NITROGEN DEPOSITION ONTO THE UNITED STATES AND WESTERN EUROPE

Summary:

This data set contains data for wet and dry nitrogen-species deposition for the United States and Western Europe. Deposition data were acquired directly from monitoring programs in the United States and Europe for time periods from 1978-1994 for wet deposition and from 1989-1994 for dry deposition and evaluated using similar quality assurance criteria to ensure comparability. A standard geostatistical method (kriging) was used to interpolate data onto a 0.5 x 0.5 degree resolution map for wet and dry deposition. Analysis of N deposition for these regions was limited by sampling density, frequency, and coverage.

These spatially explicit wet and dry N fluxes also provide a tool for verifying regional and global models of atmospheric chemistry and transport, and represent critical inputs into terrestrial models of biogeochemistry. These data can be used to construct continental scale N budgets and to evaluate recent modification of land-atmosphere N exchange and ecosystem function (Holland et al., 2004).

Data files of the site monitoring locations and monthly deposition averages are available in ASCII space-delimited format, and 0.5 x 0.5 degree gridded deposition values are available in both ASCII space-delimited format and ASCII grid format. The 11 mapped data images are available in .jpg format as companion files (e.g., Figs. 1 and 2). The complete set of 11 derived nitrogen-species 0.5 x 0.5 degree deposition maps is also available in .pdf format in the companion file ftp://daac.ornl.gov/data/global_climate/nitrogen_deposition/comp/N_Deposition_Maps_20040917.pdf. Other companion files include quality assurance plans and operating manuals available from and maintained by the United States and European monitoring networks.

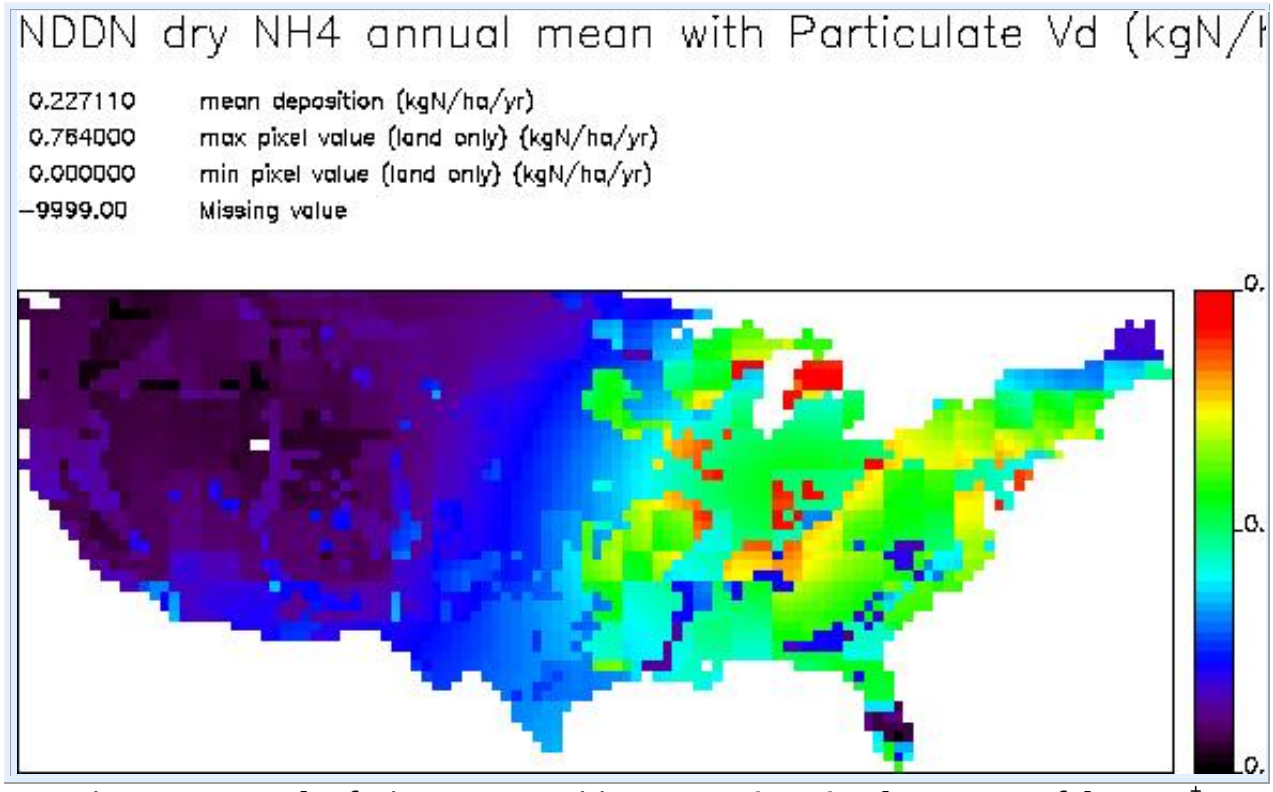

Figure 1. Example of Nitrogen Deposition Map -- 0.5 x 0.5-degree map of dry NH_4^+ deposition over the United States.

