Evaluation of Data Replacement Strategies for CASTNET Dry Deposition Modeling

Christopher Rogers[1], Thomas Lavery[2], Kevin Mishoe[3] and Ralph Baumgardner[4]

1 AMEC Environment & Infrastructure, Inc., Jacksonville, 32207, FL, USA
2 Fuss & O'Neill, Inc., Providence, RI, 02908, USA
3 AMEC Environment & Infrastructure, Inc., Newberry, FL, 32669, USA
4 U.S. Environmental Protection Agency, Research Triangle Park, NC, 27711, USA

Abstract: The U.S. Environmental Protection Agency (EPA) established the Clean Air Status and Trends Network (CASTNET) and its predecessor, the National Dry Deposition Network (NDDN), as national air quality and meteorological monitoring networks. Both CASTNET and NDDN were designed to measure concentrations of sulfur and nitrogen gases and particles and to estimate dry deposition using an inferential model. The design was based on the concept that atmospheric dry deposition flux could be estimated as the product of a measured air pollutant concentration and a modeled deposition velocity (V_d). The multi-layer model (MLM), the computer model used to simulate dry deposition, requires information on meteorological conditions and vegetative cover as model input. The MLM calculates hourly V_d for each pollutant, but any missing meteorological data for an hour renders V_d missing for that hour. Because of percent completeness requirements for aggregating data for long-term estimates, annual deposition rates for some sites are not always available primarily because of missing or invalid meteorological input data. In this work, three methods for replacing missing on-site measurements are investigated. These include (1) using historical values of deposition velocity or (2) historical meteorological measurements from the site being modeled or (3) current meteorological data from nearby sites to substitute for missing inputs and thereby improve data completeness for the network's dry deposition estimates. Results for a CASTNET site used to test the methods show promise for using historical measurements of weekly average meteorological parameters.

Key words: Dry Deposition, Deposition Velocity, Leaf Area Index, Multi-Layer Model (MLM).

1. Introduction

The U.S. Environmental Protection Agency (EPA) established the Clean Air Status and Trends Network (CASTNET) to provide data for determining relationships between changes in emissions and any subsequent changes in air quality, atmospheric deposition, and ecological effects. The monitoring network emphasized the selection of rural sites. CASTNET has its origins with the National Dry Deposition Network (NDDN), which was established in 1986 and began operation in 1987. Many of the original NDDN sites are still operational after more than 20 years and provide useful information on trends in air quality.

CASTNET was designed primarily to provide long-term measurements for seasonal and annual average concentrations and depositions. Consequently, measurements of weekly average concentrations were selected as the basic sampling strategy. An open-face, three-stage filter pack was employed to measure gaseous and particulate sulfur and nitrogen pollutants as well as concentrations of other pollutant species. The filter pack technology and sampling protocol have been used consistently over the network's history, providing a comparable data set each year and allowing for the analysis of long-term trends.

Currently, CASTNET consists of 82 monitors at 80 sites. The network is sponsored by EPA and the National Park Service (NPS), which began its participation in CASTNET in 1994. Sites are located throughout the continental United States, Alaska, and Canada. Collocated monitoring sites are operated at

Corresponding author: R.E. Baumgardner Jr., Environmental Scientist, main research fields: ecological effects, ambient air monitoring. Email: Baumgardner.Ralph@epamail.epa.gov.

Mackville, KY (MCK131/231) and Rocky Mountain National Park, CO (ROM406/206) in order to estimate network precision. More information on CASTNET can be found at http://www.epa.gov/castnet/.

Dry deposition processes are modeled as resistances to deposition [1-4]. The original network design was based on the assumption that dry deposition or flux could be estimated as the linear product of measured pollutant concentration (C; μ/m^3) and modeled deposition velocity (V_d; cm/s), which simulates the processes that control the rate of deposition of a pollutant. Measured weekly atmospheric concentrations are calculated based on the mass of each analyte in each filter extract and the volume of air sampled. Ozone (O_3) concentrations, which are measured continuously, are recorded as hourly averages. V_d is influenced by meteorological conditions, vegetation, and atmospheric and plant chemistry. The deposition velocity values are calculated for each hour of each year using the multi-layer model (MLM) [1-2].

