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FACTORS AFFECTING DRY AND WET DEPOSITION

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Contents:

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Summary

There are three mechanisms by which air pollutants can be deposited on the earth's surface namely dry deposition, wet deposition and occult deposition. A short overview is given of the processes that play a role in each of these mechanisms and the main factors that affect deposition fluxes are indicated. The influence of several parameters on dry deposition rates is discussed on the basis of a resistance layer model.

1. INTRODUCTION

For many pollutants deposition is the main removal route from the air compartment. Other relevant pollutants are removed from air after chemical conversion in the gas phase or in the aerosol phase. Wet deposition is the process by which pollutants are removed by precipitation (rain, snow). Dry deposition is the process by which pollutants are transferred from the air without the help of precipitation. As a third route occult deposition may be acknowledged. This route includes pollutant transfer by mist interception. Although, in most cases, the latter process may not be relevant as a removal process for air pollutants, it may play a significant role in the effect side of a pollutant. This is one example where it is important to realize that there are two ways to look at deposition phenomena, namely from the air compartment side and from the other side: the terrestrial ecosystem. From the air compartment side the deposition process is just a loss term for example in a large-scale modelling effort. In view of the scale of these models the information necessary to provide this loss term is also very global. For effect studies, however, the amount of material actually entering the ecosystem from the air is a key parameter. Normally ecosystems under study are much smaller than the grid size in a dispersion model. To estimate the deposition load into this ecosystem much more detailed information is necessary. This is especially true for dry deposition because the dry deposition process is very sensible to local parameters such as terrain roughness.

As an example of the different character of dry of wet depostion figures 1, 2 and 3 from Asman [1] are presented. In figure 1 the average concentration of NH_3 is given as calculated from an air pollution dispersion model and an emission data base. In figures 2 and 3 the calculated deposition fluxes are shown. Both fluxes are related to the air concentration, although dry deposition is much more sensitive to local air concentrations.

For different areas in Europe the ratio of the amount of dry versus wet deposition is not constant. As an example the estimated dry and wet fluxes for the Netherlands are presented in table 1. These figures (taken from Van Aalst [4]) are based on measurements of wet deposition fluxes (rain water monitoring network) and on estimates of dry deposition fluxes. The dry deposition fluxes are estimated from measured average air concentration (Dutch air monitoring network) and average dry deposition velocities using the formula: Flux = $-v_d.c.$*)

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*) Fluxes directed to the earth's surface have a negative sign.

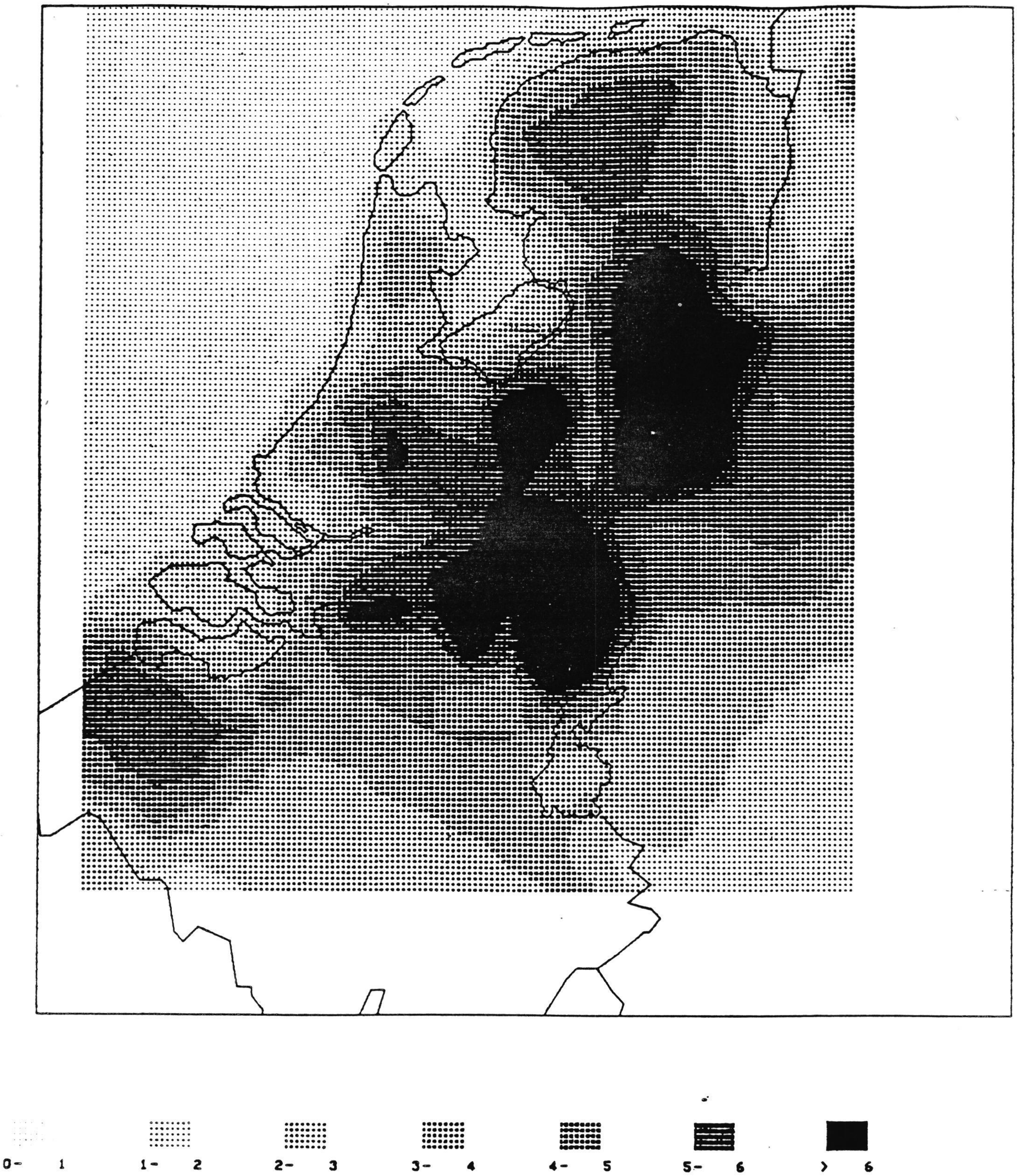


Figure 1 Calculated yearly average concentration of ammonium in air ($\mu g/m^3$), from [1].

