindberg (1992) conclude that, on average, 40% of all inorganic nitrogen input to the IFS forests was retained by the vegetation, whereas 60% is found back in the throughfall data as NO_3^- and NH_4^+. Part of the inorganic nitrogen retained by the canopy may be converted into organic substances and subsequently leached. Total nitrogen (organic + inorganic) in throughfall and stem flow is found to be about 84% of the total inorganic nitrogen deposition (Johnson and Lindberg (1992)). Lovett (1992) compared total annual N deposition estimates with throughfall measurements in these forests and found that they agree reasonably, provided a "correction factor" for N uptake is applied to throughfall fluxes.

In a comparison between throughfall and model estimates of nitrogen input to forests in Europe, Ivens (1990) found no significant difference between NO_3 throughfall fluxes and NO_y (= total oxidised nitrogen) deposition estimates, but NH_4 throughfall fluxes were significantly lower (on average, 70%) compared to NH_x (= total reduced nitrogen) deposition estimates. The correlation's between throughfall fluxes and deposition estimates from the RAINS model were very poor. This was attributed to the large deposition variability introduced by local sources and by local differences in aerodynamic and/or surface resistance not accounted for in the RAINS model, and by canopy exchange processes obscuring deposition estimates from the throughfall method.

In a separate study, NO_3 net throughfall fluxes were found significantly lower (on average, 30%) compared to NO_y dry deposition estimates using the inferential model (Erisman, 1992; 1993A). NO_x dry deposition estimates from micro meteorological measurements made over forest in the Netherlands were also somewhat lower compared to NO_x deposition estimates from this model. NH_4 net throughfall fluxes were not significantly different from NH_x dry deposition estimates, but their correlation was rather poor. NH_3 deposition estimates from micro meteorological measurements made over heather and forests in the Netherlands are found both lower and higher compared to inferential model estimates. This is attributed to the large spatial variability in NH_3 dry deposition amounts due to the impact of local sources and short atmospheric residence times of NH_3 (Erisman, 1992; 1993A).

Nitrogen cycles within ecosystems are very complex. Up to now, no reliable estimates for inorganic nitrogen deposition can be made using the watershed mass balance method.

Base cations. Na is normally considered to be more or less conservative in the canopy, showing only minor canopy exchange (Parker, 1983). Consequently, Na in throughfall was suggested to be used as a substance for modelling particle dry deposition (Ulrich, 1983). This model has been widely used (Freiesleben et al., 1986; Bredemeier, 1988; Horn et al., 1989; Ivens, 1990; Beier, 1991; Beier et al., 1992; 1993). However, canopy exchange of Na may occur; Fassbender (1977) found uptake of Na by young Spruce trees. Furthermore, Reiners and Olsson (1984) reported leaching of Na from canopies of Balsam Fir in a low input area.

Lovett and Lindberg (1984) developed an approach to estimate dry deposition from net throughfall for a number of ions based on a multiple regression model. Lindberg et al. (1988) compared this method with deposition measurements of Ca to inert surfaces to develop scaling factors to relate fluxes on scales from small plates to whole canopies. This scaling method has also been applied to the dry deposition of Na, and based on a comparison of throughfall data with deposition measurements of a large number of IFS sites in the United States, Johnson & Lindberg (1992) conclude that Na in throughfall may be considered as solely derived from atmospheric deposition. Next to the throughfall method and the surface accumulation method, also the mass balance approach in watersheds is found to provide reliable estimates for atmospheric deposition of Na (Hultberg & Grennfelt, 1992).

Unlike Na, a substantial part of Mg, Ca and K in throughfall is normally assumed to be caused by canopy leaching (Parker, 1983). However, a literature compilation made by Parker (1990) indicates that it is not clear to which degree Mg, Ca and K present in throughfall originate from atmospheric deposition and foliar leaching, respectively. Canopy leaching contributed between 10% and 80% to the total flux of these base cations reaching the forest floor. However, at coastal forest sites, Mg in throughfall may be predominantly caused by atmospheric deposition of sea-salt particles (Parker, 1983; Beier et al., 1992). Calcium in throughfall may be enhanced at sites located in areas with calcareous soils or near calcium fertilised arable land (Johnson & Lindberg, 1992). For the IFS data, Johnson & Lindberg estimated that leaching represented an average of ~70% of the annual throughfall plus stem flow flux of K^+ below 12 diverse forests, while Ca leaching represented about 40% and Mg leaching about 50%. Magnesium and calcium may also be irreversibly retained within the canopy in case of limited supplies from the soil (White & Turner, 1970; Abrahamson et al., 1976; Alcock & Morton, 1981). A new method for estimating the dry deposition of K, Ca and Mg, based on the Na deposition in a forest edge, was recently suggested by Beier et al. (1992).

