APPLICATION OF A LAGRANGIAN RANDOM PARTICLE MODEL TO FORWARD AND INVERSE AIR QUALITY MODELING

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Abstract: Two methods of using a Lagrangian random particle dispersion model in air quality simulations will be presented. For the purpose of source-oriented modeling, a hybrid Lagrangian dispersion and Eulerian chemistry were coupled to study the effects of ozone in a coastal region. The Lagrangian model was providing transport and dispersion of ozone and ozone precursors, while a chemical module was treating chemical transformations within a Eulerian framework. The Lagrangian particles had chemical dimensions that were updated after each chemical model step. This hybrid model allowed for the treatment of air parcels from numerous sources and reproduced their non-linear chemical evolution.

For the purpose of receptor-oriented modeling, the Lagrangian random particle model was adapted to be run in an inverse mode to determine the most probable sources impacting particular receptors. This inverse modeling improved source location estimates of the backtrajectory analysis and standard receptor models. These two methods indicate that the Lagrangian random particle model can be a useful tool in improving the accuracy of air quality models. Moreover, due to the fact that the Lagrangian framework can be applied to the highest resolutions, these approaches are appropriate for any complexity of regional, mesoscale, and microscale domains.

1. INTRODUCTION

Air quality models in both Eulerian and Lagrangian frameworks of reference have advantages and disadvantages when applied to various environmental research and application studies. While Lagrangian models accurately represent the transport and dispersion and do not exhibit problems related to numerical diffusion, there are problems in representing full chemistry transformations. In addition, the Lagrangian models can be used for so called "inversion modeling" that allows for estimation of the most probable emission sources based on the receptor measurements and "inverse" flows. Eulerian models currently better represent the complexity of chemical transformations and interactions and emission sources, but they are limited in representation of the dispersion and numerical accuracy. Consequently, there is a need for modeling systems that utilize the advantages of both of these approaches and promote the use of Lagrangian modeling in research and regulatory air quality applications.

2. HYBRID MODELING USING A LAGRANGIAN STOCHASTIC MODEL COUPLED WITH METEOROLOGICAL AND ATMOSPHERIC CHEMISTRY MODELS

A hybrid modeling system (Weinroth et al., 2008) has been developed that links a meteorological model Mesoscale Model 5 (MM5) (Grell et al., 1994), a Lagrangian stochastic dispersion model (Koracin et al. 2007), and a Eulerian atmospheric chemistry box model (Seefeld, 1997). The modeling system can follow individual air parcels from various sources and reproduce their nonlinear chemical transformations and concentration impacts.

Each of the m-particles in the considered domain has a multi-dimensional identifier to track its specific attributes including location, emission source, chemical composition, origin, and chemical production and loss mechanisms over time and space as well as meteorological parameters. Each particle "i" is uniquely identified by the multi-dimensional identifier:

$$Particle_i(x, y, z, T, RH, p, chm_1, ..., chm_j, ID, t)$$

where *x, y*, and *z* are the coordinates, *T* is the ambient temperature, *RH* is the relative humidity, *p* is the atmospheric pressure, *chm* is the chemical composition of each considered pollutant, *"j"* is the total number of considered pollutants, *ID* is an identification number for each particle, and *"t"* is the time. Each specie concentration at any time step is the sum of the background and particle contributions. The Lagrangian stochastic model transports and disperses hypothetical particles in every time step (usually 10-60 sec). At every prescribed "chemistry time step" (usually an hour), the particles are "frozen" and subjected to the Eulerian chemistry in each of the grid boxes where they are present.

For each chemistry time step, Δt, there are *k*-particles in each grid cell. These *k*-particles were separated into the *n*-chemical species. The mass $P_{m,n}$ of a specie *n* of each m^{th} particle were then summed to the total species mass $P_{T(n)}$.

For each chemical species, the total mass was converted into a concentration in each grid-cell. At the end of each time step (time equal to $t+\Delta t$), the predicted concentration of each species within each grid-cell was then converted into mass $P_{T(n)}|_{t+\Delta t}$.