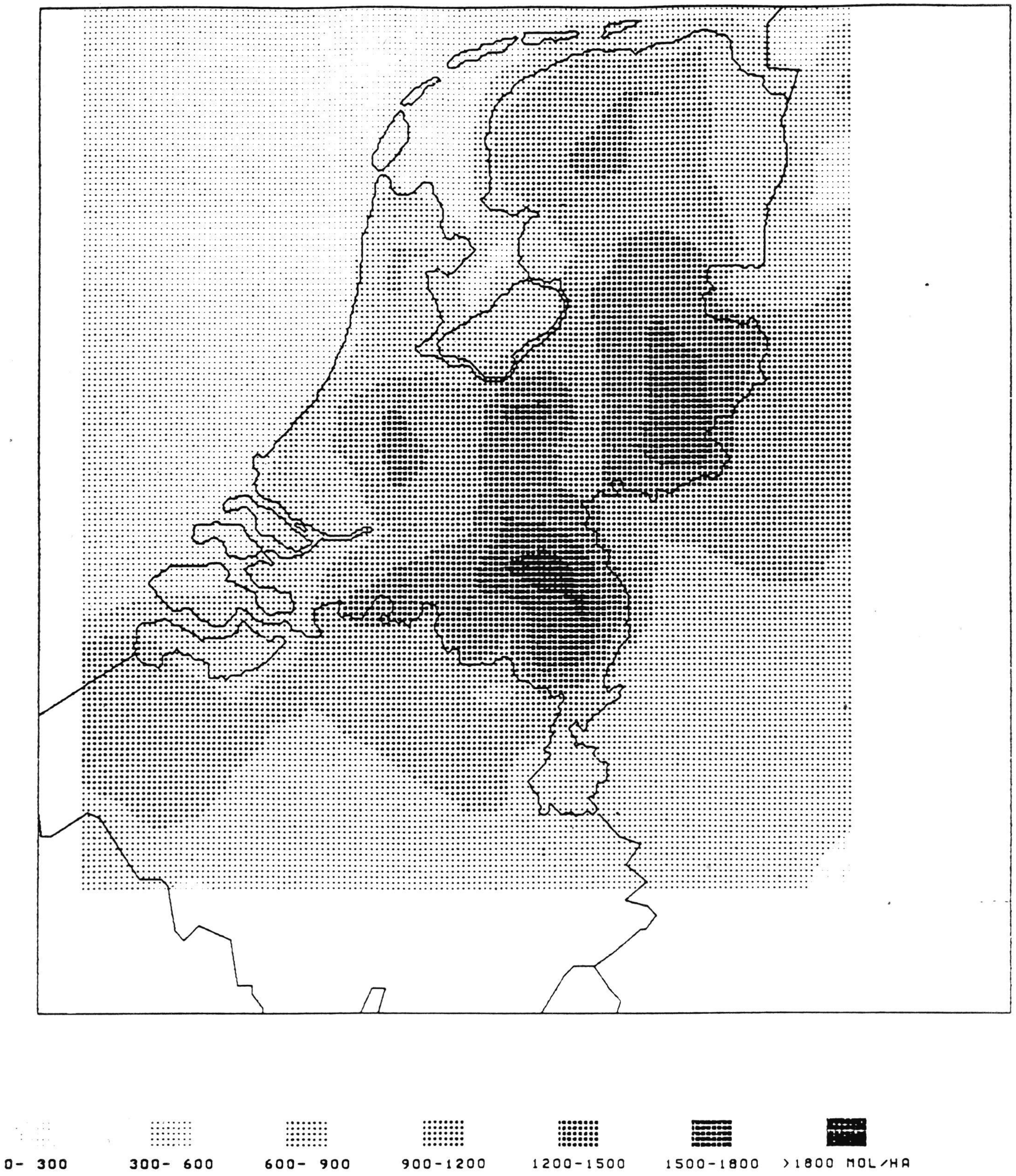


Figure 2 Calculated yearly average dry deposition of ammonium and ammonia (mol/ha), from [1].

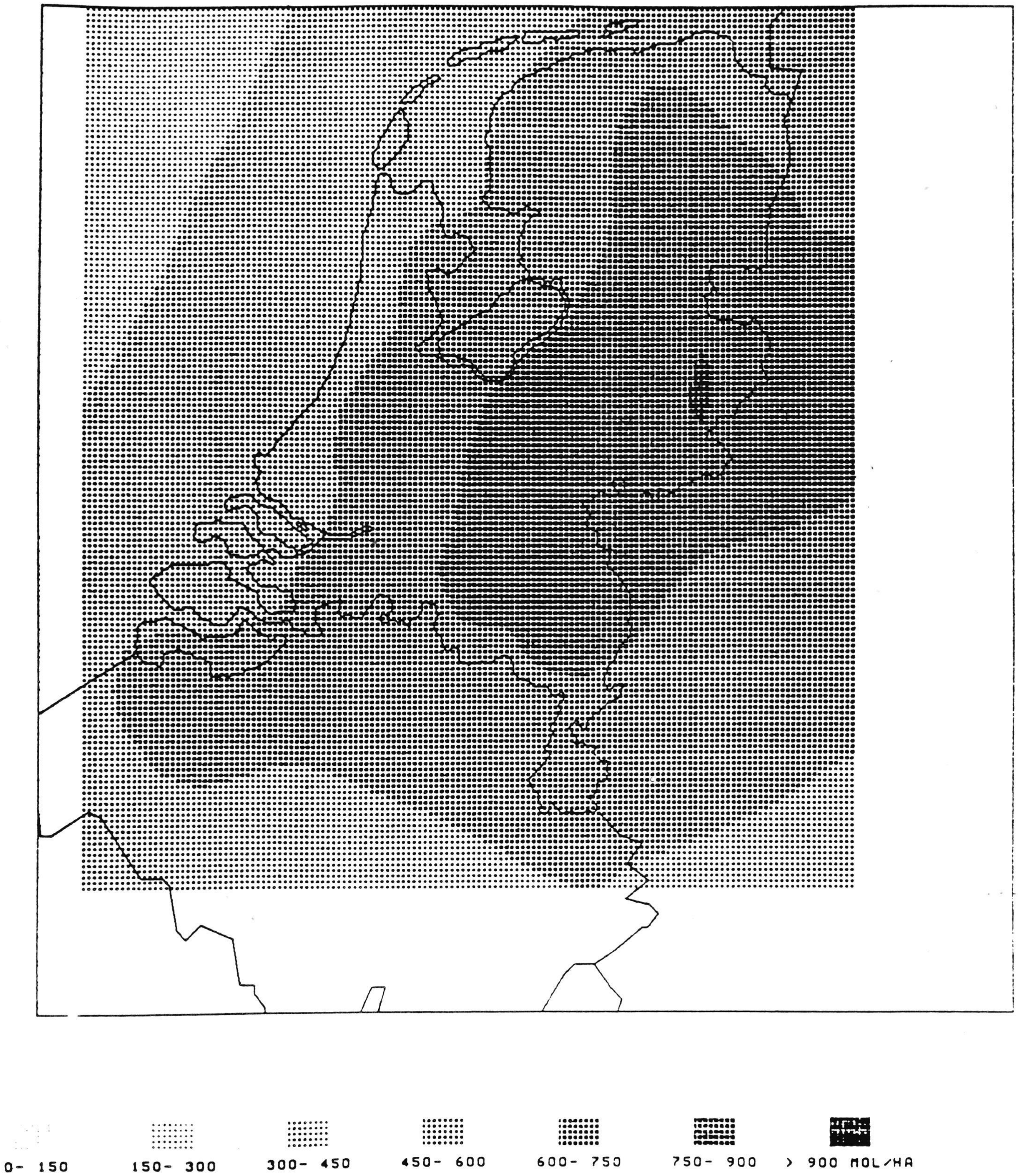


Figure 3 Calculated yearly average wet deposition of ammonia and ammonium (mol/ha)), from [1].

