Review of Dry Deposition: Available Measurements to Evaluate Models

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Dry deposition, which is the removal of gases and aerosols at the earth's surface, provides an important sink for many trace gases, e.g., ozone (O3) and sulfur dioxide (SO2), and aerosols. This sink is commonly considered in models by calculating the dry deposition flux as the product of the surface layer concentration and the dry deposition velocity (Vd). The latter expresses the efficiency of the turbulent transport, molecular diffusion (and sedimentation of aerosols) and uptake by the various substrates such as the leaf stomata of the vegetation cover, soils, snow and ice sheets and the oceanic surface layer. Hence, evaluation of the simulated dry deposition process can be done either by comparing dry deposition fluxes, which also reflects the quality of the concentration calculations and thereby the representation of other relevant processes, or the dry deposition velocity, which only expresses the representation of processes that control dry deposition. The latter only makes sense if an explicit dry deposition model has been incorporated, such as the quite commonly applied "big leaf" dry deposition scheme for large-scale models [e.g., Wesely 1989; Ganzeveld et al., 1995].

Most optimally a model evaluation should include a comparison of the fluxes as well as the dry deposition velocities. Preferably, the model comparison should also use long-term (> months to years) dry deposition measurements to evaluate the simulated seasonal cycles in dry deposition fluxes and velocities. It is not likely that large-scale models, e.g., global chemistry and transport models, realistically capture the short-term (days-weeks) variability in dry deposition fluxes related to the short-term variability in controlling parameters like radiation, turbulence and precipitation. However, changes in the dry deposition fluxes and velocities related to seasonal changes in surface cover, e.g., snow cover, vegetation activity and micro-meteorology should be fairly well represented in the models. The model evaluation should also include a selection of trace gases (or aerosols) that can provide complementary information on the model representation of the various processes. For example, the dry deposition of nitric acid (HNO3) is mainly controlled by turbulent transport to the surface whereas O3 and SO2 dry deposition velocities over (dry) vegetation are largely controlled by stomatal uptake. Including these trace gases in the evaluation of the model representation of dry deposition can thus provide an indication about the quality of simulated turbulent exchange as well as the biological processes involved.

There are generally two types of dry deposition estimates available 1) those that apply the inferential technique, which is based on a set of measurements including meteorological and biological parameters and trace gas concentrations which are then subsequently used in combination with a multi-layer dry deposition model [e.g., Meyers, 1998] to estimate the dry deposition velocity and flux and (2) direct measurements of the dry deposition fluxes using eddy covariance or equally intensive micrometeorological techniques. The disadvantage of the first set of measurements is that they involve the use of a model, which inevitably introduces inaccuracies dependent on the model representation of the processes involved. The disadvantage of the second set of measurements is that these are often limited to a small set of species. For example, eddy covariance measurements require sensors that can sample at a frequency > 1 Hz, which basically limits the use of

this technique to gases like O3 and oxidized nitrogen (NOy, which includes species such as NO, NO2 and HNO3).

Alternative measurement techniques such as the gradient and eddy-accumulation technique have been developed and deployed to cope with this problem but haven't resulted yet in a large number of reliable long-term direct dry deposition flux measurements of multiple trace gases at one site. The US EPA has a network, known as CASTNet (Clean Air Status and Trends Network), of dry deposition stations using the inferential method [e.g., Clarke et al., 1997]. A great deal of data from that network is available on the web at www.epa.gov/castnet/. Data that can be found are for example annual or weekly estimates of the concentrations, deposition velocities and fluxes of gases like O3, SO2, sulfate (SO42-), particulate nitrate (NO3-), ammonium (NH4+) and HNO3. In addition, meteorological and specific site descriptions have been provided. To indicate the quality of the dry deposition estimates provided by CASTNet, annual average precisions statistics were calculated for duplicate meteorological and concentration measurements collected at five CASTNet sites in 1991 [Clarke et al., 1997]. The precision of the calculated O3 and SO2 dry deposition flux is about 8%, for HNO3 and SO42- about 12% and 17% for the NO3- flux. A site where many of NASA's field campaigns data are posted is the Global Tropospheric Experiment (GTE) archive: http://asd-www.larc.nasa.gov/ or ftp://ftp-gte.larc.nasa.gov/.

The measurements of O3 exchange fluxes (and some NOy) measured during the ABLE campaigns (ABLE-2B, (tropical forest) [e.g., Bakwin et al., 1990], ABLE-3A (tundra) [Jacob et al., 1992] and ABLE-3B (Taiga woodland) [Bakwin et al., 1994]) can be downloaded from this site. However, these ABLE datasets are not covering a long time span, generally some weeks up to months of observations. A site where a dataset with long-term eddy covariance O3 and NOy flux measurements can be found is: http://www-as.harvard.edu/data/nigec-data.html. The measurements have been collected since 1990 at the Harvard Forest site over a deciduous forest in Massachusetts, USA [Munger et al., 1996]. At: http://medias.obs-mip.fr/idaf/, data on dry and wet deposition measured within the IDAF program at 9 sites in Africa and one in French Guyana can be found. The IDAF program has been initiated within the scope of the DEBITS (Deposition of Biogeochemically Important Trace Species) program, in order to study atmospheric deposition in tropical regions. In fact, there is not (yet) a DEBITS dataset available for the other regions where the DEBITS program is active (Asia and South America). Concerning dry deposition, the database provides an indirect estimate of dry deposition fluxes based on the measured monthly mean gas and aerosol concentrations (for more details concerning the applied technique see the internet site) and estimated dry deposition velocities based on references or models. The gas concentration measurements include SO2, HNO3, NH3, NO2, and O3. The aerosol measurements are based on collecting the bulk particles on a filter followed by chemical analysis to determine the water-soluble content and the inorganic anions and cations of the aerosol. In Europe, there is no operational dry deposition measurement network similar to that in the US. However, long-term measurements (about 5 years) of dry deposition fluxes in Europe are available for three sites: Auchencorth (CEH, UK: http://www.ceh.ac.uk/aboutceh/sections/edin_pollution.htm), Melpitz (IFT, Germany: http://www.tropos.de/FORSCHUNG/index.html) and Speulderbos (RIVM, Netherlands).

Alternative sources of deposition data are the studies that focus on regional-scale air quality issues, like the data provided by the EMEP program: http://www.emep.int/index_data.html. These data, which cover most of Europe, are not based on direct measurements of dry deposition fluxes but estimated wet and dry deposition fluxes using the EMEP Lagrangian Acid Deposition Model (LADM). Since these estimates are calculated at a fairly high spatial resolution (150 x 150 km2), use of these data to evaluate large-scale models running at a coarser spatial resolution could be considered. The data that are available are the yearly-calculated total deposition values for oxidised sulphur (SOx), NOy and reduced nitrogen (NHx) for the period from 1985 to 1996 for the whole EMEP grid. Finally, for an overview of alternative datasets (not always online available) for model evaluation we refer to Wesely [2000], who provides a comprehensive overview on the status on the research on dry deposition, and Ganzeveld et al. [1995] and Ganzeveld and Lelieveld [1998], whom collected O3, SO2 and SO42- dry deposition velocity measurements for evaluation of their global scale dry deposition model.

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