Trace Gas Exchange Above the Floor of a Deciduous Forest

2. SO2 and O3 Deposition

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The pollutant uptake capacity of the surface beneath forest canopies is poorly understood. In this study we show that the eddy correlation method can be applied within a canopy. Direct measurements of SO2 and O3 deposition to the forest floor of two deciduous forests were made with the eddy correlation method. For dry periods the surface uptake resistance for SO2 was of the order of 700 s m$^{-1}$. When the soil and litter were moist, the SO2 surface uptake resistance was small, resulting in deposition velocities at the forest floor that were about 2 times greater than during dry periods. SO2 deposition to the forest floor constitutes between 20 and 30% of total uptake to the forest ecosystem when the surface is moist. Soil surface uptake resistances for O3 were about 2000 s m$^{-1}$ for both wet and dry periods.

INTRODUCTION

Recent assessments of the total deposition (wet and dry) of nitrates and sulfur compounds suggest that dry deposition can comprise between 30 and 70% of the total atmospheric input (not including cloud water interception) to ecosystems [Meyers et al., 1991; Siros and Barrie, 1988]. These assessments are based on inferred deposition rates that are derived from measurements of ambient pollutant concentrations and estimates of the deposition velocity ($V_d$). With this approach, the flux ($F$) is determined as $F = V_d(z)C(z)$. Various schemes have been used to estimate $V_d$. In simple schemes, $V_d$ is specified according to the land use and time of year [Wesely, 1989]. In more detailed schemes like those described by Hicks et al. [1987] and Wesely and Lesht [1989], the deposition velocity for the various pollutants of interest are modeled using standard meteorological data and detailed information about the site. This includes an estimate of the leaf area index (LAI) and specification of the various plant species that comprise a particular site. Deposition of pollutants to ecosystems includes not only uptake by the vegetative components but also uptake by the ground surface that lies beneath the canopy. The ground surface is not usually bare soil (with the exception of tilled agricultural systems) but is covered with detritus from decaying organic matter and understory vegetation. The chemistry which governs the overall sink potential for a surface also depends on the amount and existence of either liquid or frozen (snow) H$_2$O covering the surface. For example, Cadle et al. [1985] found that the SO2 deposition velocity over snow depends on the snow surface temperature. Despite these effects the surface resistance is generally assumed to be a static quantity.

The diffusion of material from the atmosphere to soil surfaces takes a tortuous path that is comprised of three pathways: from the atmosphere (concentration $C_a$) to just outside a quasi-laminar layer above the ground surface (concentration $C_b$), across an unsteady viscous layer to the soil surface (concentration $C_{stfc}$), and finally from surface to reaction sites within the soil surface (concentration $C_i$). This framework for transport is analogous to flow through an electrical circuit in which the three serial pathways can be represented as resistances to diffusion across concentration differences. That is,

\[ \text{Flux} = \frac{C_a(z) - C_b}{R_a(z)} = \frac{C_b - C_{stfc}}{R_b} = \frac{C_{stfc} - C_i}{R_s} = \frac{C_a(z) - C_i}{R_a(z) + R_b + R_s} \] (1)

$R_a$ is the atmospheric diffusive resistance and is characterized by the level of vertical turbulent mixing. The near-surface transport, which is mainly controlled by molecular diffusion processes is characterized by $R_b$. The surface uptake resistance ($R_s$), also called the bulk canopy resistance, quantifies the overall pollutant uptake capacity for a given surface or surfaces, including both soil and vegetative components. This is generally not constant as it can depend on surface wetness, and how the surface was wetted (rainfall or dew). Equation (1) reduces to the commonly used deposition velocity concept, where $F = (R_s(z) + R_b + R_a)^{-1}C_a(z) = V_d(z) \cdot C_a(z)$ when the internal pollutant concentration ($C_i$) is assumed to be zero.