Table 1 Deposition of acids and acidifying components (in eq.H^+/ha/yr) in the Netherlands.

	SO_x	NO_x	NH_4^+ 1)	total
dry deposition	1860	890	1150	3900
wet deposition	890	420	790	2100
	2790	1310	1940	6000

1) Ammonium compounds are considered potentially acidifying; soil bacteria may convert ammonia into nitric acid.

The dry deposition velocity v_d is estimated on the basis of measurements of dry deposition velocities using several techniques. In this case the dry deposition fluxes are about 2/3 of the total acidic input into Dutch soils. From figures 2 and 3 it is clear that this ratio will vary over the country. Especially in source regions the amount of dry deposition may be even higher. On the other hand this ratio will be reversed for more remote areas in Europe where wet deposition may dominate the atmospheric input into the ecosystem.
In the next chapters an attempt is made to make an inventory of the factors affecting dry and wet deposition fluxes. From the above it is clear that, in any case, one important factor is the air concentration. The air concentration is of course a function of a large number of factors such as local meteorology, distance to sources etc., and all these factors will influence the deposition load into a certain ecosystem. For obvious reasons these factors will not be discussed here, but the reader is referred to the contributions on individual pollutants by D. Jost (SO_2), D. Fowler (NO_2) and R.G. Derwent (O_3).

2. WET DEPOSITION

Several mechanisms play a role in the wet scavenging process of gases and particles. In principle two routes are possible:

- rainout (in-cloud scavenging)
  In this mechanism pollutants in air are taken up in cloud-water.
  The uptake of gases in cloud-water is a function of their solubility. For some gases the total amount taken up in the liquid phase is determined by transformation processes in the liquid phase. Especially for SO_2 the oxidation in the cloud-droplet is important. The uptake of SO_2 (limited according to the Henry constant of SO_2) is enhanced by the oxidation of SO_2 in the liquid phase by H_2O_2, O_3 etc. to strong acids. The latter process causes a pH drop which in turn limits the uptake because a new equilibrium with the gas phase concentration of SO_2 will be established. Nitrogen oxides (NO, NO_2) are taken up in cloud-water mainly after transformation to HNO_3 and nitrates in the gas phase. The uptake of particles in cloud-water takes place because they act as condensation nucleï.
  The uptake of pollutants in cloud-water does not guarantee transport to the earth's surface. Many clouds evaporate ($\simeq$ 90%) and the pollutants are again suspended in air as aerosols or gases.

- wash out (below-cloud scavenging)
  In this process pollutants are taken up in raindroplets during their travel from the cloud to the earth surface. In some cases exchange of gases can take place in both directions i.e. evaporation of gases from raindroplets that have just passed through a layer of air with a higher concentration of the pollutant. For some gases (such as SO_2) the uptake may be increased in the presence of other pollutants (for example O_3, H_2O_2 in the case of SO_2). In this way the local pollutant concentration may become important for the amount of wet sulphate deposition. The transport time of the droplets however is relatively short and therefore oxidation in the droplets is less relevant than it is in clouds.

It is obvious that there is not much influence of the canopy on the amount of wet deposition. From an effect point of view of the difference in "hold up" (the amount of rain remaining in the canopy after a shower) between plant species may be relevant. The concentration of pollutants in water droplets on leaves may become extremely high when the droplets evaporate. For a certain location the amount of pollutant transferred via wet deposition is strongly dependent on the rain intensity. On a large scale the position of the receptor area with respect to source areas and the existence of predominant trajectories may be important.

3. OCCULT DEPOSITION

Occult precipitation is the process by which large aerosol particles (mist) are captured by vegetation. This process is especially relevant in coastal areas and at high altitudes. Except for these sites (where mist may occur very often) the pollutant input via this route into an ecosystem is not a very significant fraction of the yearly amount. On the other hand the concentration of acidic or oxidizing agents in the deposited water may be very high (during episodes) and the effect of occult precipitation may be larger than estimated on the bases of the yearly input. Sofar not much is known about the processes which play a role in mist deposition.

Dollard [3] shows that the deposition of fog droplets is very efficient and at wind speeds higher than 2 m/s limited by the resistance to aerodynamic transfer only. The process of occult deposition is therefore probably very sensitive to the wind speed and the surface roughness of the vegetation. The increased turbulence at a forest edge probably also causes the increased hydrological input by occult precipitation observed at forest edges [11] (see also § 4.1).

4. DRY DEPOSITION

4.1 Gases

The dry deposition flux (F) of a compound is proportional to the air concentration of the compound (c). The proportionality factor is the so-called dry deposition velocity v_d i.e.:

$$F = -v_d . c.$$

In this formula F is expressed for example in $g/m^2/s$, v_d in m/s and c in g/m^3.

Dry deposition is driven by the capture of suspended particles or gaseous material by the vegetation surface. Because of the roughness of the vegetation the air flow over it will be turbulent. This turbulence will bring air parcels from aloft to very close to the surface. Once an air parcel is in direct contact with the vegetation exchange of material may take place and gases or particles may be absorbed by the vegetation.

The final step in the uptake process can have several features. A gas molecule can be absorbed at the cuticula or taken up via the stomata. If the uptake in any form takes place the concentration close to the surface will decrease and a concentration gradient will develop. New material will be supplied from the air above and continuous transport will take place in opposite direction of the concentration gradient. The final uptake at the vegetation is of course crucial in the total process since when the gas or aerosol is not taken up there will be no deposition. Note, however, that although the uptake is crucial this does not mean that it is also the rate-determining step in the process.

The dry deposition velocity is a strong function of several parameters such as wind speed, temperature, surface roughness, biological state of the plant. Moreover for different chemical species-substrate combinations the dry deposition velocity can be completely different. It is often assumed that v_d is not a function of the air concentration, although this may sometimes be the case when a compound can be emitted as well as deposited (such as NO or NH_3). This complicating factor will not be considered here. A very useful instrument to look into factors that influence dry deposition, in a structured way is the surface resistance layer model. This model divides the deposition process into three subprocesses, namely:

1) transport through the layer of air above the surface
2) transport through the laminar boundary layer surrounding the vegetation elements
3) the final uptake at the vegetation surface.