Inferential models, such as the MLM, have long been used to estimate dry deposition of pollutants to ecosystems based on measurements of air concentrations. Some models (e.g., MLM) use measured meteorological conditions directly as model input. The Big-leaf Model (BLM), used by the Canadian Air and Precipitation Monitoring Network (CAPMoN), and other inferential models, which are used as modules within regional chemical transport models, use meteorological input data produced by numerical weather prediction models. Recent studies [5-6] have shown that flux estimates produced by inferential models can be inconsistent because of differences in model-calculated V_d values. The purpose of this study is not to discuss the uncertainties among the various inferential models but to investigate three potential methods to replace missing V_d values. The three potential methods include (1) using historical values of deposition velocity or (2) historical meteorological measurements from the site being modeled or (3) current meteorological data from nearby sites to substitute for missing data.

2. Experimental Methods

2.1 CASTNET Continuous Measurements and Filter Concentration Data

Data regularly collected from CASTNET sites and used as input for this study include continuous measurements of meteorological parameters and ozone and pollutant concentration data derived from exposed filters. Concentration data consist of laboratory results acquired through analyses of the four filters contained in the three-stage filter packs installed weekly at each site. These data products are combined by the MLM to create estimates of dry deposition, which is one of the principal objectives of the network. All data are processed and validated, and instruments are calibrated according to protocols described in the CASTNET Quality Assurance Project Plan (QAPP) [7].

Continuous data are polled from each site via cellular or telephone modem. The on-site data acquisition system (DAS) calculates hourly averages for the measured meteorological parameters and ozone. Of interest to this study, the MLM specifically uses wind speed, standard deviation of the wind direction (sigma theta), solar radiation, temperature, relative humidity, precipitation, and surface wetness to estimate hourly values of V_d. When one of these meteorological measurements is missing, V_d is not calculated for that hour.

The filter pack used at CASTNET sites consists of three types of filters arranged in the following order: Teflon, nylon, and cellulose [7]. The filter pack is open faced and has an approximate size cut of 10 micrometers. The Teflon filter collects particles, specifically sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium

(NH_4^+), chloride (Cl^-), and other earth metals. The nylon filter collects gaseous HNO_3 and some SO_2. Finally, two cellulose filters, which consist of a cellulose fiber base impregnated with potassium carbonate (K_2CO_3), collect the remaining SO_2. Among other measurement artifacts, there are known issues with the measurement of NO_3^- and HNO_3 by the filter pack because of the particle size distribution of NO_3^- particles sampled and potential volatilization of NO_3^- particles during the sampling period [8-10]. Final atmospheric concentrations are calculated by multiplying the analyte concentrations with the aggregated flow volume (in cubic meters) for the time period the filter pack was installed.

2.2 Plant Speciation Information

The MLM requires data regarding the characteristics of the plant types that immediately surround the site. The CASTNET database includes data for plant species and their weekly percent coverage, canopy height, leaf area index (LAI), and percent leaf-out for all of the vegetation within a 1-kilometer (km) radius around each site. For CASTNET applications, these data are assumed constant.

2.3 Dry Deposition Estimates from the MLM

The MLM simulates hourly deposition velocities for SO_2, HNO_3, O_3, and particles. The estimates are dependent on the specific meteorological measurements and plant information. The hourly deposition velocities are combined with the filter pack and O_3 concentrations to estimate hourly rates of dry deposition in kg/ha. The weekly filter concentrations are assumed constant for each hour of the 7-day period. The post-processor aggregates the hourly deposition velocities and deposition rates into weekly, monthly, quarterly (from weekly), seasonal (from weekly), and annual values (from quarterly or seasonal). Each aggregation step requires a percent completeness of 69 percent for the relevant underlying values. Standard CASTNET protocol is to calculate annual aggregates from the four quarterly values within a calendar year. At least three valid quarterly aggregations are required to calculate an annual value.

Current MLM modeling procedures include the use of meteorological data from “near sites” when data are missing from the site that is being modeled. One or two “near sites” have been assigned to most sites. The current “near sites” were assigned based on geographic proximity and similar terrain and land use [11]. Maps of CASTNET sites are shown in the MLM sensitivity study [11].

