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Uncertainties Associated with the Inferential Modelling of Trace Gas Dry Deposition: A Comparison of Four Models with Observations from Four Surface Types

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Abstract

For operational monitoring, it is convenient to use inferential models to estimate dry and total deposition. Such models have only been tested for a limited number of surface and atmospheric conditions. To assess the uncertainties associated with the inferential approach 4 different model formulations have been compared with ozone and sulphur dioxide dry deposition velocities derived from flux measurements. These data were collected during 4 separate field studies, each of which involved a different surface type and atmospheric conditions. Averaged over the entire study, the means of the modelled O_3 deposition velocities were less than the observed mean by from 16% to 52%, depending upon surface type. There was considerable variation among models (±50%). For SO_2 deposition there was greater variability among models and the mean modelled deposition velocity was within -20% of the observed mean. Over shorter time periods there were greater discrepancies between the observations and the model predictions. For example, a majority of the modelled values of mean daily O_3 were within 25% to 55% of the observations depending upon model and surface type. However, for each 30 minute measurement period the differences were larger, often exceeding a factor of 2. A diurnal pattern, with larger O_3 deposition velocities during daylight hours, was observed over all surfaces, but was most pronounced over a deciduous forest in the summer. The models captured the mean diurnal patterns in the summer reasonably well, but nighttime O_3 deposition velocities were generally under-predicted There were larger differences between models and observations over the deciduous forest in early spring (no leaves) and over the cotton crop.

1. INTRODUCTION

The importance of monitoring the total deposition of acidifying chemical recognized. All mechanisms of pollutant deposition, wet, dry or occult, can have an effect on the natural and human environment. It is necessary to develop models for estimating dry and total deposition due to the relative complexity of the techniques for making direct measurements of dry deposition compared to wet deposition. The instrumentation is costly and requires considerable supervision. Hicks et al. (1980) recognized the feasibility of calculating dry deposition fluxes as the product of a modeled deposition velocity and a measured air concentration. Subsequently, much attention has been paid to the development of models for estimating dry deposition velocities. The inferential approach to modelling dry deposition can produce estimates that agree to some extent with available measurements. However, the models have only been tested for a limited number of surface and atmospheric conditions and their uncertainties are not well characterized.

The method being developed in Canada to determine dry deposition is based upon the inferential approach. Observations of meteorology, surface conditions and air concentrations are being collected to run site-specific inferential models. In addition, large-scale meteorological data are being used to produce inferential estimates of dry deposition velocity across a larger domain.

Four different model formulations have been compared with ozone and sulphur dioxide dry deposition velocities derived from flux measurements. A big leaf model (Big Leaf) (Hicks et al., 1987), a hybrid big leaf/multi-layer model (Multi-Layer) (Baldocchi et al., 1987; Meyers and Baldocchi, 1988), the modified ADOM dry deposition module (Padro et al., 1991) and the RADM module (Wesely, 1989; Byun, 1990) have been run for spring (no leaves) and summer conditions in a deciduous forest and summer conditions over a vineyard and a cotton crop. The results of these model runs are described in this paper. The difference in dry deposition velocity between models and observations provides an indication of the amount of uncertainty associated with inferential modelling of trace gas deposition. The input data and the resistance parameters were examined to identify some of the sources of these differences.

2. DATA AND METHODS

Although there are significant differences among the present models, they are all based upon the multiple resistance analogue. Three main resistance terms are considered in the calculation of deposition velocity (V_d). These are the aerodynamic resistance (R_a), the laminar sub-layer resistance (R_b) and the canopy or surface resistance (R_c). The largest differences between models are associated with R_c and to a lesser degree R_a. Table I lists the input information required to run the models. The Big Leaf and Multi-Layer models rely on wind speed (u), the standard deviation of wind direction (σ_θ) and solar radiation to determine R_a (Hicks et al., 1987). RADM and ADOM calculations of Ra are more complex, utilizing the bulk Richardson number and wind speed to determine u* and L. However, ADOM uses the Louis parameterization (Louis, 1979), while RADM uses Byun's method (Byun, 1990) to compute these parameters. Differences in the R_c calculations are too great to discuss here. One of the main differences is that RADM and ADOM use land-use categories while Big Leaf and Multi-Layer consider specific information on the plant type at the location of interest. This refinement requires more input information, such as plant-specific light response constants (r_{bs}). For a complete description of the models please refer to the literature references given above.