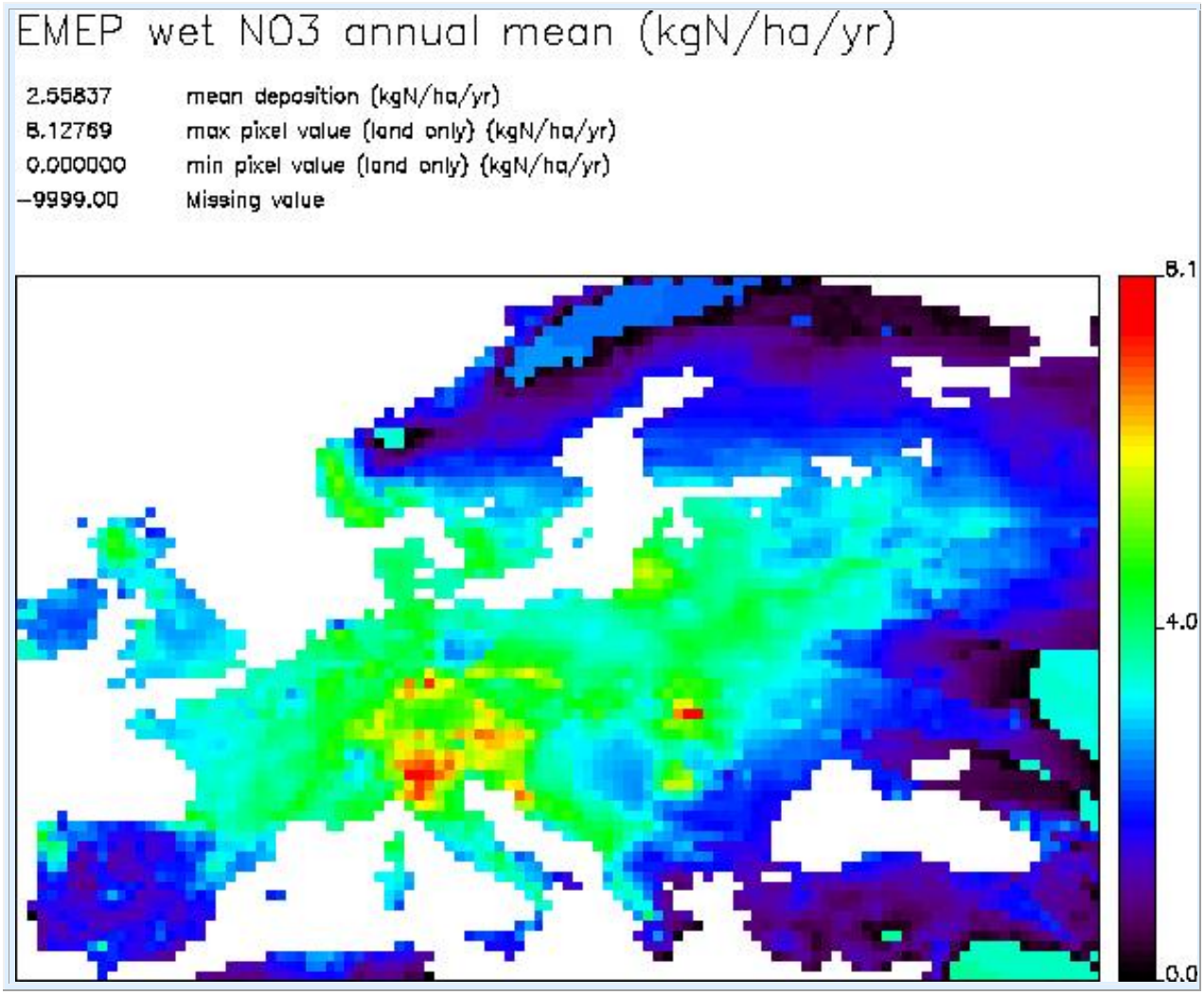

Figure 2. Example of Nitrogen Deposition Map -- 0.5 x 0.5-degree map of wet NO_3^- over Western Europe.