As a result of the percent completeness requirements, annual deposition estimates for some sites are not always available, often because of missing meteorological input data. Table 1 shows the percentage of all available site-year estimates for 1987–2007 that are valid with a breakdown of how many site-years were calculated from four valid quarters as opposed to three valid quarters. Approximately 25 percent of site-years are not available because of incomplete data. To improve data completeness, three replacement strategies were identified and tested: use of “near site” meteorological data based on comparing historical deposition velocities with calculated values, use of historical mean deposition velocities, and replacement of missing hourly wind parameter data using historical mean values.

Table 1 Summary of Valid Site-Years

	Count of Site-Years	Percent Completeness
Total	1404	
4 quarters valid	711	51 percent
3 quarters valid	291	21 percent
All valid site-years (3 or 4 quarters valid)	1002	71 percent

2.4 Ten-year Mean Deposition Velocities

For this study, ten-year mean deposition velocities for 1998–2007 were calculated for each site. Each site was compared against every other site regardless of geographic proximity. Only site-years with all four quarters used in the annual mean were used to ensure that all seasons were represented. All site combinations where the percent difference between each of the estimated deposition velocities was within ±10 percent were listed. This list was used to evaluate the current list of "near sites" used for replacing missing meteorological data when completing a MLM model run. Each site was also compared against itself by comparing the 10-year mean deposition velocity with each of the 10 individual annual mean deposition velocities from 1998–2007.

2.5 Analysis of Archived MLM Simulations

Annual aggregations for each of the 10 years from the period 1998–2007 were compared with flux estimates calculated in two ways. The first involved multiplying 10-year mean V_d for each site for each pollutant by the annual mean concentration for each pollutant. The second method involved multiplying annual mean deposition velocities for each year by the annual mean concentrations. Differences were then determined for site-years with either three or four valid quarters (all valid site-years) compared with using only site-years with four valid quarters (complete year is represented). These analyses were used to evaluate simply using 10-year mean V_d values and annual mean pollutant concentrations to estimate annual mean dry deposition fluxes.

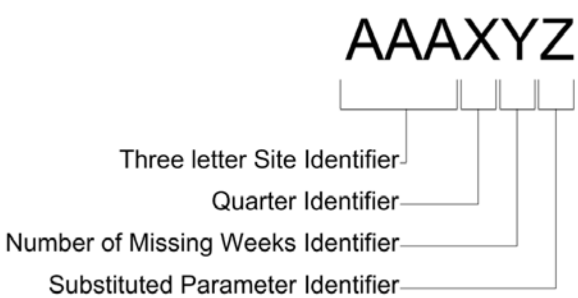
2.6 Replacement of Missing Wind Speed and Sigma Theta Data

Missing vector wind speed and sigma theta data for the CASTNET site at Lykens, Ohio (LYK123) were replaced with either historical weekly mean values from LYK123 or with data from the Deer Creek, Ohio (DCP114) site collected over the 10-year period 1998–2007 [11]. These two meteorological parameters have been shown by previous sensitivity analyses to be the most influential in the estimation of deposition velocities [12-13]. The periods of missing data were artificially created using the scheme described below. Deposition velocity and flux estimates were calculated for 2007 by running the MLM using the replaced meteorological data. In total, 48 artificial site data sets were run through the model. Each site data set represented a different combination of missing and replacement data. These estimates were compared with the archived annual flux estimates to determine if near site data or historical average values are useful sources for substitution of missing data.

To create the data set with artificial missing data, values were removed from a specific number of randomly selected weeks for several parameters. Different substitution values could then be easily adapted to the same missing data scheme. Data were removed from six different combinations of missing data in two different quarters. First and third quarters were selected because they represent the weather extremes of winter and summer seasons. A site identification code was created to represent each combination of missing data. The result was a Cartesian product of each grouping: 12 identification codes per parameter, per site. Week numbers were randomly selected for each group of missing weeks and the same weeks were used for each site in the group. For example, weeks 1 and 2 were randomly selected for the Two Missing Weeks group, and every site and parameter combination used these weeks for the Two Missing Weeks set. The value for the identified parameter was set to null for each data point within the selected weeks.

Figure 1 illustrates the construction of the site data set identification used to document the periods of missing data, describes the meaning of the X, Y, and Z positions of the site identification, and documents the weeks that were randomly removed for each scenario. Site data sets where missing data were replaced with data from DCP114 begin with a LYK site identifier. Site data sets where missing data were replaced with historical weekly averages begin with a HIS site identifier. For example, the site LYKOFS indicates that four weeks of wind speed data were removed from quarter one at site LYK123 and replaced with data from DCP114. The site HISTAG indicates that all weeks of sigma theta data were removed from quarter three at site LYK123 and replaced with historical weekly averages.