3.3. *Cloud and fog deposition*

Joslin & Wolfe (1992) concluded that net throughfall may be used as a gross estimate of total cloud S deposition through subtracting precipitation and dry inputs from total throughfall sulphate in high cloud environments. Meuller (1992) compared two techniques for estimating cloud deposition to a high-elevation spruce forest, i.e., throughfall and precipitation chemistry and a mechanistic cloud deposition model with a cloud event database. The comparison showed a discrepancy of about 30–35% of the total throughfall sulphate, throughfall being higher than model estimates.

4. Deposition in complex terrain

Within forested areas, a large deposition variability is observed which is attributed to a large extent to differences in tree species, tree height, canopy density and edge effects (Van Ek and Draaijers, 1991; Draaijers et al., 1992; Beier et al., 1993; Draaijers, 1993).

Results from throughfall measurements (Potts, 1978; Hasselrot & Grennfelt, 1987; Draaijers et al., 1988; Beier & Rasmussen, 1989; Draaijers, 1993) and deposition modelling (Wiman & Agren, 1985; Bosveld & Beljaars, 1987; Van Pul et al., 1992) have shown that dry deposition in forest edges is considerably enhanced compared to deposition inside the stand. Dry deposition is found to decrease exponentially with distance to the forest edge. The width of the zone with enhanced dry deposition never exceed five edge heights. The deposition enhancement in forest edges is found to be influenced by stand density, edge aspect and pollution climate (Wiman, 1988; Draaijers, 1993). Up to now, edge effects have not been incorporated in present-day regional scale and receptor oriented deposition models: deposition is treated as an one-dimensional transfer to homogeneous surfaces with infinite length. For the Netherlands, approximately 30% higher dry deposition amounts for forests are computed when taking into account edge effects (Van Pul et al., 1992).

Wiman & Agren (1985), Bosveld & Beljaars (1987) and Van Pul et al. (1992) presented numerical models describing transport and deposition in single forest edges. Klaassen (1992) presented a multi-layer transfer model through which regional fluxes of momentum were calculated, taking into account advection processes by using a simple mixing length closure model. Local advection was found to increase the regional momentum flux by up to 50% of the value that would have been obtained if a one-dimensional transfer was assumed with immediate adjustment of atmospheric properties to the underlying surface. Up to now, there seem to be no physical based models which can adequately account for the impact of landscape architectural complexity on regional deposition amounts. Regional scale deposition modellers trying to cope with edge effects have to deal also with transitions in surface roughness and surface resistance, and with the impact of shorter atmospheric residence times, and hence the reduced long range transport of gases and particles induced by edge effects.

If the surface is flat, describing small scale roughness should be sufficient as an input parameter of terrain roughness for inference deposition models. However, mountains and valleys may have a significant effect on the overall roughness of a terrain. Smith & Carson (1977) developed an empirical model for grid cells of 10 × 10 km to estimate the influence of orography on roughness length. They used the average height range between peaks and valleys in the grid and the average distance between peaks separated by valleys or successive ridges as input parameters for their computations. More physically based models describing neutral stratified boundary flow over hilly terrain are presented by, e.g., Walmsley et al. (1986) and Beljaars et al. (1987).

Much information on the influence of hill slopes on deposition estimates has been obtained from studies made in Great Dun Fell in the UK (Fowler et al., 1988; Dollard et al., 1983; Gallagher et al., 1991). The influence of topography on cloud deposition was clearly demonstrated. Rates of cloud droplet deposition to vegetation were found to be similar to rates of momentum deposition. These findings provide the basis for estimates of cloud deposition inputs of major ions to uplands.

5. Synthesis

In this paper, the available knowledge on monitoring methods is summarised. Furthermore, it is tried to inter compare results obtained using different methods. It might be concluded that throughfall, micro meteorological methods (supplemented/supported by inference) and watershed balance methods (S-saturated systems) yield similar estimates of the annual mean total deposition of sulphur, within generally acceptable uncertainly limits ($\sim 30\%$). A larger uncertainty exists to estimate reduced or oxidised nitrogen and base cation fluxes.

It is clear that for individual ecosystems deposition in general, and dry deposition in particular, can still not be quantified with sufficient accuracy. The various methods have different advantages and drawbacks and the choice of a certain method for estimation of the flux of a specific pollutant to a specific ecosystem may in many cases depend on the purpose of the study and on requirements on accuracy and costs. For now, it is impossible to obtain an accurate annual average deposition map of Europe based on actual deposition measurements. Dry, cloud and fog deposition show very strong horizontal gradients due to variations in ambient concentrations, in land use, in surface conditions and in meteorology. Therefore, deposition maps should be generated based on a combination of models and measurements.