Data Citation:

Cite this data set as follows:

Holland, E. A., B. H. Braswell, J. M. Sulzman, and J.-F. Lamarque. 2005. Nitrogen Deposition onto the United States and Western Europe. Data set. Available on-line [http://www.daac.ornl.gov] from Oak Ridge National Laboratory Distributed Active Archive Center, Oak Ridge, Tennessee, U.S.A. doi:10.3334/ORNLDAAC/730.

Table of Contents:

1. Data Set Overview:

Project: Climate Collections

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Overview:

The documented acceleration of NH_3 and NO_x ($NO + NO_2$) emissions over the last 150 years has accelerated N deposition, compromising air and water quality and altering the functioning of terrestrial and aquatic ecosystems worldwide. To construct continental scale N budgets, we produced maps of N deposition fluxes from site-network observations for the US and Western Europe. Increases in the rates of N cycling for these two regions of the world are large and have caused profound modifications of land-atmospheric N exchanges, and ecosystem function.

We compiled observations from monitoring networks in the US and Europe, in order to construct 0.5 x 0.5 degree resolution maps of N deposition by species. In the United States, wet deposition data were provided by the National Atmospheric Deposition Program/National Trends Network (NADP/NTN, 1995; NADP/NTN, 2000; Lamb and Van Bowersox, 2000). Measurements of ambient air concentrations, used to calculate dry deposition fluxes, were provided by the National Dry Deposition Network (NDDN) and the Clean Air Status and Trends Network (CASTNet). In Europe, measurements of wet deposition fluxes of NO_3^- and NH_4^+ and ambient air concentrations of HNO_3, NO_2, particulate NO_3^-, and particulate NH_4^+ were provided by the Cooperative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe (European Monitoring and Evaluation Programme). Both the US and European measurements span the 1978-1994 for wet deposition and 1989-1994 for dry deposition with some sites beginning later than others.

The maps are necessarily restricted to the network measured quantities and consist of statistically interpolated fields of aqueous NO_3^- and NH_4^+, gaseous HNO_3 and NO_2 (in Europe), and

particulate NO_3^- and NH_4^+. A number of gaps remain in the data set including organic N and NH_3 deposition.

The interpolated spatially continuous fields allow estimation of regionally integrated budget terms. Dry deposition fluxes were the most problematic because of low station density and uncertainties associated with exchange mechanisms at the land surface. We estimated dry N deposition fluxes by multiplying interpolated surface air concentrations for each chemical species by model-calculated, spatially explicit deposition velocities (Holland et al., 2004). Our analysis of N deposition for these regions was limited by sampling density. The framework we present for quantification of patterns of N deposition provides a constraint on our understanding of continental bio-atmospheric N cycles. These spatially explicit wet and dry N fluxes also provide a tool for verifying regional and global models of atmospheric chemistry and transport, and represent critical inputs into terrestrial models of biogeochemistry.

2. Data Characteristics:

Data Descriptions

Measured Deposition Data

Data were compiled from wet deposition and dry ambient air concentration monitoring networks in the US and Europe and processed as described in Section 5 to derive monthly wet nitrogen deposition for each site (monthly dry deposition data for each site is not provided). These data were then interpolated into the 0.5 x 0.5-degree gridded annual deposition files as described below.

Data Source

For data sources refer to References/Measured Deposition Data Sources.

Measured Wet Deposition Data

Temporal Characteristics

Temporal Coverage

NADP: Wet deposition 1978-1994

EMEP: Wet/bulk deposition 1978-1994

Temporal Resolution

NADP: Data are monthly precipitation weighted means of wet deposition totals derived from weekly measurements.

EMEP: Data are monthly precipitation weighted means of wet deposition totals derived from daily measurements.

Measured Wet Deposition File Formats

NADP and EMEP wet deposition data are in the same space-delimited ASCII file format. The chemical form of nitrogen is specified in the filename (e.g., NADP_wet_deposition_nh4_sitedata_monthly.txt).

Parameter/Variable

Variable	Column Number	Units
Latitude (negative southern)	1	degrees
Longitude (negative western)	2	degrees
Elevation	3	meters_amsl
Year	4	yyyy
Month	5	1-12
Precipitation weighted mean concentration	6	mg N species/L*
Precipitation	7	cm/month
Deposition per area	8	Kg N species/ha/month*
Type of measurement**	9	Unitless

* - data are reported for "N species" NH4-N and NO3-N

** - 0 for bulk deposition and 1 for wet only deposition

Example Measured Wet Deposition Data File Record

(EMEP_wet_bulk_deposition_nh4_sitedata_monthly.txt)

emep_NH4-N_wet/bulk_deposition

latitude longitude elevation year month precipitation_weighted_mean_concentration precipitation deposition type_of_measurement

degrees degrees meters_amsl yyyy 1-12 mgNH4-N/L cm/month kgNH4-N/ha/month 0-bulk_1-wetonly