Measurements of SO2 uptake by calcareous soil surfaces have been reported to be large [Garland, 1977], with surface uptake resistances that were not significantly different from zero. No significant effect of soil moisture was observed. In laboratory experiments, Abelles et al. [1971] and Terraglio and Manganelli [1966] found that SO2 uptake was strongly influenced by the availability of soil moisture. Average deposition velocities ranged from 0.3 to 0.6 cm s$^{-1}$ for wet soils compared to 0.2 cm s$^{-1}$ for dry soils.

Garland [1977] found an increase in the deposition velocity to soils with higher pH. Payrissat and Belkhe [1974] report that a pH increase from 4.5 to 7.7 corresponds to a 10-fold decrease in the soil surface resistance to SO2 uptake. Petit et al. [1977] found that the soil surface resistance ranged between 300 and 400 s m$^{-1}$ for soils with a pH of 3 and were roughly a factor of 10 lower for soils with higher values of pH.

Early studies suggested that deposition to soils beneath vegetation was relatively small compared to the uptake of an active canopy. For example, Fowler and Unsworth [1979]...
estimate that the deposition to the soil beneath a cereal canopy is only 5 to 10% of the total flux to the agricultural plot. However, direct measurements of sulfur deposition to the forest floor have yet to be made.

Deposition rates of similar magnitudes to soils have been found for O₃. In laboratory experiments, Turner and Waggoner [1973] found surface uptake resistances for cultivated and sandy loam soils were of the order of 0.4 s cm⁻¹, with lower resistances reported for dryer soils. From the same study, the combined resistances below a height of 2.5 cm above the soil surface yielded a total resistance of about 200 s m⁻¹ (i.e., \( V_d = 0.4 \text{ cm s}^{-1} \)). An analysis by Galbally [1971] yielded similar results with combined resistances that were of the order of 150 s m⁻¹. Using measured gradients above a tobacco field and accounting for uptake by the vegetative elements, Leuning et al. [1979] estimated the soil surface resistance to be 100 s m⁻¹, which accounted for 50% of the total flux measured above the tobacco field. Much larger resistances (1000 s m⁻¹) were reported by Wesely et al. [1981] from measurements obtained over cold bare soil saturated with water, which yielded a maximum \( V_d \) of \( \approx 0.1 \text{ cm s}^{-1} \).

In any case, uptake of SO₂ by the ground surface can constitute a significant fraction of the overall deposition to an ecosystem. Dry deposition to the soil or litter surface represents a component of the total deposition that cannot be measured using either leafwashing or throughfall methods. Models which are used to infer seasonal and annual deposition rates of O₃ and SO₂ have relied upon the aforementioned studies to parameterize the sink capacity of the forest floor for pollutants. However, actual measurements of dry deposition to ground surfaces within forests are lacking. The direct input to soils is important in the characterization of biogeochemical recycling in forest ecosystems. As a result, several experimental investigations were conducted to evaluate the role of the forest floor as a sink for O₃ and SO₂. Results from experiments conducted at two research sites are presented and discussed. The effect of water availability on the magnitudes of the dry deposition rates is also explored. The implications regarding the effect these results have on estimated seasonal and annual SO₂ deposition budgets are discussed.

Site Characteristics and Data Acquisition

Walker Branch Watershed, Oak Ridge, Tennessee

The data and analysis presented in this report were taken from three studies. The first study period ran from April 29 to May 10, 1988. The second experiment was conducted between September 14 and 30, 1988. Both of these experiments were conducted at Walker Branch Watershed, located on the U.S. Department of Energy’s Reservation near Oak Ridge, Tennessee (35°57'30"N, 84°17'15"W). The field site is located on a gentle ridge in moderately complex terrain at 365 m above mean sea level. The site is a forested, mixed, uneven-aged stand of oak and hickory (spp. Quercus, Carya) with some small patches of loblolly pine (spp. Pinus) dispersed within the area. The average tree height is about 23 m with LAI of 5 [Hutchison et al., 1986] during years that are not affected by drought. The first experimental period was preceded by a multiyear drought that resulted in a lower LAI (3.2) than what is normally expected (4.9) for this forest.
the first experiment at Oak Ridge, the soil and litter layer was moist with four rain events occurring over the course of 10 days. The soil gravimetric water content was never less than 0.3 g g⁻¹. During the fall study, the soil was moist at the beginning of the experiment (θₑ = 0.23). With only 9 mm of rain over a 12-day period, θₑ dropped to 0.14 g g⁻¹.