A possible method could be to link time dependent concentration maps derived from long-range transport models and/or air concentration measurements, with inferred sub-grid deposition velocities (Erisman, 1992; 1993A; van Pul et al., 1992). Deposition velocities can be estimated using the resistance analogy. Individual resistance's can be inferred from land use data, data on surface conditions and meteorological parameters (Hicks et al., 1987). These data determine the spatial resolution of the dry deposition estimates. Wet deposition maps can be obtained from LRTAP models and/or measurements (EMEP, national data). This method is described in Erisman and Baldocchi (1993). In such a method, measurements are supplementary to models, regarding spatial and temporal scales. Furthermore, measurements are used for developing process descriptions and for evaluation of model results. Finally, measurements can act as an independent tool for assessing policy targets (trend detection). These issues require different measuring/monitoring strategies.

Process oriented studies. The process oriented studies are primarily used to derive insight into deposition processes for different components, and to obtain process descriptions and parameters to be used in models. For these purposes, micro meteorological methods provide the best methods. In those cases where micro meteorological methods can not be used, such as complex terrain and within forest stands, throughfall measurement is the only available method up to now. Process oriented studies can be used to test or verify simple/cheap measuring methods, which might be used for other purposes such as monitoring.

Evaluation of models. For evaluation or validation of model results, preferably simple and cheap monitoring methods are desired. In general, monthly to annual average fluxes are used for validation. The uncertainty in results obtained by these monitoring methods should be within acceptable limits. Furthermore, results should be representative for areas used as receptor areas in the model. Validation of LRTAP model results can be done by area representative measurements of wet deposition and of ambient concentrations. Micro meteorological measurements suitable for monitoring might be used for evaluation of model dry deposition fluxes (Hicks et al., 1991; Erisman et al., 1993A; Erisman and Wyers, 1993). Throughfall measurements might be used as a validation method for spatial variability in S, Na and Cl dry (and total) deposition, provided that several criteria on the method and siting are met (e.g., Beier and Rasmussen, 1989; Ivens, 1990). It is advisable to equip several monitoring locations in Europe with dry deposition monitoring systems, wet-only sensors and cloud and fog deposition measuring methods. These locations should be selected based on pollution climates and type of vegetation. Furthermore, the area surroundings should be homogeneous and no sources should be near to the site.

Detection of trends. If the purpose of measurements is trend detection, the annual averages should be measured as accurately as the magnitude of the trends. For trend detection ambient concentration and wet deposition measurements can be used (EMEP monitoring network). The trend in concentrations is representative for the dry deposition trend, which can not be measured accurately enough at present. The disadvantage of using only concentration measurements is that a change in dry deposition due to ecosystem response (as a result of reduced loads or climatic change) or due to changes in surface conditions (interaction with other gases, etc.) can not be detected. Extensive deposition monitoring (see previous section) might be useful for trend detection, especially at larger emission reductions.

6. Gaps in knowledge

Based on information presented in this synthesis several gaps in knowledge regarding deposition monitoring can be identified.

There is a great need for increasing our understanding on the factors of importance for the range and variability in deposition in order to take such factors into account in the design of monitoring programmes, in modelling and in the assessment and evaluation of deposition measurements.

There is a great need for further knowledge about canopy processes in order to distinguish the deposition and the canopy contribution to the throughfall flux. This may involve further use of radioactive tracers, more detailed canopy sampling in both time and space and comparisons of surface sampling techniques with micro meteorological methods. In general there is a need for direct method inter comparisons.

There is a general need for further development of sensors (especially for NH_3 and particles) and micro meteorological methods to be used for process studies. Furthermore, micro meteorological methods should be developed for routine application. Process oriented studies need to be extended to obtain parametrizations of parameters such as R_c. These parametrizations have to be incorporated in deposition estimates using inference.

Cloud and fog deposition measuring methods need further development and testing.

Existing monitoring programs (EMEP, NADP) should be extended with dry and cloud and fog deposition measurements or with measurements needed for the application of inferential techniques. Furthermore, monitoring programmes should be extended with several extensive monitoring locations and with many simple and cheap routine monitoring sites (e.g., throughfall sampling). Network design and representatives of sites with regard to homogeneity, type of vegetation, and pollution climate need special attention.

Extrapolating these fluxes or derived deposition parameters to larger areas is still a great problem, because of varying surface properties and roughness characteristics and accordingly non-homogeneous turbulent behaviour. There is a need to develop methods to extrapolate point measurements to regions, especially in complex terrain.

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