47.77 16.77 117 1989 2 0.926 2.85 0.26391 1

47.77 16.77 117 1989 3 1.397 1.93 0.269621 1

47.77 16.77 117 1989 4 1.241 7.05 0.874905 1

47.77 16.77 117 1989 5 0.856 5.36 0.458816 1

Gridded Deposition Data

The monthly average nitrogen deposition for EMEP (European) and NADP wet deposition monitoring networks were spatially interpolated onto a 0.5 x 0.5-degree grid as described in Section 5. For the dry deposition networks of EMEP and NDDN ambient air concentrations of nitrogen species were interpolated onto a 0.5 x 0.5-degree grid in the same fashion as wet deposition and an explicit dry deposition velocity was calculated for each grid cell. For each grid cell a dry deposition flux is calculated by multiplying the dry deposition velocity for a specified nitrogen species by the ambient concentration for that nitrogen species. Only the annual gridded dry deposition flux data are provided.

Spatial Characteristics

The nitrogen species deposition data for European and United States monitoring networks were spatially interpolated onto a 0.5 degree latitude by 0.5 degree longitude grid as described in Section 5. Nitrogen deposition estimates are for the center of the grid cell.

Spatial Coverage

The grid used for the EMEP data set (negative values are Western Longitude and Southern Latitude).

Corner	Latitude (degrees)	Longitude (degrees)
NW	70.50	-10.0
NE	70.50	44.50
SW	35.50	-10.0
SE	35.50	44.50

The grid used for the NADP/NDDN data set (negative values are Western Longitude and Southern Latitude).

Corner	Latitude (degrees)	Longitude (degrees)
NW	49.0	-124.0
NE	49.0	-66.5
SW	25.0	-124.0
SE	25.0	-66.50

Spatial Resolution

For both EMEP and NADP/NDDN the resolution is 0.5 degree vertical (latitude) by 0.5 degree horizontal (longitude)

Temporal Characteristics

Temporal Coverage

NADP: Wet deposition 1978-1994

EMEP: Wet/bulk deposition 1978-1994

NDDN Dry: 01/01/1989 - 12/31/1994

EMEP Dry: 01/01/1989 - 12/31/1994

Temporal Resolution

Data were gridded as an Annual mean of N deposition

Projection

Projection of Geographic (latitude-longitude)

Grid Description

The EMEP grid is 109 columns (longitude) by 70 rows (latitude). The NADP/NDDN grid is 115 columns (longitude) by 48 rows (latitude).

Gridded Data File Format

The gridded data files are in space-delimited ASCII format with the value for each pixel a floating point number.

Parameter/Variable Characteristics

Wet Deposition

Network	Deposition	Deposition Units	Gridded Deposition File
EMEP	NO3	KgN/ha/yr	EMEP_wet_deposition_no3_0.5x0.5_grid_annual.txt
EMEP	NH4	KgN/ha/yr	EMEP_wet_deposition_nh4_0.5x0.5_grid_annual.txt
NADP	NO3	KgN/ha/yr	NADP_wet_deposition_no3_0.5x0.5_grid_annual.txt
NADP	NH4	KgN/ha/yr	NADP_wet_deposition_nh4_0.5x0.5_grid_annual.txt

Dry Deposition

Network	Concentration		Deposition Velocity		Deposition Units	Gridded Deposition File
EMEP	HNO3+NO3	x	HNO3	=	KgN/ha/yr	EMEP_dry_deposition_hno3conc_no3_hno3vd_0.5x0.5_grid_annual.txt
EME	HNO3+N	x	NO3	=	KgN/h	EMEP_dry_deposition_hno3conc_no3.no3vd_0.

P	O3				a/yr	5x0.5_grid_annual.txt
EMEP	NO2	x	NO2	=	KgN/ha/yr	EMEP_dry_deposition_no2conc_no2vd_0.5x0.5_grid_annual.txt
EMEP	NH4	x	Particulate	=	KgN/ha/yr	EMEP_dry_deposition_nh4conc_particulatevd_0.5x0.5_grid_annual.txt
NDDN	HNO3	x	HNO3	=	KgN/ha/yr	NDDN_dry_deposition_hno3conc_hno3vd_0.5x0.5_grid_annual.txt
NDDN	NH4	x	Particulate	=	KgN/ha/yr	NDDN_dry_deposition_nh4conc_particulatevd_0.5x0.5_grid_annual.txt
NDDN	NO3	x	NO3	=	KgN/ha/yr	NDDN_dry_deposition_no3conc_no3vd_0.5x0.5_grid_annual.txt

Example Gridded Data File Record

NADP_dry_deposition_nh4conc_particulatevd_0.5x0.5_grid_annual.txt (1 record, wrapped multiple times shown. Full file has 115 columns and 48 rows.)