Figure 1. Site Identification Codes



Description of X, Y, and Z coding

X	Y	Z
Selected Quarter Identifier	Number of Missing Weeks Identifier	Missing Parameter Identifier
O ≥ Quarter One	T ≥ Two Weeks	S ≥ Wind speed
T ≥ Quarter Three	F ≥ Four Weeks	G ≥ Sigma Theta
	S ≥ Six Weeks	
	E ≥ Eight Weeks	
	N ≥ Ten Weeks	
	A ≥ All Weeks	

Randomly Selected Weeks Removed

Number of Weeks	Week Numbers
Two	1, 2
Four	2, 3, 8, 9
Six	5, 6, 9, 10, 11, 12
Eight	1, 2, 4, 5, 7, 8, 9, 10
Ten	1, 2, 3, 4, 5, 6, 7, 8, 9, 12
All	All

The historical weekly averages were obtained by calculating the average of all valid sigma theta or vector wind speed for a particular week for the 10-year period of 1998–2007. This created a data set of 54 weekly averages for these two parameters. Missing hourly data were replaced by determining the week of the missing hour and then substituting the average value for that specific week. For the study model runs, no time was provided for spin up of the soil moisture budget, which requires typically a few simulation weeks. All runs were initialized with soil moisture values of 20 mm. Because all runs started with the same value, the spin up is not important to the conclusions of the study. Also, scalar wind speed was not used in the model runs. For routine model runs, scalar wind speed can be used as a backup when vector wind speed is either invalid or missing.

3. Results and Discussion

3.1 Analysis of Near Sites by Comparing Ten-year Mean Deposition Velocities

To identify pairs of sites where the 10-year mean deposition velocities were similar, the means for 1998–2007 for each site were compared with the 10-year mean values for all other sites. Relative percent differences (RPD) were calculated for SO_2, HNO_3, and particulate V_d. Current pairs of sites that are assigned as near sites with values within ±10 percent are shown in Table 2. The current collocated sites (MCK131/231, KY and ROM406/206, CO) are also included. The N/S (north/south) and E/W (east/west) distance columns show the distance in kilometers that separate the sites.

Only five current "near site" pairs [11] were identified through this analysis as a good match for replacing missing meteorological data. Most of the matches shown in this table are between sites that are not in close proximity. Therefore, they cannot be used as near sites because the good comparison is fortuitous and unrelated to prevailing synoptic scale meteorological conditions.

The comparison between the collocated sites is also informative. These pairs are located at the same monitoring location and should have the best comparisons. The RPD for SO_2 V_d is approximately 5 percent for MCK131/231 and -5 percent for ROM406/206. The RPD for HNO_3 V_d and particulate V_d are closer to zero.

3.2 Analysis of Historical V_d Data by Comparing Archived MLM Simulations

Fluxes based on (1) 10-year mean deposition velocities and (2) annual mean deposition velocities were compared with archived historical annual flux values by constructing histograms of percent differences. Comparisons were made using annual mean and 10-year mean values when either three or four quarters were valid (one quarter of the year may not be represented in the annual mean) and a second set of comparisons were made using 10-year mean and annual mean values when all four quarters were valid (the entire year is covered). Figures 2 and 3 present histograms of percent differences versus number of site-years based on annual mean values when all four quarters were available. Figure 2 is based on annual mean deposition velocities, and Figure 3 is based on fluxes estimated from 10-year mean deposition velocities. Each histogram includes a bar for each of five pollutants: SO_2, HNO_3, SO_4^{2-}, NO_3^-, and NH_4^+.

Table 3 summarizes the results from the use of 10-year and annual mean fluxes. The two columns labeled percent of site-years within ±20 percent of the historical flux estimate show that for the analysis done when all quarters were valid, well over 90 percent of all site-years have a percent difference within the ±20 percent window, except for NO_3^-, which is lower.