Each layer represents a resistance against the transport of material analogous to Ohm's law. In this picture the flux of material across a layer is equivalent to the electrical current, whereas the concentration difference between the air and the vegetation is equivalent to the voltage difference across a resistance. The sum of these series of resistances presents a total resistance to transport from the air compartment to the vegetation.

So, similar to Ohm's law $c_{air} - c_{veg} = F.R_{tot}$.

When the gas is efficiently absorbed by the vegetation the concentration in the vegetation will be negligible compared with the concentration in the air, and from the definition of v_d it follows that

$$\frac{1}{v_d} = R_{tot} = R_a + R_b + R_c.$$

The aerodynamic resistance R_a is the resistance of the layer of air between some reference z and the air vegetation interface.

R_b is the resistance of a small quasi laminar boundary layer existing close to the vegetation elements.

The eventual resistance against uptake is the surface resistance R_c (or canopy resistance). The final uptake can consist of a number of parallel routes. In figure 4 an example of a resistance model is given. It can be seen that for gas molecules there are several routes for uptake, for instance through the stomata, followed by "bio"chemical conversion or physical binding of the molecule. Another route may be the dissolution of the molecule in a water layer on the vegetation leaves. As with the electrical analogue one of the parallel resistances can short circuit the other ones.

Using the structure of the model all factors affecting dry deposition fluxes will be treated in the sequel.

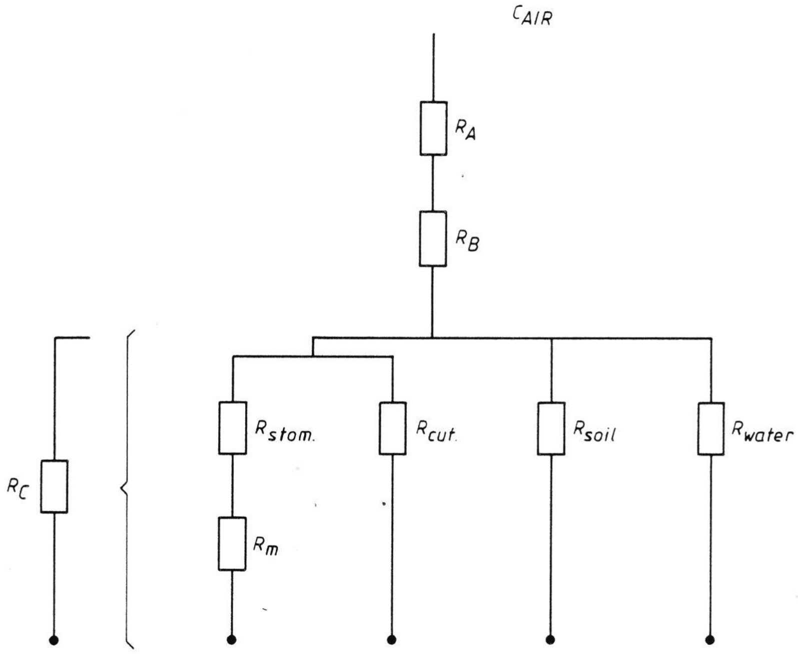


Figure 4 Simple resistance model for dry deposition.

R_a = aerodynamic resistance
R_b = resistance of the boundary layer
R_{stom} = stomatal resistance
R_m = mesophyl resistance
R_{cut} = cuticular
R_{water} = resistance of water surfaces
R_{soil} = resistance of soil

Aerodynamic resistance R_a

When the wind blows over a large flat terrain covered with vegetation a vertical wind speed gradient will be established. This gradient is caused by the drag force of the vegetation elements. Figure 5a shows such a wind speed profile. Under neutral conditions the wind speed at height z will be proportional to log (z). Neutral conditions occur in the atmosphere when there is so much cloud cover that there is no insolation or radiative cooling of the earth. When there is considerable insolation, turbulence will be increased because of buoyant forces acting on air parcels. This situation is called unstable and the wind speed gradient will be less steep than logaritmic. During the night when there is radiative cooling (on clear cloudless nights) the air parcels close to the ground will be cooled and will not have the tendency to rise and mechanically generated turbulence will be suppressed. In this situation very strong wind speed or concentration gradients can exist. The effect of stability on the logaritmic wind speed profile is accounted for in the following formula:

$$U_z = \frac{U_*}{k} \left(\ln \frac{z}{z_o} - \Psi_m \left(\frac{z}{L}\right)\right) \qquad (1)$$

where k is von Kármán's constant and Ψ_m is an empirical correction function for stability; Ψ_m is a function of one stability parameter only: the Monin-Obukhov length L. The Monin-Obukhov length contains all information to describe the stability situation in the atmosphere. Many important turbulent magnitudes are a simple function of the dimensionless number z/L (z in height above ground). The Monin-Obukhov length can be estimated from wind speed and temperature profiles or often with sufficient accuracy from synoptic meteorological information. U_* is the so-called friction velocity which by definition is equal to $\sqrt{-\tau/\rho}$, where τ is the surface stress (and momentum flux) and ρ the air density.

It is easy to understand that a rougher vegetation causes a larger drag force on the wind flow. In formula 1 the surface roughness is described by the roughness z_o. Essentially this is an integration constant and its value in a certain situation can easily be found by extrapolating the wind speed profile to the height were the wind speed theoretically would become zero. It is clear, however, that this value is related to the height and structure of the vegetation. Roughly z_o is of the order of magnitude of 10-20% of the vegetation height, for example the roughness length of forest is in the order of magnitude of 0.5-1 metre and grass is in the order of magnitude of 5-20 cm. Of course the architecture of the stand (especially the density) will also be important for the value of z_o (very dense canopies will be smoother). The resistance to momentum transfer is the ratio of the momentum concentration ($\rho\ U_z$) and the momentum flux ($\tau=-\rho\ U_*^2$), i.e.

$$R_a = \frac{U}{U_*^2} \tag{2}$$

The influence of z_o and the stability can be seen by rearranging (1) and (2) to

$$R_a = \frac{1}{k\ U_*} (\ln \frac{z}{z_o} - \Psi_m (\frac{z}{L})) \tag{3}$$

For gases the aerodynamic resistance from z to z_o can be calculated in a similar way introducing the integrated flux profile relation for gases $\Psi_c(z/L)$, i.e.

$$R_a = \frac{1}{k\ U_*} (\ln \frac{z}{z_o} - \Psi_c (\frac{z}{L})) \tag{4}$$

The turbulent processes that play a role in the dry deposition of gases are very similar to the processes that are important in the transport of sensible heat from the earth to the air and visa-versa.
As a consequence of this similarity Ψ_c is in practice often replaced by Ψ_h. Values for Ψ_h as a function of z/L are given in the literature. The influence of wind speed, surface roughness and stability on gas fluxes is therefore equal to the influence on heat fluxes. For example during inversion situations strong concentrations may develop.
In figures 5b and c an example of simultaneously measured vertical gradients of NH_3 and NH_4^+ over heather is shown. From these graphs the different behaviour of NH_3 and NH_4^+ ions in particles is obvious because under the same meteorological conditions NH_3 gives a much stronger gradient than NH_4^+. This leads to the conclusion that NH_3 deposits much faster than NH_4^+ particles.