0.000 0.000 0.000 0.043 0.045 0.047 0.048 0.036 0.037 0.039 0.040 0.027 0.029 0.030 0.031
0.040 0.042 0.043 0.059 0.060 0.061 0.062 0.041 0.042 0.043 0.074 0.076 0.077 0.062 0.080
0.082 0.059 0.085 0.072 0.073 0.057 0.058 0.060 0.061 0.063 0.065 0.067 0.069 0.071 0.074
0.077 0.079 0.082 0.086 0.089 0.092 0.095 0.098 0.176 0.182 0.187 0.149 0.154 0.158 0.163
0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000
0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000
0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000
0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000 0.000

ASCII GRID versions of the original ASCII data files

As a service to our GIS users, we have provided ASCII GRID versions of the original ASCII data files included with this data set. ASCII GRID format includes 6 lines of header information (pulled directly from the documentation accompanying this data set) followed by cell values in row-major order. The file names and file format are described in the accompanying readme file.

3. Data Application and Derivation:

These spatially explicit wet and dry N fluxes allow verification of regional and global models of atmospheric chemistry and transport. The data and maps are critical inputs for terrestrial models of biogeochemistry. The data can be used to construct continental scale N budgets and to evaluate recent modification of land-atmosphere N exchange and ecosystem function. The data can be used to produce 0.5 x 0.5 degree maps of mean annual wet and dry nitrogen-species deposition for the United States and Western Europe.

4. Quality Assessment:

For quality assurance (QA) and quality control (QC) of the site data used in the interpolation, the user is referred to the NADP QA/QC documentation (Lehmann and Bowersox, 2003), which is

provided as a companion file, and the NADP web site [http://nadp.sws.uiuc.edu/QA/]. The NADP quality criteria 1-3 were applied to EMEP data. Criterion 4 does not pertain to the EMEP data set because data from a separate rain collector were not reported.

For the European EMEP data sets, we followed a similar quality assurance plan as that used by NADP to ensure comparability. The NADP quality assurance criteria required in order to include a site for a year are the following:

1) There must be valid samples for at least 75% of the summary period.

2) For at least 90% of the summary period there must be precipitation data either from the rain gauge or from the sample volume.

3) There must be valid samples for at least 75% of the total precipitation reported for the summary period.

4) For the entire summary period the total precipitation as measured for the sample volume must be at least 75% of the total precipitation measured by the rain gauge for all valid samples where both values are available (NADP only).

5. Data Acquisition Materials and Methods:

For description of collection methods of the European Monitoring and Evaluation Programme (EMEP), the user is referred to the EMEP operations manual (EMEP 2001), which is provided as a companion file, and the EMEP web site [http://www.nilu.no/projects/ccc/submission.html].

For description of collection methods of the wet deposition NADP site data the user is referred to the NADP operations manual (Dossett and Bowersox, 1999), which is provided as a companion file, and the NADP web site [http://nadp.sws.uiuc.edu/lib/manuals/opman.pdf].

For description of collection methods of the ambient air concentrations, used to calculate dry deposition fluxes, of the National Dry Deposition Network (NDDN) and the Clean Air Status and Trends Network (CASTNet), the user is referred to the CASTNet web site [http://www.epa.gov/castnet] (US EPA, 2003).

Data and Methods

We compiled observations from monitoring networks in the US and Europe, in order to construct 0.5 x 0.5 degree resolution maps of N deposition by species. In the United States, wet deposition data were provided by the National Atmospheric Deposition Program/National Trends Network (NADP/NTN, 1995; NADP/NTN, 2000; Lamb and Van Bowersox, 2000). Measurements of ambient air concentrations, used to calculate dry deposition fluxes, were provided by the National Dry Deposition Network (NDDN) and the Clean Air Status and Trends Network (CASTNet). In Europe, measurements of wet deposition fluxes of NO_3^- and NH_4^+ and ambient air concentrations of HNO_3, NO_2, particulate NO_3^-, and particulate NH_4^+ were provided by the Cooperative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air

Pollutants in Europe (European Monitoring and Evaluation Programme). Both the US and European measurements span the 1978-1994 for wet deposition and 1989-1994 for dry deposition with some sites beginning later than others.