Huntington Forest, Newcomb, New York

The third study was conducted during July 20–30, 1990, at Huntington Forest (43°59'N, 74°14'W). Located near Newcomb, New York (Essex County), Huntington Forest is a property of the State University of New York, College of Environmental Science and Forestry. As part of the Adirondack State Park of New York, the site is used as a research facility for long-term studies of wildlife ecology, forest ecosystem biogeochemical cycling studies, and silviculture. The elevation within Huntington forest ranges from 466 to 806 m. The site can be classified as a northern deciduous forest dominated by Acer saccharum Marsh. (sugar maple) and Fagus grandifolia Ehrh. (American beech). The leaf area index in the area where the eddy flux measurements were made is about 6.5 (70% maples, 30% beech) with an average tree height of 21 m. The total woody biomass (stems, branches, boles) is about 190 kg m⁻².

The soil at Huntington Forest is classified as a coarse-loamy, mixed, frigid, Hapludalf. The soil profile (about 1 m in depth) overlies a bedrock of gneiss which contains a hardpan derived from parent material of glacial till. The soil contains a considerable fraction of coarse fragments with high organic matter concentrations in the mineral horizons. The pH of solution in the litter layer is 4.42 and increases to 4.9 at a depth of 50 cm [Johnson and Lindberg, 1992]. Two instrumented towers are currently in use at the Huntington Forest research site. A 30-m triangular tower (18-inch base), instrumented with meteorological and chemical samplers, has until recently been operated as part of the Integrated Forest Study (IFS) [see Lindberg and Johnson, 1989]. A walk-up scaffolding tower (38 m) was recently erected for the purpose of making micrometeorological measurements.

The topography at Huntington Forest is considered complex. In the azimuth window from about 90° to 180° the terrain is moderately rolling and the land use is generally homogeneous (hardwood forest) for at least 2 km. This window was considered the best possible fetch. To the north, the elevation increases significantly. There is also a large body of water (Arbutus Pond) located about 300 m northeast of the tower. An eddy correlation system was operated at 2 m above the forest floor on a portable platform. Air temperature and the three components of the wind vector were determined with a commercial version of the sonic anemometer/thermometer described by Coppin and Taylor [1983]. Water vapor density was measured with an open path, fast response infrared gas analyzer [Auble and Meyers, 1992]. Fast response measurements of O₃ were determined with a chemiluminescent method based on the design by Ray et al. [1986]. Experiments conducted earlier indicate a frequency response of better than 5 Hz. Measurements of SO₂ were in a similar fashion as previously described. Mean concentrations of SO₂ and O₃ (2.0-m Teflon intake lines) were determined with a Monitor Labs model 8850 and a TECO (model 49), respectively.

The analog signals from the fast response instrumentation were sampled and digitized at 20 Hz (Campbell Scientific Instruments, model 21X, Logan, Utah) before being sent to a desktop computer via RS-232 communication protocols. Short-haul modems were used to insure the integrity of the RS-232 signals over a distance of 200 m. The incoming data were written to portable storage media for post processing.