In figure 6, taken from Duym [12] the aerodynamic resistance is presented as a function of the wind speed and surface roughness for several Pasquill stability classes. Class D is the neutral stability class (overcast skies, high winds). Classes E and F are nighttime inversion situations were turbulence and hence exchange is suppressed, whereas A, B and C are daytime sunny conditions with increased turbulence.

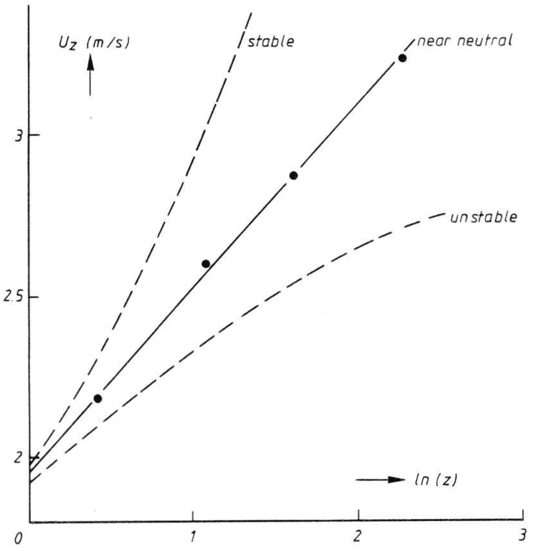
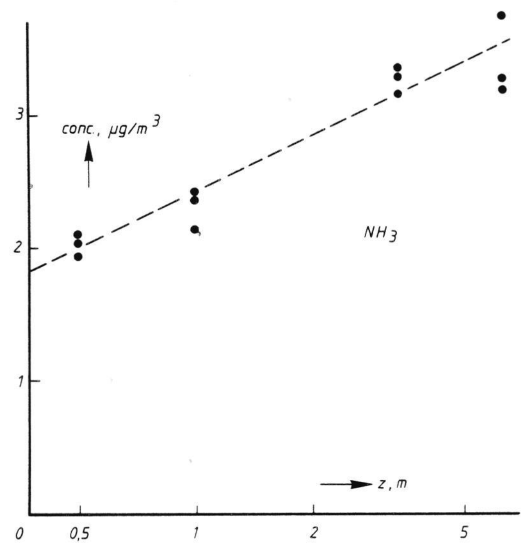


Figure 5a Half-hour averaged wind speed as a function of height above ground for different stabilities.

Figure 5b Ammonia concentrations as a function of height z over heather.



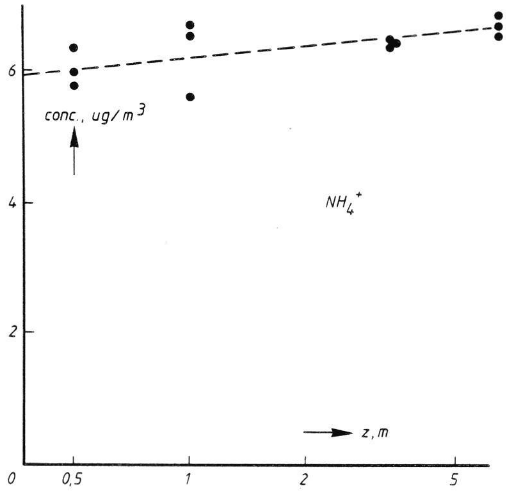


Figure 5c Ammonium concentrations as a function of height z over heather.

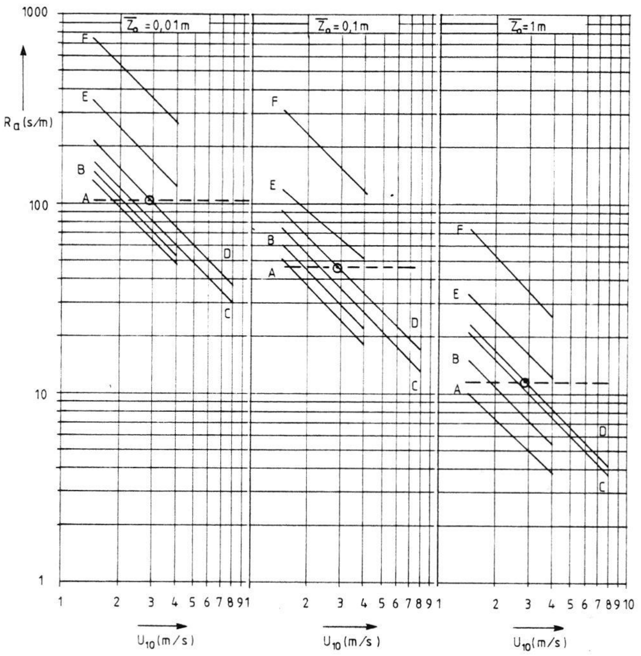


Figure 6 Aerodynamic resistance R_a and the dependence of wind speed U, roughness length z_o and for several stability classes (from [12].

From figure 6 the relevance of the different parameters can be seen. For example, by changing the value of z_o from 0.1 m (high grass) to z_o=1 m (forest), the aerodynamic resistance is lowered by a factor of four.
In a situation were a gas is very efficiently absorbed by the vegetation the deposition velocity is also increased by the same factor.
The influence of the wind speed is of the same order of magnitude: if the wind speed increases from 2 m/s to 8 m/s the aerodynamic resistance goes down by a factor of 4. The influence of stability is also dramatic: in a nighttime situation (F) the atmospheric resistance can be about an order of magnitude higher than under daytime conditions (A) with the same wind speed.
From the above it can be concluded that the deposition velocity at a certain location very much depends on local meteorology (average wind speeds, occurrence of different stability classes) and surface roughness. As a consequence its value will show daily and seasonal cycles caused by meteorological as well as biological cycles. For example the growing of leaves on decidious trees will significantly change the surface roughness and the deposition velocity.

Boundary layer resistance R_b

When a plant leaf is held in the direction of the mean flow of the wind the drag force of the leaf on the plant will be generated by molecular diffusion. Holding the leaf perpendicular to the flow will augment the drag considerably because of bluff body forces acting on the leaf. The transpiration of the leaf, however, will practically be the same in both cases, because the first stage in transport of water molecules from the leaf to the mean flow will be molecular diffusion in both cases.

The resistance to transfer of momentum is therefore always lower than the resistance to transfer of gas. This excess resistance is the resistance of the small laminar boundary layer (of a few mm) through which transport of gases or heat takes place by molecular diffusion.
It is easily understood that this boundary layer resistance is a function of the thickness of the layer and of the molecular diffusion constant of the species.
Very often the boundary layer resistance is calculated from:

$R_b = \frac{1}{U^*B}$, where B is the sublayer Stanton number i.e.