Wet Deposition Measurements

At the NADP/NTN sites within the US, precipitation is accumulated in buckets that are triggered to open at the onset of rain. Samples from precipitation buckets are gathered weekly and are sent to a central laboratory for chemical analyses. The chemical analyses include measurement of concentrations of hydrogen ions H^+, associated anions (SO_4^{2-}, NO_3^-, Cl^-), and base cations (NH_4^+, Ca^{2+}, Mg^{2+}, K^+, and Na^+). Quality control is also checked by evaluation of the charge balance of the samples.

In Europe, the method of sampling varied from location to location, country to country, and, in general, differed from the method used in the US. In Europe, samples of precipitation were collected daily using either the precipitation-only samplers (described above) or bulk sampling devices which are continually open to the atmosphere, and are thus more subject to contamination. Chemical sampling and analyses were usually done on a country-by-country basis. Inter-laboratory quality control tests were conducted annually . Some of the sites collected data monthly rather than daily (Lazaropole, former-Yugoslovia; Neuglobsow, Germany; Jarczew, Poland; Rarau, Seminic, Paring, Fundata, Turia, and Masun Romania; and Ivan Sedlo, Bosnia-Herzogovina). These data were the only available observations for large areas and so we included them in our analysis despite the potential problems associated with monthly sampling. The European data were published as the EMEP data report for 1995 (Hjellbrekke et al., 1997).

The wet deposition data were averaged annually for each measurement site. For the US, monthly means were acquired directly from the NADP/NTN and were summed for an annual measurement. For the European EMEP data sets, we followed the same quality assurance plan used by NADP to ensure comparability. The quality assurance criteria required in order to include a site for a year are the following: 1) There must be valid samples for at least 75% of the summary period. 2) For at least 90% of the summary period there must be precipitation data either from the rain gauge or from the sample volume. 3) There must be valid samples for at least 75% of the total precipitation reported for the summary period. 4) For the entire summary period the total precipitation as measured for the sample volume must be at least 75% of the total precipitation measured by the rain gauge for all valid samples where both values are available. Criterion 4 does not pertain to the EMEP data set, because data from a separate rain collector were not reported. In the US, the number of sites which met the completeness criteria varied between 21 and 203 with a mean of 156 sites over the time period. The number of sites was smallest as the network was starting in the late 1970s, and largest between 1985 and 1994, when the number of sites rose to approximately 200. Across Europe, data from 113 out of 188 sites satisfied the completeness criteria. There were 18 sites at the beginning of the monitoring period, growing to 50-75 participating sites during 1986-1994, with a mean of 50 sites in any given year.

For both the United States and Europe, precipitation weighted mean concentration was calculated as follows:

$$C_w = \frac{\sum_{i=1}^{n} C_i P_i}{\sum_{i=1}^{n} C_i P_i} \qquad (1)$$

C_w is the precipitation-weighted mean in units mg/L, calculated from the n valid samples for the season. (Each individual valid sample concentration C_i is weighted by the individual precipitation amount P_i for the sample. This calculation makes the assumption that the chemistry of the invalid samples is represented by the chemistry of the valid samples for that summary period.

$$F_{wet} = C_w P_t \quad (2)$$

where F_{wet} is the wet deposition flux, mmoles liter^{-1}, P_t, mm $H_2O$is the total precipitation over the averaging time period (i.e. (mmoles molecular weight l^{-1} mm precipitation)/100=kg ha^{-1}).

Dry Deposition Measurements

For both regions, dry deposition fluxes were calculated by multiplying the mapped ambient air concentration of the chemical species by a calculated mapped deposition velocity. In the US, atmospheric concentrations of trace species were measured by pumping ambient air through a filter pack containing 3 filters: a Teflon filter for dry deposition of aerosols (particulate NO_3^- and NH_4^+), a nylon filter for collection of gaseous HNO_3, and dual potassium carbonate-impregnated cellulose filters for collection of SO_2 (Edgerton, 1992). The filter packs were changed weekly, and filter extracts were analyzed in a central laboratory within 72 hours of filter extraction. Similarly, in Europe the atmospheric concentrations of trace species were made by pumping ambient air through a filter pack . However, the filters and flow rates varied from country to country. For example, in Italy, particulate ammonium was sampled using a Teflon filter (Gelman Zeflour with a 1 micron pore size) with a flow rate of 17 m^3 day^{-1}. In Hungary, particulate ammonium was sampled using a Teflon filter (Kipzer Paraplan) with a flow rate of 25 m^3 day^{-1}. There were similar contrasts for gas sampling. For example, in Italy, NO_2 concentrations were measured by a chemiluminescence technique while in Hungary, NO_2 was measured using a Triethanolamine solution with a 0.5 m^3 day^{-1} flow rate. The measurement techniques are summarized in Tables 1.1-1.3 on pages 43-46 of the EMEP data report . In addition, there was year to year variation in the measurement techniques. Hicks et al. (1991) compare various approaches and review the associated problems.