Determination of Vertical Fluxes

Vertical turbulent fluxes were computed in real time using a digital recursive filter (200-s time constant). Tests have shown that the computed flux is not sensitive to the choice of the filter time constant [see Baldocchi et al., 1988]. The recursive filter allows for a determination of average values from which the instantaneous values were subtracted to obtain the fluctuations. The real-time vertical turbulent eddy fluxes were determined as

\[
\frac{w'\chi'}{n} = \sum_{i=1}^{n} \frac{(w - \langle w \rangle)(\chi - \langle \chi \rangle)}{n}
\]

where \(w\) is the vertical velocity and \(\chi\) is the scalar of interest. Here, the bracketed quantities denote a running mean that is being subtracted from the instantaneous values. The averaging period of 30 min was used during all experiments. For experimental sites that are not perfectly flat, the coordinate system of the sonic anemometer can be mathematically rotated to obtain a zero mean vertical and transverse velocity (\(w = \chi = 0\)). This is usual practice to correct for small misalignment errors of the sonic anemometer as well as to determine the vertical turbulent flux across the mean horizontal streamline, which generally follows the contour of the land surface. Since the fast O₃ and SO₂ analyzers were not open path systems but were fitted with sampling tubes with intakes in the vicinity of the sonic anemometer, a 2-s time lag was introduced. Computer software compensated for these time lags during data acquisition and in the calculation of the fluxes.

RESULTS AND DISCUSSION

Micrometeorological Considerations

The validity of the flux measurements at 2 m above the forest floor can be addressed by examining the heat balance. That is, on average, do the measured sensible (H) and latent energy (LE) fluxes balance the sum of net radiation (R_n) and ground storage (S) fluxes? For the first two experiments at Walker Branch Watershed, Baldocchi and Meyers [1991] concluded from independent measurements of all the components that the energy balance was closed. The energy balance residual was not significantly greater than zero. Evaporation from the forest floor with a wet litter surface often exceeded 40 W m⁻² and was closely linked to available energy (\(R_n - S\)). For dry surface conditions, evaporation was well correlated but weakly driven by available energy and LE values never exceeded 40 W m⁻².

During the experiment conducted at Huntington Forest, net radiation was measured above the canopy but not at the forest floor. For proper measurements of radiation within forest canopies, traversing tram systems are required to
the large scales to penetrate to the lower canopy. However, a recent study by Shaw and Zhang [1992] suggest that turbulence in the lower canopy levels is generated mainly by pressure perturbations created aloft.

A slope of +1, which is observed for the data from the SO2 analyzer, is typical of a white noise spectrum when $nS(n)$ is plotted against $n$ on a log-log plot. This feature is expected when trace gas analyzers are operated during periods when the ambient concentrations are low and electronic noise represents a sizable fraction of the signal variance [Wesely and Hart, 1985].

The normalized cospectral estimates ($nC_{wx}(n)$) between the scalar quantities and the vertical velocity $w$, in general, show spectral peaks that occur at or near the same frequency. This includes the cospectral estimates for SO2 (Figure 2) which for the same periods, had a signal that was dominated by noise (Figure 1). Although the noise constitutes a sizable fraction of the overall variance for SO2, the cospectral estimates of SO2 with $w$ (i.e., the flux) are consistent with the spectral estimates from the more noise-free scalars (H2O and CO2) since the noise that is superimposed on the signal does not correlate with $w$. This illustrates the noise rejection capability that is inherent in the eddy correlation method. The strong similarity in the normalized cospectra for the fast response open-path analyzers (H2O and CO2) and the closed-path slower-responding sensors (SO2 and O3) demonstrates that these sensors were adequate for flux measurements within the canopy. The flux contribution at frequencies $>1$ Hz is minimal.

The two prominent peaks observed in Figure 2 suggest a strong similarity in the turbulent transfer mechanisms for scalars with upward fluxes (H2O and CO2) as well as for trace species that are taken up by the surface (O3 and SO2). From Figure 2 the majority of the covariance occurs at frequencies less than 0.05 Hz, indicating that a large fraction of vertical fluxes are dominated by infrequent coherent turbulent structures. This supports the hypothesis suggested by Shaw and Zhang [1992] that turbulence in the subcanopy trunk space is a consequence of pressure perturbations imposed on the canopy from above. These pressure fluctuations are of lower frequency than velocity [Sigmon et al., 1983], resulting in a larger fraction of the power and cospectral energy occurring in the low-frequency range.