The results for SO_2, HNO_3, NO_3^-, and NH_4^+ show that the calculated fluxes are somewhat higher than the archived MLM results. The exception is for SO_4^{2-} which shows lower modeled values. Use of the 10-year mean allowed for the recovery of 131 additional site-years (37 percent). Many of the missing site-years are the start-up or shut-down years for a specific site. Use of the annual mean allowed for the recovery of only 8 additional site-years (2 percent) but was useful in examining how flux estimates calculated with annual average concentrations and deposition velocities are comparable with those aggregated using the current protocol.

Table 2 Current Near Sites or Collocated Sites with a Percent Difference of ± 10 Percent for each Deposition Velocity

Site ID (1)	Site ID (2)	N/S Distance (km)	E/W Distance (km)	Sulfur dioxide V_d RPD	Nitric acid V_d RPD	Particulate V_d RPD
ALH157*	BVL130*	-131	-100	2.1	-2.6	-5.3
ARE128	WSP144*	-43	-195	-4.3	4.4	5.2
*DCP114**	*LYK123**	*-142*	*-21*	*4.4*	*5.9*	*-1.0*
HOX148*	PRK134	-114	389	-7.7	-3.9	-2.9
LYK123*	OXF122*	154	138	6.5	8.4	1.4
MCK131	MCK231	0	0	5.5	2.5	1.3
ROM206	ROM406	0	0	-5.4	-0.8	-0.8

Note: Italicized text indicates "near site" pair selected

Table 3 Summary of 10-Year Mean and Annual Mean Calculated Flux Comparisons (Percent Differences)

Parameter	± 4 Percent Difference		± 20 Percent Difference	
	3 or 4 Quarters	All	3 or 4 Quarters	All
Sulfur dioxide 10-yr Flux	31	36	92	95
Sulfur dioxide Annual Flux	53	60	96	99
Nitric acid 10-yr Flux	43	47	94	97
Nitric acid Annual Flux	60	68	97	99
Sulfur trioxide 10-yr Flux	31	37	91	96
Sulfur trioxide Annual Flux	42	49	96	98
Nitric oxide 10-yr Flux	24	25	79	83
Particulate nitrate Annual Flux	24	26	83	87
Particulate ammonium 10-yr Flux	34	38	93	96
Particulate ammonium Annual Flux	55	61	96	98

Table 3 and Figure 2 show that the simulations based on individual annual V_d values better match the MLM results. For SO_2, approximately 99 percent of the calculated fluxes are within 20 percent of the MLM deposition rates, and about 60 percent of the calculated values are within 4 percent of the MLM results. Table 3 and Figure 3 show that SO_2 results based on the 10-year mean deposition velocities indicate that approximately 95 percent of the calculated fluxes are within 20 percent of the MLM values and about 36 percent are within 4 percent.

For HNO_3, SO_4^{2-}, NO_3^-, and NH_4^+, Figures 2 and 3 again show the flux estimates based on individual annual V_d values better match the MLM results. The fluxes based on 10-year mean V_d values for HNO_3, SO_4^{2-}, and NH_4^+ are somewhat better than the SO_2 results. More than 95 percent of the flux values based on 10-year mean V_d values are within 20 percent of the MLM annual values. The results for NO_3^- are not as good. Particulate nitrate concentrations are lower and more uncertain than the other parameters.

Figure 2. Histogram of Percent Difference versus Number of Site-Years for Annual Mean Deposition Velocity Comparisons