$B^{-1} = a(Re_x)^m (Sc)^n + b$, where Re is the surface layer Reynolds number; Sc is the Schmidt number. The empirical factors m and n are in the order of magnitude of 0.2 to 0.45 for m to -0.5 to -1 for n [4]. Typically, B^{-1} is of the order of magnitude of 4. The dependence of R_b of the diffusion coefficient (D) is weak; of the order of magnitude of $D^{2/3}$.
Typical values of R_b for forest are 5 s/m and for lower vegetation as heather 10 s/m.
Summarizing, the boundary layer resistance is a function of surface roughness, local meteorology and the molecular diffusion coefficient of the gas. The value of R_b is often relatively low compared to the value of R_a or the surface resistance R_c.

If the surface resistance is close to zero the quantity $1/R_a+R_b$ presents the maximum deposition velocity any compound can have under these circumstances. Fowler [5] estimated maximum depostion velocities over different vegetation types. In table 2 the values for grass, maize and forest are presented.

Table 2 Maximum deposition velocities over different vegetation types (after Fowler [5]).

	Height (m)	v_d (cm/s)
Grass	0.4	3.3
Maize	2.0	8.3
Forest	10.0	10.0

Figures of this kind can be used to judge the results of measurements of dry deposition velocities obtained using techniques such as throughfall or stemflow. The deposition velocity obtained from these measurements can never exceed the values calculated from wind speed, surface roughness and stability data. If v_d exceeds the maximum value the measurements are probably distorted by artefacts such as exchange processes (leaching of plant leaves) inside the canopy.
When using routine meteorological data to do these calculations one should realize that the 10 m wind speed measured on the routine site may be systematically different from the wind speed at the measuring site because of the different surface roughness. For instance over forest the average wind speed may be a factor of 1.4 lower than over a normal standardized measuring site [6].

Surface or canopy resistance R_c.

Finally the gas is absorbed at the vegetation surface. Several routes are possible for this uptake. A molecule can be taken up through opened stomata and subsequently converted inside the plant. Uptake via the cuticula is often not very important. For some, very soluble, gases (such as HNO_3 or NH_3) cuticular uptake may be especially important when the surface is wetted. In figure 4 these parallel routes are represented as parallel resistances. Derived from this picture the canopy resistance can be calculated from

$$R_c = [[R_s \, Sc \, Pr^{-1} + R_m]^{-1} + R_{cut}{}^{-1}]^{-1}/LAI$$

All resistances are per unit of leaf area.
R_s is the stomatal resistance controlled by stomatal opening. Factors affecting stomatal opening are the intensity of photosynthetically active radiation, the leaf temperature, the leaf water potential and the water vapour deficit. For obvious reasons the behaviour of the stomata is different for different biological species. Much experimental work has been carried out showing the influence of stomatal behaviour on the dry deposition of SO_2 and O_3. For O_3, however, the additional route through the cuticula cannot be neglected because even at night ozone is absorbed at the vegetation surface. The diffusion coefficient of the gas is accounted for by the introduction of the Schmidt number (Sc). The solubility of the gas in the cell liquid (Henry constant) is of course relevant. A mesophylic resistance R_m is used to account for the reaction path the gas may undergo after uptake. Unfortunately not very much information is available for R_m for the different gases.
For a wet canopy (wetted by dew, mist or often rain) the situation may be completely different because the stomatal route may be blocked.
For O_3, and NO_2 the dry deposition velocity is therefore probably lowered in the presence of waterfilms. In the case of SO_2 the uptake may be limited by the acidity of the waterfilm (at a pH of approximately 3). In this case NH_3 deposition can become relevant because this process will increase the pH. Although this effect seems reasonable from a chemical point of view, no outdoor measurements support this hypothesis up to now.
The influence of the leaf area index (LAI) on the canopy resistance is more complex than expressed by the model, because of its many different aspects: when the LAI increases, the number of stomata increases and the canopy resistance decreases as is accounted for by the model. However, when the top layer of the canopy is very dense, the lower leaves will be shaded and the stomatal resistance will increase lower in the canopy. Moreover the dense top will decrease the turbulent exchange coefficient in the canopy. All these effects cannot be accounted for in the present formulation of the surface layer model.
In principle the canopy resistance may be determined from laboratory fumigation experiments. However, the translation of these results to outdoor situations is a difficult task. Using micrometeorological techniques the canopy resistance can be determined directly from field measurements. As an example the results of the measurements of the dry deposition flux of NH_3 and O_3 over heather performed by our group, are given in figure 7. The contribution of the surface resistance to the total resistance can be seen from this graph. It is clear that heather much more efficiently absorbs NH_3 than O_3. In addition, it can be concluded that NH_3 is even better absorbed by a wet canopy.

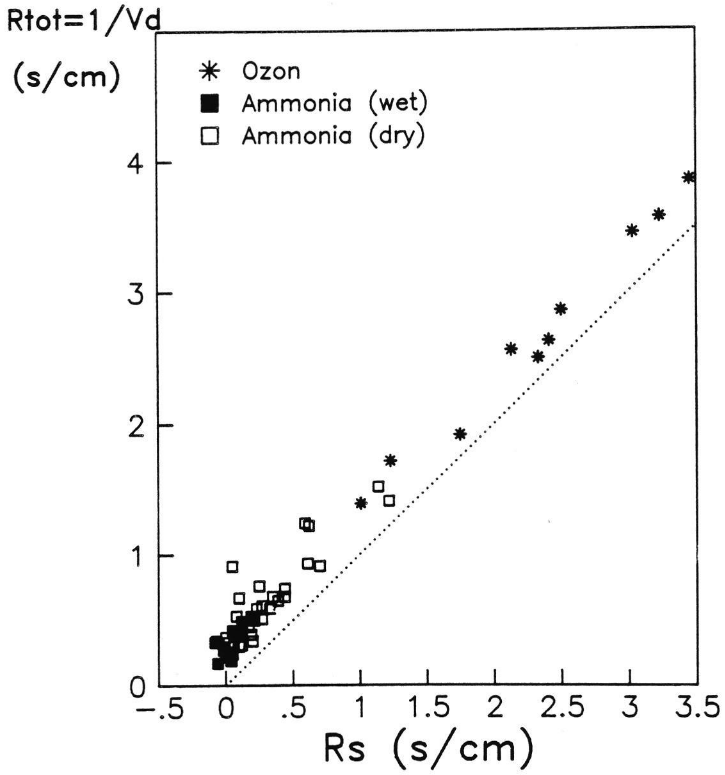


Figure 7 Total resistance to deposition ($1/v_d$) of O_3 and NH_3 over heather plotted against the surface resistance (R_s).

