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A computationally efficient model for estimating background concentrations of NO_x, NO_2, and O_3

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ABSTRACT

We formulate a Lagrangian model to supplement comprehensive Eulerian grid models such as *CMAQ*, to estimate concentrations of NO_x, NO_2, and O_3 averaged over a spatial scale of the order of a kilometer over a domain extending over hundreds of kilometers. The model can be used to estimate hourly concentrations of these species over time periods of years. It achieves the required computational efficiency by separating transport and chemistry using the concept of species age. The model computes concentrations by tracing the history of an air parcel reaching a receptor through back trajectories driven by surface winds. Chemical reactions within the parcel are modeled through the Generic Reaction Set (GRS) chemistry model, which approximates the photochemical processes that lead to the production of ozone. The model is evaluated with concentrations measured over two years, 2005 and 2007. Evaluation with data measured at 21 stations distributed over the California South Coast Air Basin (SoCAB) during 2007 indicates that the model provides an adequate description of the spatial and temporal variation of the concentrations of NO_2, and NO_x. Estimates of maximum hourly O_3 concentrations show little bias (less than 10%) compared to observations, and the scatter ($s_g^2 \leq 2.56$–95% confidence interval of the ratio of predicted to observed concentrations) is comparable to those associated with more computationally demanding models. The model was also evaluated with data collected at monitors in the San Joaquin Valley Air Basin (SJVAB) in 2005, and it shows similar performance to that at SoCAB. The paper also illustrates the application of the model to 1) screening regions for attainment of statistically based air quality standards, such as that for the daily maximum 8-h average O_3, and 2) improving methods to interpolate observations.

Software availability

Program title: LBM-Lagrangian Background Model.
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Software: All platforms supporting Matlab.

1. Introduction

Exposure studies in urban areas require concentrations associated with emissions from a large number of sources, such as vehicles and power generators, distributed over the urban area. Computing the contributions from these sources necessitates computational resources that can become impractical even with current computers, especially when it is necessary to conduct sensitivity studies over long averaging times. The current approach to reducing these computational needs is to use separate dispersion models for different spatial scales so that sources at different distances from the area of interest are treated with different levels of source aggregation. The concentration at a receptor has three components: a regional contribution computed from a long-range transport model with grid spacing of the order of tens of kilometers, an urban "background" contribution from sources aggregated over kilometer-sized grids, and a local contribution from models that estimate concentrations at meters from a receptor. This approach to estimate the contributions from emissions at varying distances from a receptor was pioneered by Brandt et al. (2001a, 2001b, 2001c, 2001d, 2001e, 2003) in developing an integrated operational air pollution forecast system called *THOR*.

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Urban background concentrations of species such as NO_2 and ozone can be computed using comprehensive models such as *CMAQ* (*Community Multi-scale Air Quality Model*; Byun and Schere, 2006). Although the previous studies show that comprehensive models can provide reasonable estimates of background concentrations of species such as NO_2 and ozone, they can become computationally cumbersome if concentrations are required for long averaging times.

Most exposure studies avoid the use of dispersion models altogether by analyzing data from monitoring networks through land-use regression (*LUR*) statistical models (Brauer et al., 2003; Ross et al., 2007) that interpolate between observations to identify small-scale gradients in concentrations. While such models are useful for estimating current or past exposure, they cannot be used to estimate the impact of changes in emissions on concentrations and their performance is highly dependent on the measured data that is used to calibrate them (Jensen et al., 2009). Furthermore, they cannot be used to estimate concentrations at short averaging times because they do not account for the impact of varying meteorological conditions (Gulliver and Briggs, 2011).

For the reasons given in the previous paragraphs, we need a dispersion model that connects concentrations explicitly to emissions and meteorology, and at the same time is computationally convenient for long-term exposure studies. The outputs from such a model can be used to interpolate observations or serve as background concentrations for a model applied at a smaller scale of tens of meters. This paper describes such a model, which is designed to estimate hourly concentrations of selected pollutants. These concentrations can be averaged over longer periods, relevant to health studies.