Distribution of newly produced chemical species takes into account the diffusion, time scale, mixing height, and turbulence intensity in terms of an empirical factor f (in this study f=0.05) for the intermixing efficiency. The apportionment was calculated by:

$$P_{(m,n)}|_{t+\Delta t} = B_{(n)} * f \Big/ N_n \qquad (1)$$

where N_n is the number of particles without chemical species (n) and $B_{(n)}$ is the production result between the time steps (t, $t+\Delta t$) for particles that had the new chemical species. Thus,

$$P_{(m,n)}\,|_{t+\Delta t} = \left(P_{(m,n)}\,|_t \Big/ P_{\mathrm{T}(n)}\,|_t \right) \left[p_{\mathrm{T}(n)}\,|_{t+\Delta t} - \left(B_{(n)} * f \right) \right] \tag{2}$$

In summary, the particle contribution of each species was calculated in each 3-D grid-cell by summation of particle-masses within the cell. The resulting concentrations are then converted back to masses and the multi-dimensional identifiers are re-assigned for each particle. After that, the Lagrangian model is then resumed for transporting and dispersing the particles. This process is then repeated over the simulation period.

A schematic in Figure 1 shows the grid structure of the Lagrangian and Eulerian modules, and illustrates ozone as one of the chemical components.

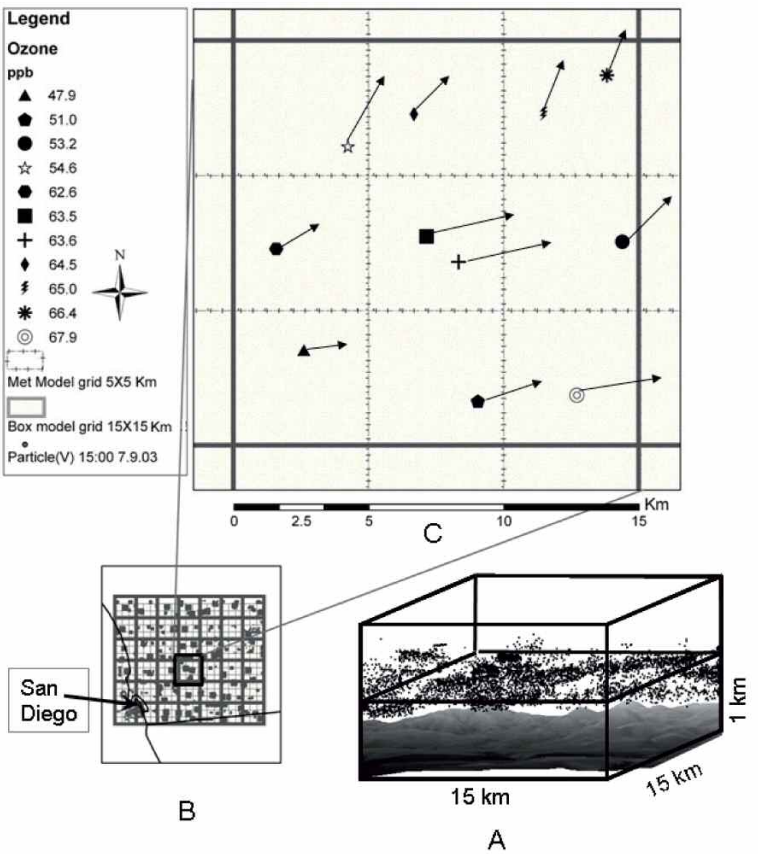

Figure 1. Illustration of the grid structure for the hybrid modeling system (atmospheric, Lagrangian, and box chemistry models) with an example of one chemical component – ozone.

This hybrid modeling system was applied to a case study of elevated ozone over San Diego, California. The evaluation of the modeling system using aircraft measurements is encouraging (Weinroth et al., 2008), and indicates the possibility of using this type of model for other research and possibly regulatory application in the future.