![Fig. 1. Power spectral estimates (nS(n)) for H2O, CO2, O3, and SO2. Units are (g/m$^3$)$^2$ s$^{-1}$ for H2O, (mg/m$^3$)$^2$ for CO2, ppbv$^2$ s$^{-1}$ for O3, and (μg/m$^3$)$^2$ s$^{-1}$ for SO2.](image)

![Fig. 2. Cospectral estimates ($C_{wx}$) of H2O, CO2, O3, and SO2. Cospectra are normalized by the average flux for the period.](image)
TABLE 1. Description of the Quadrants

<table>
<thead>
<tr>
<th>Quadrant</th>
<th>Plane, w - c</th>
<th>Emission, $\bar{w}'c' &gt; 0$</th>
<th>Deposition, $\bar{w}'c' &lt; 0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>($w' &gt; 0$, $c' &gt; 0$)</td>
<td>burst/ejection</td>
<td>outward interaction</td>
</tr>
<tr>
<td>2</td>
<td>($w' &gt; 0$, $c' &lt; 0$)</td>
<td>outward interaction</td>
<td>burst ejection</td>
</tr>
<tr>
<td>3</td>
<td>($w' &lt; 0$, $c' &lt; 0$)</td>
<td>sweep/gust</td>
<td>inward interaction</td>
</tr>
<tr>
<td>4</td>
<td>($w' &lt; 0$, $c' &gt; 0$)</td>
<td>inward interaction</td>
<td>sweep/gust</td>
</tr>
</tbody>
</table>

Quadrant Analysis

Conditional sampling is another analysis technique that is used to evaluate the turbulent events and processes that contribute to the time average vertical covariance. In particular, contributions from infrequent turbulent events such as a downward sweep of fast moving air or an ejection of slower air moving away from the surface can be evaluated. Most applications of quadrant analysis to canopy flows have focused on the structure of the kinematic Reynolds stress. For this quadrant analysis, the scalar fluxes of H$_2$O, CO$_2$, O$_3$, and SO$_2$ from the Huntington study will be examined.

In quadrant analysis, individual components of the vertical turbulent scalar flux ($\bar{w}'c'$) are examined on a $w'$-$c'$ plane, where the y axis represents positive and negative $w'$ fluctuations and the x axis represents the positive and negative c fluctuations. For scalar fluxes the definitions of the quadrants depend upon whether the flux is positive (surface is a source) or negative (surface is a sink). Table 1 describes the quadrants.

From the raw data time series, the relative importance of each turbulent event is assessed from

$$H = |\bar{w}'c'|/|\bar{w}c'|$$

where $H$ represents a hyperbolic hole size in the $w'$-$c'$ plane. By increasing the value of $H$, the importance of individual turbulent events relative to the mean can be determined. Following the notation of Shaw et al. [1983], the stress fraction $S_{i,H}$, for quadrant i and hole size $H$, can be determined as

$$S_{i,H} = \langle w'c' \rangle_{i,H}/|\bar{w}c'|$$

where

$$\langle w'c' \rangle_{i,H} = \frac{1}{T} \int_0^T w'c'(t)I_{i,H}(t) \, dt$$

$I_{i,H}$ is equal to 1 if the product $w'c'$ lies in the $i$th quadrant and $|w'c'| \equiv H|\bar{w}'c'|$. Otherwise, $I_{i,H}$ is set equal to 0. When the stress fractions are normalized by the average covariance, then

$$\sum_{i=1}^{4} S_{i,0} = 1$$

Similarly, the fraction of time ($F_{i,H}$) for each flux fraction ($S_{i,H}$) is determined as