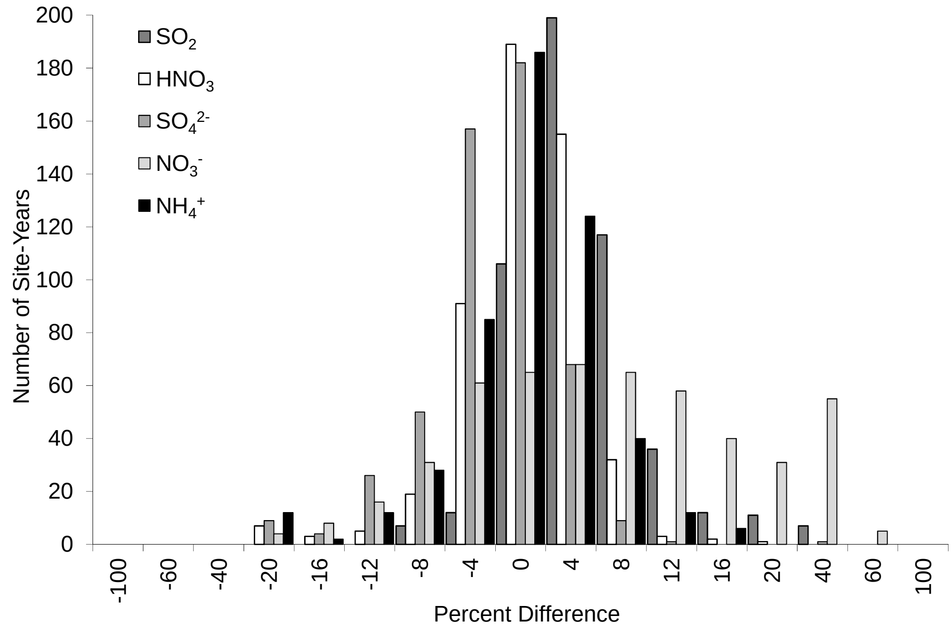
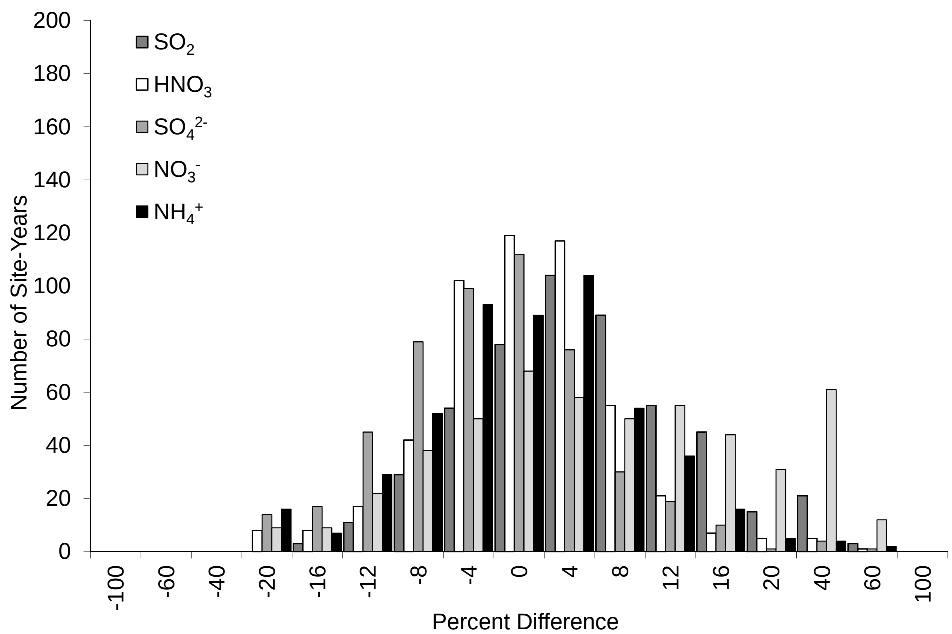


Figure 3. Histogram of Percent Difference versus Number of Site-Years for 10-Year Mean Deposition Velocity Comparisons



The analysis of the historical deposition velocities demonstrates that the use of 10-year mean V_d values and annual mean pollutant concentrations provides acceptable estimates of annual mean fluxes if a 20 percent uncertainty is acceptable.

3.3 Analysis of Replacement of Wind Speed and Sigma Theta Data with Historical Values

Model runs were performed by replacing missing vector wind speed and sigma theta data with either historical weekly mean values or "near site" values. The "near site" pair of LYK123 and DCP114 was selected for the analysis with LYK123 as the primary site. Model runs were completed for 2007 and then compared with the archived deposition velocity estimates. LYK was selected because archived V_d values had a near perfect level of completeness for 2007 with all 52 weeks having a percent completeness greater than 90 percent.

The results of the analyses of replacing missing sigma theta and wind speed data at LYK123 with historical weekly mean values from LYK123 and with data from DCP114 are illustrated in Figures 4 though 7. Figure 4 is a bar chart that illustrates the analyses of the HISOiS results where i = T through A (Figure 1). In other words, Figure 4 shows the results of replacing missing wind speed data for six combinations of missing weeks during the first quarter with historical weekly wind speed data from LYK123. The x-axis gives the number of missing weeks and the y-axis presents mean absolute percent differences (MAPD) between archived V_d values for three pollutants and V_d values based on running MLM with the substituted historical wind speed values. All the results are less than 5 percent. Generally, the MAPD values increased with the number of weeks with missing data. Also, the results for V_d for HNO_3 were somewhat worse than for SO_2 and particles. Although not shown, the results for sigma theta were similar. Figure 5 presents the results from LYKOiS based on the substitution of weekly values of wind speed from DCP114. All but three MAPD values are less than 5 percent. The results for sigma theta were similar albeit with five MAPD values greater than 5 percent.