3. INVERSE MODELING USING A LAGRANGIAN STOCHASTIC DISPERSION MODEL

A study supported by the U.S. EPA focused on evaluation of the multivariate and trajectory-based receptor models for regional source apportionment relevant to the USEPA Regional Haze Rule. Model-simulated data for two northeastern U.S. sites has been used to evaluate the receptor models: Brigantine National Wildlife Refuge (NWR), New Jersey, and Great Smoky National Park (NP), Tennessee. The one objective of this project was to produce the synthetic data sets that could be used to assess and provide guidance for using receptor models for regional source apportionment. Simulated data were generated with the Community Multiscale Air Quality (CMAQ) model (U.S. Environmental Protection Agency, 1998). This synthetic data include concentrations of PM2.5 mass, ions (sulfate,

nitrate, chloride, ammonium, and sodium), elemental and organic carbon, major crustal species, other metal oxides, and trace elements. The selected modeling domain (Fig. 2) includes seven source regions in the eastern and central U.S. for which the MM5 model was previously run for the year 2002 at a resolution of 12 km. This spatial resolution is needed to generate accurate backward trajectories for the trajectory-based receptor models that are being evaluated as part of this study.

As a part of the back-trajectory analysis, a Lagrangian stochastic particle dispersion model (Koracin et al., 2007) was used in an inverse mode to estimate the most probable source areas starting from receptor locations. By using inverse modeling, probabilities were established for the receptor-source relationship between a single receptor and many source elements. The inverse modeling included explicit calculation of a source-receptor relationship in terms of linear transformations (i.e., in a matrix form) to describe the relative importance of specific subsets of the source to the impact at a certain receptor site. The elements of the receptor-source matrix simply represent the residence time spent in the respective source grid cell by the particles released from the receptor.

As an example, a backward simulation for 7 days was conducted with the particle model that started at 20 July 2002 at 00 UTC (local time: 19 July 2002 at 1900 EST). The CMAQ-derived sulfate concentrations showed a value of 18968 (ng/m^3) on 19 July 2002 with relative maximum contributions from regions 1, 3 and 5. Figure 5 shows the instantaneous position of the hypothetical particles at -168 hours obtained from the particle model. The direction of the particle transport corresponds well with the HYSPLIT back trajectories.

A preliminary investigation showed that the correlation coefficients between the CMAQ simulated average daily sulfate concentrations and the particle concentrations from the particle model for regions 1 to 7 lie in the range 0.78-0.97. Notice that the spread of the particles indicates a range of probability of determining most probable source areas. Since the stochastic particle model accounts for uncertainties in accurate predictions of the turbulence, it is important to assess relative ratios (not occurrence of individual particles) among the regions to identify the most likely sources.

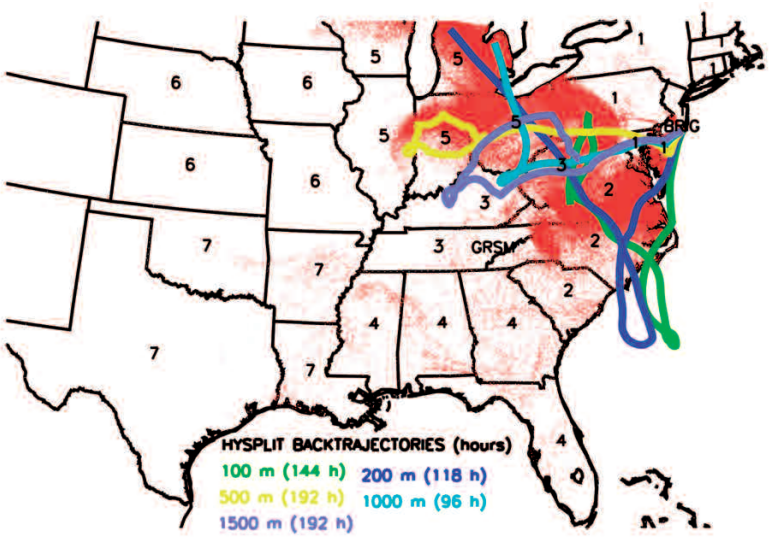

Figure 2. Instantaneous position of the simulated particles obtained from the Lagrangian stochastic dispersion model at 7 days in the inverse run mode (for clarity, particles over the ocean and water bodies are excluded in the plot). HYSPLIT derived backtrajectories from receptor site BRIG at different heights (100, 200, 500, 1000, and 1500 m) are overlaid on the plot.

An enhanced capability of air quality modeling using this hybrid modeling system and inverse modeling, among other qualities, indicates that Lagrangian stochastic models should be an essential component of research and also regulatory air quality modeling.

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