Neither the US nor the European dry deposition measurements spanned the whole 1978-1994 time period. In the US, some sites began measurements in 1989 with additional sites added in 1990 for a total of 65 sites. In Europe, the number of sites varied with the chemical species

considered: 33 sites sampled gaseous HNO_3 and particulate NO_3^-, 39 sites sampled particulate $NH_4^{+,}$ and 50 sites sampled gaseous NO_2. For both the US and Europe we used the available data to calculate monthly average concentrations taking into account the differences in sampling frequency.

We calculated the deposition velocity in a way that was consistent for both the US and Europe, estimating surface exchanges of chemical species based on the resistance approach developed by Wesely (1989). The dry deposition flux, F_{dry}, molecules m^{-2} s^{-1}, was calculated as the product of deposition velocity (V_d, $m\ s^{-1)}$ and concentration (C, molecules m^{-3}):

$$F_{dry} = V_d C \quad (3)$$

and the deposition velocity was assumed to be inversely proportional to the sum of three resistances

$$V_d = (r_a + r_b + r_c)^{-1} \quad (4)$$

where r_a is the aerodynamic resistance, r_b the resistance of the laminar sub-layer between the surface and the turbulent boundary layer, which depends on the diffusivity of the species, and r_c the bulk surface resistance that depends on land surface characteristics and on the solubility and reactivity of the chemical species. In general, r_b is small, r_a and r_c depend on a variety of parameters which in turn depend on both the land cover and the chemical species considered. For particulate fluxes, we assumed that the relevant particles had a diameter between 0.1 and 1 μm (Wyers and Duyzer 1997), and applied the u* parameterizations suggested by Wesley (1985) and Erisman and Draaijers 1995.

The V_d calculation requires maps of meteorological variables and land cover. Site specific meteorological data were not available for many of the European sites. Therefore we used model results from the NCAR/NCEP re-analyses to provide a regionally consistent and spatially-explicit data set containing the meteorological information necessary to compute deposition velocities: pressure, temperature, wind, and precipitation.

To assign land cover classes, we used the 8-km resolution land cover map of Defries and Townsend (1995), spatially re-aggregated to 0.5 x 0.5 degrees, in order to assign land cover classes. Implementation of the resistance model (which has its own land surface types) was accomplished by matching the Wesley land cover classes to the DeFries and Townsend classes. To match the relatively high resolution of the land cover data, the meteorological data were also interpolated from their original resolution (T42 or approximately 2.8) to 0.5 x 0.5 resolution. All V_d output was mapped at 0.5 x 0.5 resolution globally. Finally, dry deposition fluxes were mapped by statistically interpolating the ambient concentrations of each chemical species and then multiplying the interpolated concentrations by the calculated deposition velocities for each 0.5 C grid cell.

Critical evaluation of the initial V_d maps for the US and Europe uncovered a problem introduced by the mismatch in spatial resolution of the land cover map (8 by 8 km) and the meteorological data (2.5 by 2.5). The high wind speed modeled over the oceans introduced spuriously high

deposition velocities for some of the coastal grid cells because the meteorological data was produced on a much larger grid than the land cover data. The actual V_d calculations were done in an intermediate grid size (0.5 by 0.5) to reduce the problems associated with the spatial mismatch, but did not solve the problem entirely. The deposition velocities calculated on land with the high ocean wind speeds were excluded from our calculations by generating a cell masking routine. The average deposition velocity of surrounding grid cells with the same vegetation type was re-substituted to complete the maps of V_d. The problem underscores the difficulty and importance of careful spatial analysis.

Deposition of aerosols on forests differs from deposition onto low-stature vegetation . A physically-based parameterization distinguishing the two classes would be ideal but is problematic for a one-dimensional model because of subgrid scale horizontal heterogeneity. Therefore, we relied on the empirical derivation of Wesley (1989) to calculate the deposition velocities for particulate NO_3^- and NH_4^+. To further examine the sensitivities of the calculation, we revised the model to include the influence of forest type as described by Jonas and Heinemann (1995):

$$V_{d,i} = \varepsilon V_{d,ref} \quad (5)$$

where $V_{d,ref}$ is the deposition velocity of the "reference forest type", ε is a multiplying factor, and $V_{d,j}$ represents the modeled V_d for individual forest types in the domain. The multiplying factor was calculated as the mean of the factors calculated for all of the forest types sampled to yield 6.71 for deciduous forests, and 10.4 for evergreen forests, the only vegetation types to which we applied equation 5.