Table 1 The main input information required to run the inferential models.

	Big Leaf	Multi-Layer	ADOM	RADM
Atmospheric.	u, T, σ_θ, hv, wetness, pre.	u, T, σ_θ, hv, wetness, pre	u, ΔT, pre., wetness	u, ΔT, hv, RH, pre.
Surface	species(%), LAI, % leaf, z_0, R_{stom}(min), r_{bs}	species (%), profile type, LAI, % leaf, z_0, R_{stom}(min), r_{bs}	season, land-use (8), z_0, LAI, C_{stom}, lat., long.	season, land-use (11), z_0, R_{stom}(min)

Information on the observed V_d data that were used in the model comparison is given in Table II. Mean 30 minute (min) O_3 fluxes were determined using eddy correlation over 4 different surfaces. The number of 30 min periods with valid observation ranged from 565 over cotton during CODE (California Ozone Deposition Experiment) to 1196 over a summertime maple forest (EMEFS-I, Eulerian Model Evaluation Field Study). There was a limited amount of SO_2 flux data which were only collected during EMEFS-II also using eddy correlation. These data are not as certain as the O_3 data and negative SO_2 V_ds were excluded from the analysis. However, the magnitude of the V_ds and the general diurnal pattern are believed to be true.

Table 2 Description of the Field Studies providing V_d Measurements

Study	Surface	Time Period	No.	Poll.
EMEFS I	maple-full leaf	July 15-Aug. 30, 1988	1196	O_3
EMEFS II	maple-no leaves	Mar. 17-Apr. 27, 1990	996	O_3
			247	SO_2
CODE	vineyard	July 11-Aug. 6, 1991	1183	O_3
CODE	cotton crop	July 15-Aug. 6, 1991	565	O_3

Model comparisons were only based upon time periods when all models were able to compute V_d values. This was not possible for the entire time periods indicated in Table II because model input data were not available for every 30 min interval. However, for a majority of the time the model values were determined, except for RADM. During the CODE studies no input data were available and for all studies RADM was only capable of determining hourly V_d values.

3. RESULTS

Observed and modelled deposition velocities were compared for a variety of time scales ranging from 30 min to multiple weeks. In Figures 1a and 1b the individual 30 min observations are compared to the Multi-Layer and ADOM estimates. Overall, for both models there were more underpredictions of V_d and there tended to be an upper limit to their V_d predictions. There were also a substantial number of observations where the discrepancy between modeled and observed V_d was greater than a factor of 2. For the Multi-Layer and ADOM models, 36% and 35% of the predictions were off by more than a factor of 2. This percentage increased to 44 and 47 for the Big Leaf and RADM models, respectively. As shown on Figures 1a and 1b the model biases differed between field studies. For example, the Multi-Layer model tended to underestimate the O_3 V_d values over the vineyard and cotton crop and overestimate V_d over the leafless maple forest. In contrast, ADOM underestimated V_d over the leafless forest.

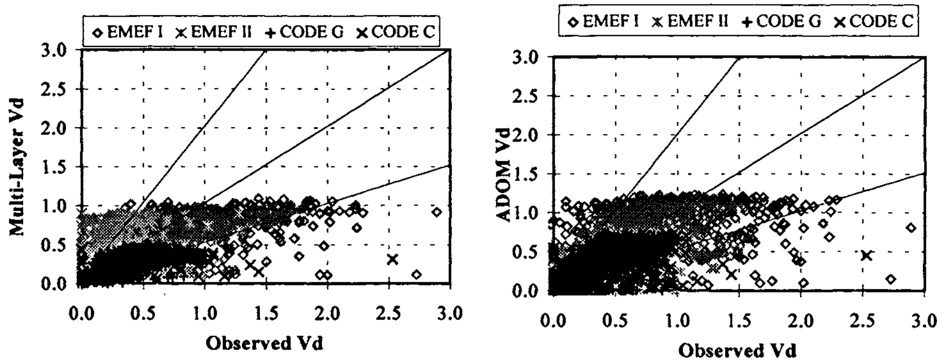

Figure 1a, b Comparison of observed and modelled V_d (30 min) for (a) Multi-Layer and (b) ADOM (cm s^{-1}).