Typical values for the surface resistance of SO_2 and O_3 (and probably NO_2) are in the range of 1 s/cm and 2.5 s/cm. The surface resistance for NO is probably very high (up to 20 s/cm), whereas it is close to zero for HNO_3. For wet canopies the surface resistance for O_3 and NO_2 is probably around 10 s/cm.

Linking up all resistances gives a picture of the cycles the dry deposition velocity of certain compounds undergo during different periods.
Figure 8 presents data of this kind taken from Hicks [7]. This figure is plotted from measured values of the aerodynamic resistances and a model based on formula 4 for the surface resistance. The picture shows some features typical for dry deposition:

1. The diurnal cycle of the wind speed or the aerodynamic resistance shown in the profile of the HNO_3 deposition velocity which is only limited by the atmospheric resistance.
2. The different behaviour of the deposition velocity of different chemical species (compare HNO_3 with O_3 and NO_2).
3. The influence of the wet surface during a part of the night on the dry deposition velocity of SO_2.

Although much of the information that goes into the model is hardly verified quantitatively by experiments, it gives a clear picture of the factors relevant for gas dry deposition. Using the surface layer model, the influence of several cycles (annual, seasonal, daily) on individual processes (resistances) can be estimated from separate data.

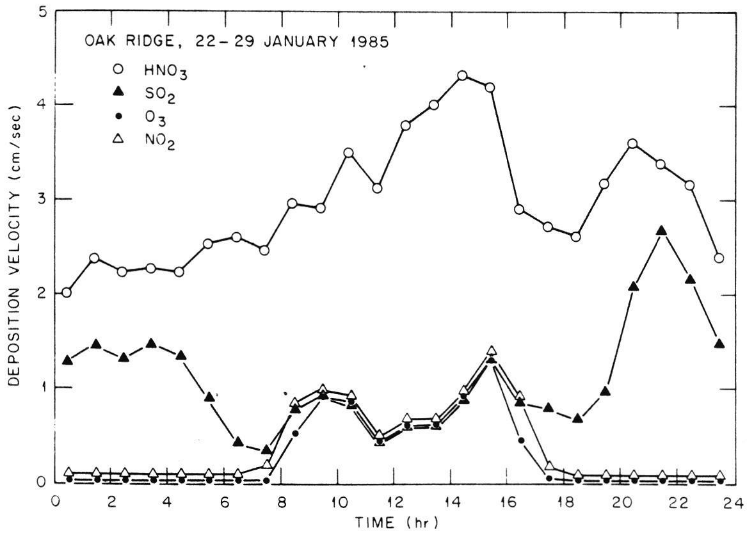


Figure 8 The weekly-average diurnal cycle of deposition velocity for HNO_3, SO_2, O_3 and NO_2, taken from Hicks et al. [7].

The use of the model is limited to flat homogeneous plant surfaces. Normally, however, natural vegetation is far from homogeneous and there are many transitions both in surface resistance and in surface roughness. If the surface resistances before and after the transition are known (from measurements over ideal sites) the problem can be solved simultaneously with the surface roughness transition (using second order closure models, see for instance Bosveld [8]).

In some forests with many open spots the amount of forest edges may form a significant fraction of the forest and it is relevant to study the effects of edges separately. Grennfelt [9] obtained interesting results from measurements of the throughfall on several distances from the forest edge. His measurements show a sharp decline in the amount of pollutant throuhgfall going from the edge into the forest. Many processes may explain this result but a possible cause for this effect is also the increased turbulence over the edge resulting from the adaptation of the wind profile to the new (higher) surface roughness of the forest. Close to the forest edge the wind speed, and by consequence the turbulence intensity, is higher than is normal over such a rough terrain (when the wind is fully adapted) and will decrease with increasing distance from the edge into the forest. In any case these results show clearly that the choice of the measuring site for throughfall measurements is very important.

The present formulation of the model is probably also not suitable to describe the processes taking place during the deposition of reactive compounds such as O_3 or NO. These compounds may undergo (photochemical) reactions during transport and hence their flux will not be constant with height. The constancy of the flux with height is a basic prerequisite for the use of the simple resistance layer model.

4.2 Dry deposition of particles

Particles are transported from the air to the earth's surface by turbulent diffusion (like gases) or by sedimentation. Sedimentation is only relevant for large, heavy particles or in cases were turbulence is very low. However, if one is interested in pollutants present in heavier particles, sedimentation may be important. The viscous sublayer is passed by several mechanisms such as Brownian diffusion (only relevant for particles smaller than 0.1 μm), sedimentation, impaction and phoretic effects. What mechanism is dominating for a certain aerosol highly depends on the particle diameter. The sedimentation velocity is a function of the aerosol diameter and density: $v_s \sim \rho d^2$. Inside the canopy sedimentation will be important (because the wind speed is low) and particles sediment on horizontal leaves. Therefore sedimentation inside the canopy is sensitive to the LAI.

Interception occurs when a particle is about to pass an obstacle at a distance closer than its dimensions. This process is relevant only when the element and the particle have comparable dimensions. When particles are not able to follow the small-scale streamlines close to vegetation elements because of their inertia they will diverge from the streamlines and impact on the leaves. Once a particle has reached the surface it depends on the possibility of absorbtion of its momentum whether the particle will remain there or bounce off.

For particles smaller than 5 µm bounce off is probably not very important. For these particles the presence of hairs on the leaves is important because hairs thicken the boundary layer and slow down the particle before the impact. A slow particle will not bounce off very easily.

The stickiness of the leaf (caused by wax or surface water) is relevant for larger particles.

Another relevant aspect is the influence on the dry deposition of hygroscopic particles exercised by vertical moisture gradients over the vegetation. When the relative humidity near the surface is higher than aloft a particle may grow very fast when it comes in contact with this humid air and deposit easier because of its higher inertia.

Adding up all processes gives the well-known picture of the dry deposition velocity of particles as a function of particle size. An example of this dependency taken from [2] and based on a model described by Sehmel [10] is shown in figure 9. The minimum dry deposition velocity around 0.1 µm is consistent with the observed atmospheric residence time of these particles. This particle size range is relevant because it carries most of the atmospheric sulphate, nitrate, ammonium etc.

The dependency of the dry deposition velocity can be studied from the same model formulation. For instance in figure 10 the influence of the surface roughness is presented [2].

So far measurements of dry deposition velocities have not shown very coherent results. One important problem especially with micrometeorological techniques is the low deposition velocity of particles in the important size region. Another problem may be the (re)suspension of (deposited) particles. Because of the influence of the microstructure of the leaf surface the use of artificial collectors to measure dry deposition does not seem very valuable except perhaps for very large particles. Throughfall and stemflow measurements still suffer from problems related to canopy exchange.