$$F_{i,H} = \frac{1}{T} \int_0^T I_{i}(t) \, dt$$

For all the gases (H$_2$O, CO$_2$, O$_3$, and SO$_2$), at least 50% of the total flux consists of turbulent events that are 10 times greater ($H = 10$) than the mean flux for the period (Figure 3). Yet, these events occur less than 10% of the time during the averaging period (Figure 4). Ejection and sweep events comprise at least 75% of the turbulent events for values of $H > 10$. Almost identical results were obtained by Baldocchi and Meyers [1988a, b] from a momentum flux study conducted at Oak Ridge with measurements at 2 m above the forest floor. Similar results were obtained by Shaw et al. [1983] for momentum flux within a corn canopy. In their study, about 50% of the momentum flux occurred from events ($H > 4$) that occupied only 6% of the time in the averaging period.

SO$_2$ Deposition to the Forest Floor

The SO$_2$ deposition velocities from all the experiments were classified into “wet” and “dry” periods. Soils with gravimetric water contents less than 0.15 were considered dry. The deposition velocities ($V_d$) at 2 m above the forest floor, shown in Figure 5, are plotted against the standard deviation of the vertical wind velocity ($\sigma_w$) measured at 2 m above ground level. From Figure 5 it is evident that the deposition velocities measured during the dry periods are significantly lower than those from the other periods, even though turbulent mixing (as indicated by $\sigma_w$) tended to be greater during the dry periods. Because the surface uptake resistance for SO$_2$ appears to be small for moist soil conditions, the level of turbulence in the canopy flow field becomes a controlling factor in characterizing the deposition rate. The strong dependency of deposition on the level of turbulence is observed for HNO$_3$ [Huebert and Robert, 1985; Meyers et al., 1989], which is known to have a negligible surface uptake resistance when compared to the aerodynamic and boundary layer terms. For significant surface resistances, as evident in the dry periods, the depo-
position velocities are relatively insensitive to the local turbulent flow field.

The large deposition velocities observed during moist surface conditions indicate a negligible surface resistance for uptake for SO₂. If we assume that \( R_s = 0 \) for moist conditions, an estimate of the SO₂ surface uptake resistance for dry periods can be obtained. For periods when the surface was wet, and assuming \( R_s \ll R_a \) or \( R_b \), the deposition velocity can be determined as \( V_d = (R_a + R_b)^{-1} \). From Figure 5, \( V_d \) can be approximated as a linear function of \( \sigma_w \). By including this parameterization of \( V_d \) (and therefore \( R_a + R_b \)) into a complete resistance equation, the residual or surface uptake resistance can be determined. Upon substitution, the average surface uptake resistance for dry periods was \( 660 \text{ s m}^{-1} \). This finding is consistent with the laboratory studies by Payrissat and Beilke [1975] who found surface uptake resistances of the order of \( 400 \text{ s m}^{-1} \) for soils of similar pH. Garland [1977], using the gradient method inside a wind tunnel, determined the surface uptake resistance for a dry, low pH soil to be \( 0.4 \text{ cm s}^{-1} \), much lower than the numbers reported here. The deposition velocities for dry, acidic soils (\( pH < 4 \)) reported by Garland [1977] were, on average, \( 1.3 \text{ cm s}^{-1} \) with 20% larger values when these same soils were moist. Using a similar bulk resistance analogy as used here, Fowler and Unsworth [1979] found SO₂ soil uptake resistances of the order of \( 1000 \text{ s m}^{-1} \) for a cultivated agricultural site. These resistances are consistent with our reported numbers for dry soils.

The differences in the surface uptake resistance between wet and dry surfaces results in a factor of \( \approx 2 \) in the deposition velocity. This general conclusion is commensurate with findings from laboratory studies assessing the effect of moisture on SO₂ uptake on a variety of soil types and characteristics [Terraglio and Manganelli, 1966; Payrissat and Beilke, 1975; Lockyer et al., 1978; Judeikis and Stewart, 1976; Judeikis and Wren, 1977].