In the future, Vd values from site-specific inferential models are expected to be used with Canadian Air Precipitation Monitoring Network (CAPMoN) measurements of SO_2, SO_4^{2-} and HNO_3 to compute dry deposition fluxes. These air concentration data are collected daily for integrated 24 hour periods. The observed and modelled V_d's were, therefore, averaged for each individual day during the field studies to examine how the models would perform for 24 hour periods. Mean percent RMS errors between modelled and observed 24 hour V_d values, which are listed in Table 3, varied between models and field studies. Errors were smallest over the vineyard (30%) and largest for SO_2, for which the errors ranged from 39% for the Multi-Layer model to 130% from ADOM. The day to day variations in mean O_3 V_d for EMEF I are shown in Figure 2a. A similar plot corresponding to SO_2 is included in Figure 2b. Only days with more than 17 of the possible 48, 30 min periods were included in these figures and in Table 3. The models tended to underestimate mean daily O_3 V_d (15 of 27 days for EMEF I). There were only 16 days with sufficient SO_2 results. The model predictions were above and below the observations with ADOM and the Big Leaf model deviating most from the observed values.

Table 3 Mean Percent RMS Errors in Daily Mean Deposition Velocity

	Multi-Layer	Big Leaf	ADOM	RADM
EMEFS I	25%	32%	28%	41%
EMEFS II	53%	55%	50%	37%
EMEFS II-SO2	39%	85%	130%	81%
Vineyard	29%	22%	23%	-
Cotton	50%	40%	37%	-

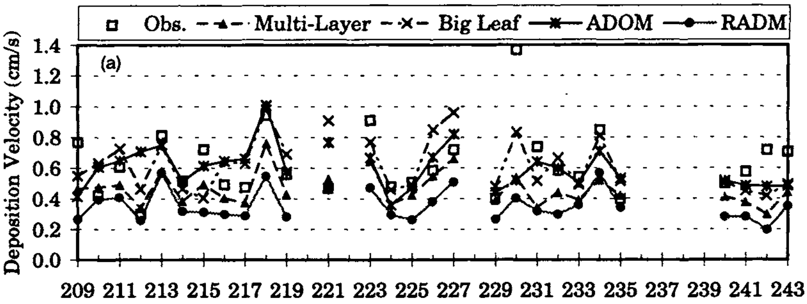

Figure 2a Mean daily V_d for O_3 during EMEFS I (cm s^{-1}).

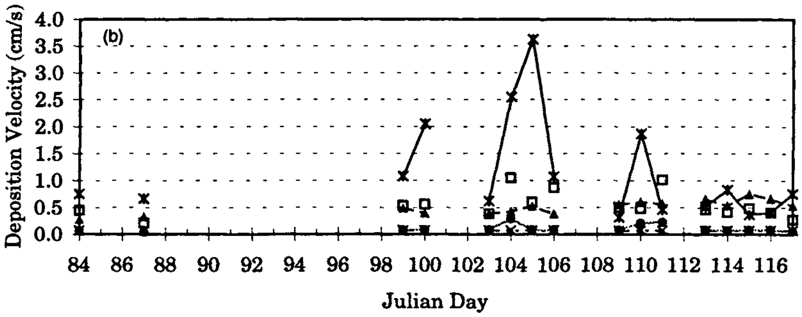

Figure 2b Mean daily V_d for SO_2 during EMEFS I (cm s^{-1}).

Mean diurnal variations in V_d for each case listed in Table I are shown in Figures 3a-e. According to the observations there tended to be a daytime peak in V_d for all surface types and for O_3 and SO_2. When there were leaves on the vegetation the peaks tended to occur at midday, when incoming solar radiation was strongest. This behaviour supports the belief that stomatal control of pollutant uptake, which effects R_c, is very important for O_3 and SO_2. The models produced similar diurnal patterns, with the exception of the Big Leaf model for EMEFS II for both O_3 and SO_2 and RADM for SO_2. The Big Leaf model exhibited no diurnal variation for the leafless forest conditions. This was because the leaf area index (LAI) was very small and thus, R_c was set to large constant value corresponding to the soil resistance. There was also very little diurnal variation in the SO_2 R_c for RADM over the leafless forest (Wesely, 1989). Given that there were no leaves on the trees, it is somewhat surprising that a diurnal pattern in O_3 V_d was observed and that 3 of the models also predicted such a pattern. RADM increased V_d during the day because Wesely (1989) assumed a R_c dependence on solar radiation even though there are no leaves. In contrast, the ADOM diurnal pattern was produced because of an assumed temperature effect on R_c. Thus, these models both resulted in reasonably good predictions of the mean diurnal pattern, but for different reasons.