Figure 4: Mean Absolute Percent Difference between Weekly Estimates from Historical Data

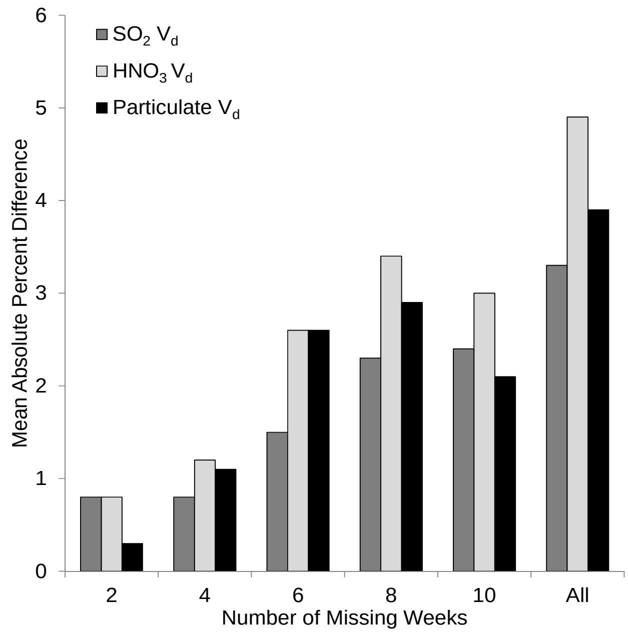
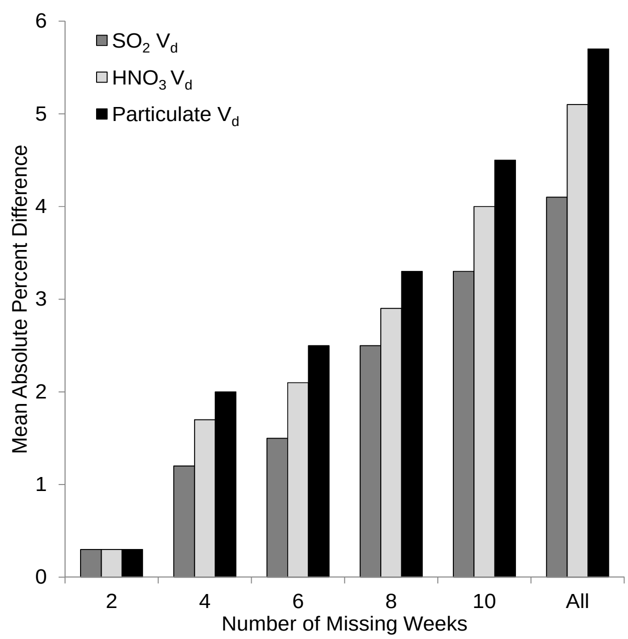


Figure 5: Mean Absolute Percent Difference between Weekly Estimates from Near Site Data



Figures 6 and 7 present results for HISOiS and LYKOiS, respectively, based on calculating MAPD between archived annual mean V_d values for three pollutants and annual mean V_d values based on running MLM with the substituted historical wind speed values.

Figure 6: Absolute Percent Difference between Annual Estimates from Historical Data