O₃ Deposition to the Forest Floor

Ozone deposition velocities from all studies were plotted in a similar manner (versus \( \sigma_w \)) to SO₂ (Figure 6). The deposition velocities for O₃ show a slight dependency on turbulence (\( \sigma_w \)), similar to what was observed for SO₂ during dry conditions. In general, O₃ deposition appears to be limited by the surface uptake resistance for both moist and dry surface conditions. The surface resistance for O₃ was determined in a similar manner to that above for SO₂. The computed value from the Oak Ridge site (\( 22 \pm 8 \text{ s cm}^{-1} \)) was in agreement with that determined for the Huntington Forest location (\( 28 \pm 11 \text{ s cm}^{-1} \)). A plot of the deposition velocity against \( \sigma_w \) with \( R_s = 2000 \text{ s m}^{-1} \) fits the observed data well with the exception of one point, although this value was associated with one of the largest standard deviations. The large surface resistance calculated for O₃ is consistent with measurements by Wesely et al. [1981] over bare wet soil. O₃, because of its low solubility in water, is not expected to have high deposition rates to moist surfaces. The average deposition velocity was \( 0.05 \text{ cm s}^{-1} \) for this period. The trend displayed in Figure 6 is what would be expected for a diffusion process that is limited by a large surface uptake resistance. At low turbulence levels the aerodynamic and boundary layer resistance are relatively

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure6.png}
\caption{Fig. 6. The deposition velocity of O₃ (\( V_{d,O₃} \)) at Oak Ridge and Huntington Forest versus the standard deviation of the vertical velocity (\( \sigma_w \)) at 2 m above the forest floor; \( n \) is the number of half hourly periods (\( n \)) averaged together for each data point.}
\end{figure}
large components of the overall resistance to diffusion. However, at higher turbulence levels, deposition is limited by the presumably constant surface uptake resistances. This can be contrasted to the results obtained for SO\textsubscript{2} (Figure 5), in which the forest floor was moist and the uptake resistance was negligible. As the turbulence levels increase (higher $\sigma_\text{w}$), the deposition velocity increases in an almost linear fashion (similar to HNO\textsubscript{3}) because the aerodynamic and boundary layer resistances are believed to be the limiting factors in the diffusion process. However, this can only be true if the pH of these moist surfaces is well buffered. Measurements of soil litter leachate by Lindberg and Johnson [1989] found values of pH ranging from 5 to 6.

**Summary**

Eddy correlation measurements of $O_3$ and SO\textsubscript{2} deposition were made at 2 m above the surface within two deciduous forests. A spectral analyses of the scalar quantities measured in the lower canopy at Huntington Forest indicate a much steeper roll-off in the inertial subrange than in the atmospheric surface layer (a slope of $-1$ compared to $-2/3$). Similar spectra have been observed for wind velocity spectra at the Oak Ridge site. This finding is consistent with recent suggestions that turbulence in the lower canopy is in part generated by low-frequency pressure perturbations. A quadrant analysis of the raw time series data showed that at least 50% of air-surface exchange at the forest floor is dominated by turbulent events that provide, on average, 10 times the mean flux. These events, however, only occupy less than 10% of the time during the averaging period.

The deposition of SO\textsubscript{2} was found to vary with surface moisture. When the surface was moist, the surface uptake resistance ($R_s$) for SO\textsubscript{2} was negligible which resulted in a deposition rate that was generally a factor of 2 higher than to dry surfaces. For dry surface conditions, $R_s$ was estimated to be of the order of 700 s m\textsuperscript{-1}. The deposition of O\textsubscript{3} was invariant with surface moisture conditions and was associated with surface resistances that were of the order of 2000 s m\textsuperscript{-1}. For moist surface conditions, deposition of SO\textsubscript{2} to the forest floor comprises a significant fraction of the total flux to the ecosystem and during the dormant season has the potential to be a dominant sink for SO\textsubscript{2}. Total deposition estimates based on throughfall measurements [Lindberg and Lovett, 1992] need to account for this additional fraction since it is not measured by the throughfall method.

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