The amount of agreement between the mean hourly observations and the model results varied with surface type, model and time of day. Over cotton (Fig. 3d) all three models (RADM was not used) underestimated V_d. The largest errors were during the day, when the observations tended to be twice as large as the model results. ADOM and Big Leaf both predicted a sharp decline in V_d in the early afternoon. Big Leaf exhibited this behaviour because of its simplified approach to partition the incoming solar radiation into shaded and sunlit leaves. Apparently, for overhead sun conditions this approach leads to an unrealistic reduction in photosynthetically active radiation to the leaves, particularly at the more southern latitudes. This pattern was also predicted for the summertime maple forest, but due to the more northern latitude of the EMEFS II site the drop was not as pronounced as during CODE. Possible reasons for the ADOM decline in V_d over cotton were discussed by Padro et al. (1994).

During EMEFS I, there was a general tendency for the models to underestimate V_d at night. In particular, in the early morning hours the observations were 2 to 3 times larger than the model values. The low bias at nighttime was also evident in the Multi-Layer and Big Leaf results for both CODE studies. The nighttime biases were not as consistent for ADOM (i.e., ADOM overestimated for the vineyard).

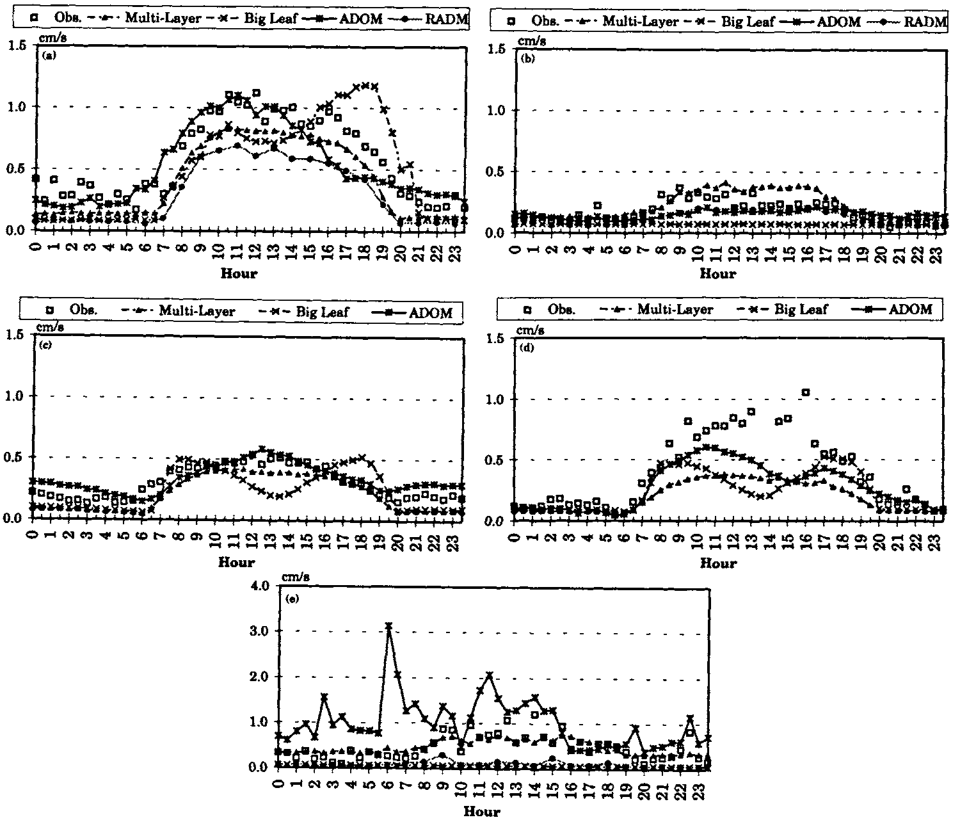

Figure 3 Mean diurnal patterns in V_d (cm s^{-1}). (a) EMEFS I. (b) EMEFS II. (c) Vineyard. (d) Cotton (e) EMEFS II SO_2

Model performance for SO_2 was generally not as good as for O_3. According to the observations there was an increase in V_d during the day (Fig. 3e), but no obvious midday peak. The Big Leaf model and RADM did not match this pattern. Reasons for this behaviour were discussed above. The Multi-Layer model performed surprising well with no apparent bias during periods without sunlight and only a slight low bias during the day. ADOM predicted some structure in the hourly variations of V_d and there was a tendency for a peak in the early morning hours. This may have been a result of surface wetness.