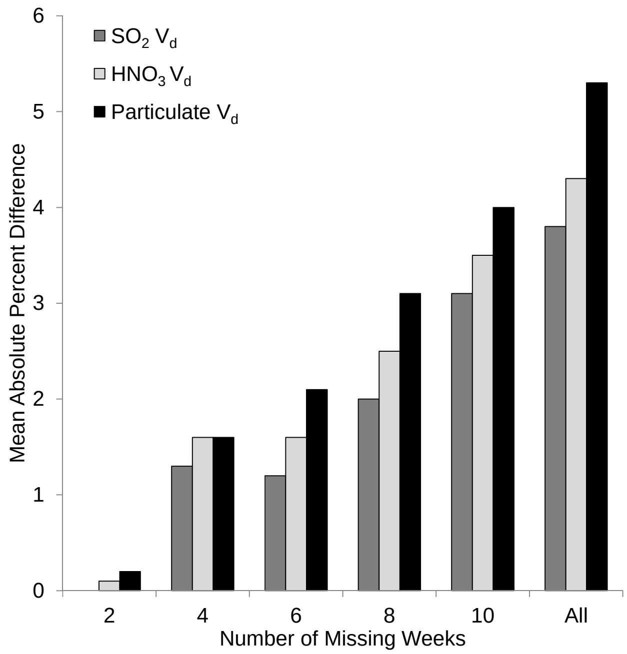


Figure 7: Absolute Percent Difference between Annual Estimates from Near Site Data

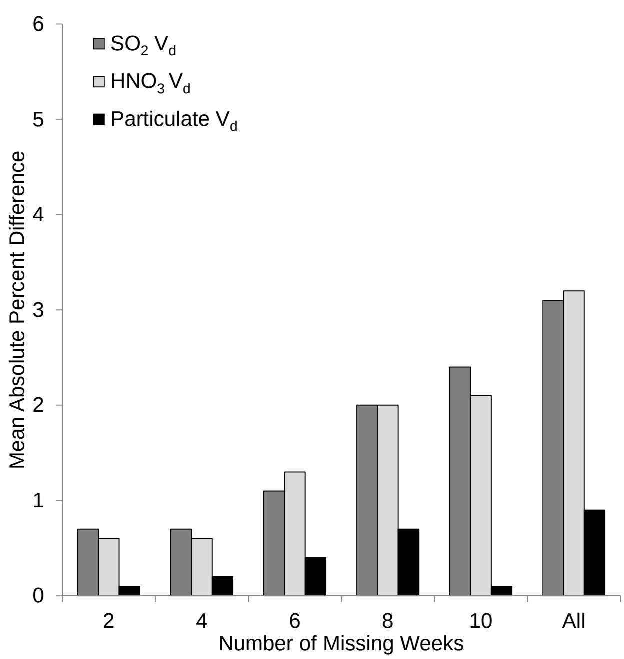


Figure 6 is based on substituting for missing wind speed values with data from LYK123 and Figure 7 is based on data from DCP114. The MAPD values in the two figures are less than 5 percent. The results were somewhat worse for particles than the other two pollutants.

4. Conclusions

The analysis of the comparison between 10-year mean deposition velocities shows that the practice of determining “near sites” based on geographic proximity and similar terrain and land use is not supported by an analysis of the data produced by the model. Of the 89 site pairs identified by seeking matches where the SO_2 V_d, HNO_3 V_d, and particulate V_d are all within ±10 percent, only five are currently used as “near sites,” and most of the site pairs identified were not located nearby geographically. From this analysis, it is recommended that the use of “near sites” for missing data replacement be terminated.

Evaluation of the results shows that flux estimates calculated using 10-year mean deposition velocities and annual mean concentrations compare reasonably well. Most comparisons varied between -20 and 20 percent. Results obtained using 10-year mean deposition velocities calculated using only years with four valid quarters were better than those calculated using all valid site-years (either three or four valid quarters). These results show that reasonable approximations of annual deposition estimates might be obtained using a summary deposition velocity such as a 10-year mean.

The most promising results of this study come from the replacement of missing meteorological parameters (specifically vector wind speed and sigma theta) with historical weekly averages of these parameters using data from the same site. These model runs were better than the results obtained by substituting for missing data using “near site” meteorological data.

Details of the replacement protocol will require further development including the number of years required to calculate the averages, the backup to weekly averages if they are unavailable, and the remedy for missing data at new sites without a historical record. Similar situations were successfully dealt with when CASTNET converted to calculation of atmospheric concentrations using local atmospheric conditions [14].

This study only examined a single site for one year. Further work in support of the development and testing of the final replacement protocol should perform a similar analysis on all sites for multiple years. Also, it would be beneficial to reconstruct maps and time series of dry and total deposition that are presented in CASTNET Annual Reports, e.g., Figures 3-3 through 3-10 from the CASTNET 2009 Annual Report, using the results herein and review the differences [15]. If results of the time series analyses are approximately the same, the finding would lend support and validation to the adoption of the replacement scheme.

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