A further complication was introduced by the fact that in Europe, HNO_3 (g) and particulate NO_3^- were combined and reported as a single value by EMEP. It was impossible to partition the sums into their individual components based on the available data. Thus, for the sum of HNO_3 (g) plus particulate NO_3^-, we calculated two dry deposition fluxes as a means of bracketing the estimate: one as if the flux were made up entirely of particulate NO_3^- and another as if the flux were made up entirely of the HNO_3 (g). This necessarily introduces an additional uncertainty because the deposition velocities of the HNO_3 (g) and particulate NO_3^- differ by as much as 5 fold.

Geostatistical Methodology

To create the interpolated fields of wet deposition flux and dry species concentrations, we used a variant of a standard geostatistical method, the Moving Window Residual Kriging (MWRK) algorithm developed by Haas (1990, 1995). This analysis/interpolation tool was developed originally for use with atmospheric deposition network data. Kriging is a statistical method of providing unbiased estimates of variables in regions where the available data exhibit spatial autocorrelation, and "Kriging estimates" are obtained in such a way that they have minimum variance . In the Haas (1995) algorithm, a moving window is used to isolate sub-regions of the data for calculation of variograms, which is critical because the distribution of deposition at the continental scale is typically not normal. This kriging application also allows the application of prior information (covariates) that are correlated with the data of interest and available at all grid cell locations.

We found the MWRK method to be a satisfying alternative to other interpolation methods (e.g. cubic splines, bilinear/nearest neighbor approaches), for several reasons: (1) the use of spatial covariates such as precipitation and elevation; (2) temporal correlations in the data to be included in the local model; and (3) estimates of uncertainty via cross-validations (site-by-site removal) and by including confidence intervals for the estimates (based upon kriging variance). The uncertainty estimates also provide a means to evaluate the importance of the inclusion of spatial covariates, and the value of including the temporal correlation in the spatial estimate. The spatial-temporal aspect of the analysis also allows examination of regionally integrated trends using simple regression techniques, thereby avoiding the need for multi-layer hypothesis testing (e.g., Stoddard 1994, Stoddard et al. 1998), which is a common approach.

Kriging of the wet deposition measurements and the dry deposition measurements were handled somewhat differently. Spatial interpolation of the annual mean site observations of wet deposition of NO_3^- and NH_4^+ were performed using moving window (no time dimension) kriging (Haas, 1990) with precipitation and elevation covariates. The analysis was performed using 237 sites for the wet deposition estimates within the United States, and 115 sites for the wet deposition estimates within Europe. Data from three European sites were excluded from the kriging analysis because they contained questionably high deposition values: Lesogorrsky, Russia; Stina de Vale, Romania; and Ispra, Italy.

The choice of precipitation and elevation as covariates was made after evaluating the effects of a wide range of variables (e.g., temperature, humidity) on kriging model statistics. These statistics were based on a series of cross-validation studies where each site (in turn) is withheld from the analysis, and the distribution of residuals (modeled minus predicted deposition) is examined. For the US, we used gridded precipitation and elevation information from the VEMAP Phase I data set . For Europe, we used the Leemans and Cramer (1991) global data set. All covariates and our desired output base map for both regions are defined as before on a 0.5 x 0.5 grid.

Kriging of the ambient concentrations of dry species (HNO_3, NO_2, particulate NH_4^+, particulate NO_3 , HNO_3 plus particulate NO_3^-, and particulate NH_4^+ plus NH_3) was performed using a modification of the MWRK called Moving Cylinder Residual Kriging . This approach extends the circular spatial window along the time axis to form a cylinder. The time dimension is included in order to exploit the existence of temporal correlations in the observations, which are significant in the dry species concentrations. This approach is based on the addition of temporal variograms in the kriging estimation. Cross-validation analysis yielded a "cylinder length" of 3 months so that an estimate at time *t* used information (observations) from times *t*-1 (month) and *t*+1 month. In Europe, the dry deposition data lacked the temporal coherence needed to defend application of the Moving Cylinder Residual Kriging technique. Thus, kriging of the ambient concentrations of NO_2, HNO_3 and particulate NO_3^- , and particulate NH_4^+ were done using the MWRK with no covariates.

6. Data Access:

This data is available through the Oak Ridge National Laboratory (ORNL) Distributed Active Archive Center (DAAC) or the EOS Data Gateway.

Data Archive Center:

Contact for Data Center Access Information:

E-mail: uso@daac.ornl.gov
Telephone: +1 (865) 241-3952
FAX: +1 (865) 574-4665

Product Availability:

Data can be accessed electronically on the ORNL DAAC's anonymous FTP site or provided on CD-ROMs.

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