At the moment there is no best model available by which all experimental results can be explained in a satisfactory way. In a qualitative sense, however, it is clear that the dry deposition velocity of particles is a strong function of particle diameter, turbulence (and hence wind speed and especially surface roughness), leaf area index and plant species (especially with respect to leaf microstructure and stickiness). The uncertainty in dry deposition velocity of particles is at the very least a factor of two.

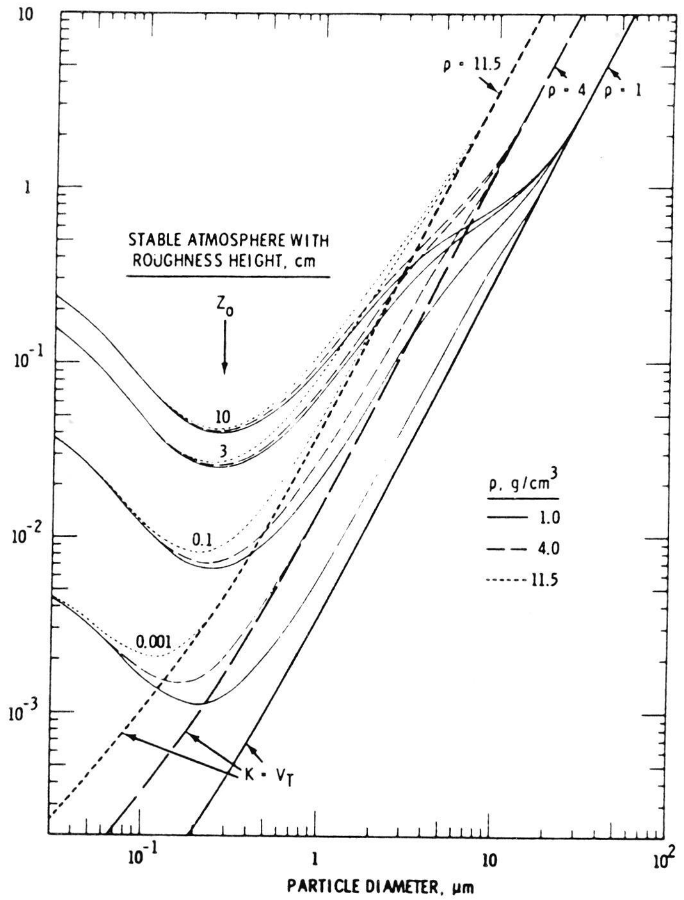


Figure 9
Dry deposition velocity of particles as a function of particle diameter for different particle densities, roughness length (after Sehmel [10]).
The lines with $K=V_T$ represent the sedimentation velocity.

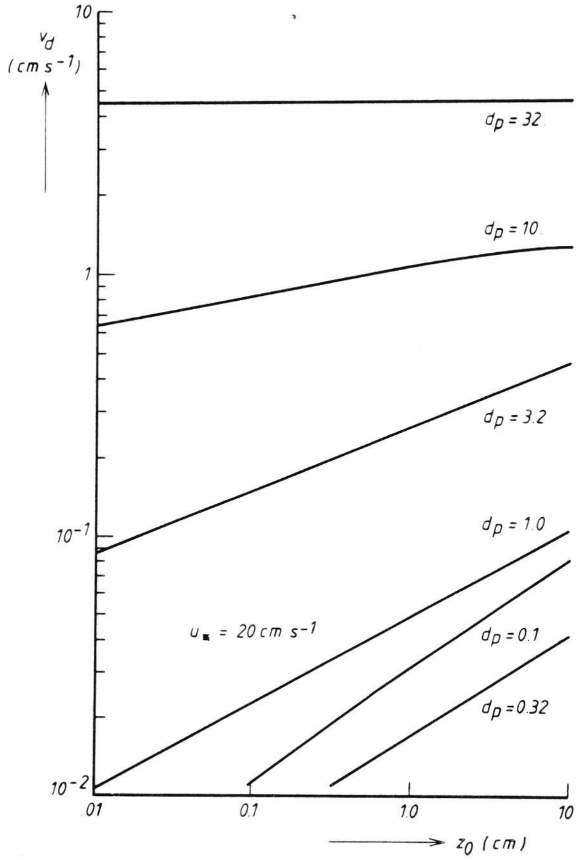


Figure 10
The deposition velocity of particles as a function of particle diameter according to the model of Sehmel and Hodgson [10]; friction velocity 20 cm/s.

5. CONCLUSIONS

For many pollutants deposition is the main route by which they are removed from the atmosphere. Other pollutants are deposited after chemical conversion. Two main routes can be identified i.e. wet deposition and dry deposition. A third route "occult deposition" (deposition of pollutants from mist or cloud water) is also important for the effects on plants at sites where heavily polluted mist is frequent.

In any case the amount of deposition entering an ecosystem is strongly related to the concentration of pollutant in the air. The amount of dry deposition is even directly proportional to the pollutant concentration in the air. For wet deposition the relation is less direct. The amount of pollutant in wet deposition at a certain site is related to the amount of rainfall at that site, the trajectory of the air mass, and the pollution level during the travel of the air mass. Because interception of large fog or cloud droplets is a very efficient process the amount of occult deposition is strongly related to the vegetation structure (roughness) and the wind speed.

Dry deposition of gases is a relatively well understood process. The large number of measurements reported has led to the construction of surface layer models that seem to describe rather adequately the deposition of gases. Although these models have not yet been verified extensively the influence of several subprocesses can be identified.

The absorption of SO_2, O_3 and probably NO_2 is dependent on a large number of meteorological factors such as wind speed, surface roughness and atmospheric stability. On the other hand uptake by the plant is also controlled by the stomata and therefore the uptake shows several cycles associated with stomatal opening and closure. For O_3 cuticular uptake seems to be relevant at night when stomata are closed. This situation may change when the plant surface is wetted by dew or rain. A waterfilm may block the stomata and decrease the uptake of O_3 and NO_2. For SO_2 the uptake in waterfilms will be enhanced theoretically until the pH reaches a certain limit. For very reactive gases such as NH_3, HNO_3 and HF absorption on wet as well as dry surfaces will be very efficient. The deposition velocity of these gases is limited only by the rate at which they can be supplied from the air above. Therefore the deposition rate of these gases is a strong function of surface roughness, wind speed and stability and less of the stomatal condition.

Although there is general consensus as to what processes play a role in particle dry deposition the present knowledge does not allow precise estimation of the deposition flux of pollutants associated with particles. The deposition velocity of particles is a very strong function of the diameter. Large particles (>10 μm) are transported to the vegetation by sedimentation, smaller particles mainly by turbulent diffusion (like gases). Close to or in the canopy other processes such as interception, impaction, sedimentation and Brownian diffusion are the dominant processes. In a qualitative way it is clear that dry deposition of particles is influenced by micrometeorological parameters such as wind speed, surface roughness and stability. The microstructure of the vegetation (hairy leaves, waxiness of leaves, leaf area index) is also important.

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