The simple Urban Background Model (*UBM*, Berkowicz, 2000) is typical of the response to the need for urban scale dispersion models with small computational demands. *UBM* achieves computational efficiency through two simplifications: a straight-line steady dispersion model, and ozone chemistry based on photo-stationarity, which neglects the role of hydrocarbons. In this paper, we focus on a model that is intermediate between comprehensive photochemical models and the simple *UBM*. This model estimates urban "background" concentrations of NO_x, NO_2, and O_3, averaged over a scale of a few kilometers to tens of kilometers. The lower limit on the grid size is determined by the assumption that the concentration is well mixed through the depth of the mixed layer, and the upper limit depends on the validity of using surface winds to represent transport in the atmosphere. The model treats unsteady meteorological conditions with trajectories that reflect space and time varying surface winds, and it reduces the computational requirements of photochemical models by separating transport and chemistry using a method described in Venkatram et al. (1998). The model is evaluated by comparing model estimates of relevant species with data from measurements made in the South Coast (SoCAB) and the San Joaquin Valley (SJVAB) air basins in California, USA.

2. The Lagrangian Background Model

The model described here is similar to the Lagrangian model used in Europe to estimate long-range transport of sulfur (Eliassen and Saltbones, 1983). It estimates the concentration of a pollutant by tracing the history of the air parcel associated with the concentration at the receptor of concern at a specified time. Back trajectories are calculated in 1-h time steps using the hourly averaged wind speed (at 10 m above the ground level), and wind direction from the meteorological station closest to the receptor. Each trajectory is extended backwards in time for 24 h, which assumes that sources beyond this travel time make a negligible contribution to concentrations at the receptor. This assumption was evaluated using sensitivity studies. To facilitate the use of the model, meteorological inputs are taken directly from the surface input files used by AERMOD (Cimorelli et al., 2005) which are generated by the AERMOD meteorological preprocessor (AERMET). In a single layer model, the choice of the height of the wind used to compute trajectories is arbitrary; the choice of the 10 m wind is justified a posteriori through comparison of model estimates with observations.

The air parcel has horizontal dimensions of the grid square used to represent emissions of NO_x and *VOC* (volatile organic compounds) over the domain. The height of the air parcel corresponds to the local mixed layer height. In order to account for horizontal dispersion, we examined the approach used in *UBM* (Berkowicz, 2000), in which the concentration at a receptor is taken to be the average of the concentrations corresponding to slightly different wind directions ($\Delta\phi = 3^\circ - 5^\circ$) centered on the average wind direction. We found that perturbing the back trajectories using this approach made little difference to the results. Consequently, this approach was dropped to improve computational efficiency.

Emissions are injected every hour into the box at the grids traced by the back trajectory, and then mixed through the volume of the box. The concentrations are stepped from the $(i-1)^{\text{th}}$ to the i^{th} time step through

$$C_i = C_{i-1}\min\left(\frac{z_{i-1}}{z_i}, 1\right) + \frac{\Delta m_i}{z_i} \quad (1)$$

where C_i is the concentration of the species at time i, Δm_i is the mass of pollutant injected into the air parcel, and z_i is the mixed layer height. The term within the parenthesis on the right hand side of the equation ensures that the concentration does not increase when the mixed layer decreases during a time step. The loss of mass when the mixed layer height decreases ensures that the near surface concentration is affected primarily by material that is less than 24 h old. The mixed layer heights used in this model are generated through AERMET, which calculates the height of the convective boundary layer (CBL) through a simple one-dimensional energy balance model (Carson, 1973). The height of the stable boundary layer (SBL) is based on the formulation described in Venkatram (1980).

Currently the model does not account for dry deposition, although this process can be readily incorporated into Equation (1). The mass of pollutant injected per unit surface area of the air parcel is $\Delta m_i = q_i(\vec{r},\tau)\Delta t$, where $q_i(\vec{r},\tau)$ is the emission density at the location of the parcel, $\vec{r}$, injected at a time from the initiation of the trajectory, τ, and Δt is the time step of the trajectory calculation.

The incremental concentration during the last hour of the air parcel's path is computed with a steady state dispersion model (Venkatram and Cimorelli, 2007) that accounts for incomplete vertical mixing,

$$\Delta C_i = \sqrt{\frac{2}{\pi}}\frac{q}{\sigma_w}\ln\left(1 + \frac{\sigma_w \Delta t}{h}\right) \quad (2)$$

where q is the emission rate per unit area, σ_w is the standard deviation of the vertical velocity fluctuations, and h is the initial vertical spread of surface emissions which is taken to be 1 m. The equation is modified if the pollutant is well mixed through the boundary layer during the last time step before the parcel reaches the receptor.

Once the concentrations of the primary pollutants are estimated, the model calculates the effective age of each species in the