There are a number of other interesting features in Figures 3a-4e, which are currently being examined in more detail. For example, the Big Leaf model predicts a large peak in O_3 V_d in the late afternoon and early evening during EMEFS I. This behaviour has not been fully explained, but some of it was due to the variation in R_c. Figure 5 is a plot of the mean diurnal pattern in R_c+R_b. The Big Leaf model resistances were smallest at approximately 1830 LST and remained below the ADOM, Multi-Layer and RADM values until 2000 LST or later.

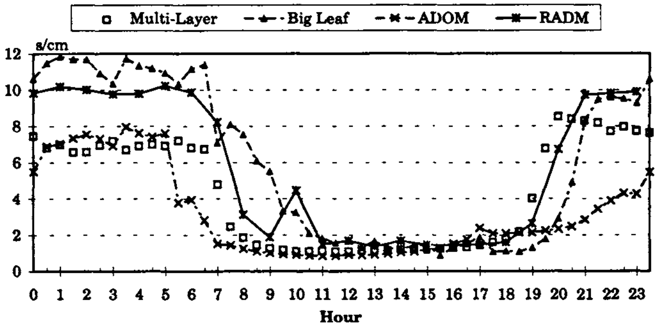

Figure 4 Diurnal pattern in modelled Rc+Rb (s cm^{-1}) for EMEFS I.

Mean V_ds were determined for the entire durations of the field studies. These values are listed in Table 4. For O_3, the number of 30 min values that were included in the average ranged from about 500 to 1000, depending upon the study. Fewer observations were available for SO_2. With the exception of the Multi-Layer model during EMEFS II and ADOM over grape, the overall tendency was for the models to underestimate V_d. For O_3, the average amount of bias (difference between the observed V_d and the mean of the 3 or 4 models) was -17% for the leafless forest, -22% for the forest with leaves, -52% for cotton and -16% for grape. For SO_2 the average bias was -18%. As seen in previous figures the models differed from one another substantially. Compared to the mean V_d across all 3 or 4 models, the range of mean V_ds for the individual models were: +19% to -29% for EMEFS I; +59% to -52% for EMEFS II; +14% to -16% for cotton and; +28% to -18% for grape. The variability among models was larger for SO_2 ranging from +140% to -84%.

Table 4 Comparison of Overall Mean Deposition Velocities (cm s^{-1})

Site	Obs.	Multi-Layer	Big Leaf	ADOM	RADM
EMEFS I	0.67	0.50	0.62	0.60	0.37
EMEFS II	0.18	0.23	0.07	0.15	0.13
Cotton	0.50	0.20	0.24	0.27	-
Grape	0.31	0.22	0.24	0.34	-
Stratified by Stability (EMEFS I)					
stable	0.44	0.28	0.42	0.44	0.18
unstable	0.91	0.73	0.84	0.79	0.59
B90-SO_2	0.52	0.52	0.07	1.02	0.10

CONCLUSIONS

There are a variety of approaches to modelling and/or parameterizing the deposition velocity of acidifying pollutants to surfaces. In this paper, 4 approaches were compared for 4 different sets of conditions. The amount of discrepancy between models and observations varied among models and from one surface type to the next. In addition, the agreement between models and observations decreased with increasing temporal resolution and reasons for agreement or lack of agreement varied among models. These results clearly suggest that the models are not adequately representing the underlying processes involved in pollutant dry deposition. Results of comparisons, such as those presented here, can be used to improve the models. However, before models can be fully tested, there is a need for more flux measurements for a larger variety of surface and atmospheric conditions and chemical species. More research is needed.

Given the range of predictions that arise from different models it is apparent that for monitoring total deposition of air pollutants it will be important for all groups/countries involved to coordinate their modelling efforts.

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