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A backward-time stochastic Lagrangian air quality model

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ABSTRACT

We describe a backward-time Lagrangian air quality model based on time-reversed, stochastic particle trajectories. The model simulates the transport of air parcels backward in time using ensembles of fictitious particles with stochastic motions generated from the Stochastic Time-Inverted Lagrangian Transport model (STILT). Due to the fact that STILT was originally developed out of the HYSPLIT lineage, the model leverages previous work (Stein et al., 2000) that implemented within HYSPLIT a chemical scheme (CB4). Chemical transformations according to the CB4 scheme are calculated along trajectories identified by the backward-time simulations. This approach opens up several key advantages: 1) exclusive focus upon air parcels that affect the receptor's air quality; 2) the separation of transport processes—elucidated by backward-time trajectories—from chemical reactions that enables implications of multiple emission scenarios to be probed; 3) the potential to incorporate detailed sub-gridscale mixing and transport phenomena that are not tied to Eulerian gridcells.

The model was used to simulate concentrations of air quality-relevant species (O_3 and NO_x) at eight measurement sites in the Canadian province of Ontario. The model-predicted concentrations were compared with observations, and comparisons show that simulated O_3 concentrations usually agree well with observations across sites in rural areas, small towns, and big urban regions. Furthermore, the backward-time model showed improved performance over the previous approach involving forward-time particle trajectories, especially for O_3. However, the model under-estimated NO_x at sites away from the big cities, possibly due to the inability of the coarsely gridded emission grids to resolve fine-scale NO_x sources.

Influences of cross-border transport of U.S. emission sources on the test sites were investigated using the model by turning off anthropogenic and natural U.S. emission sources. The model results suggest that total U.S. emissions contributed more than 30% of O_3 concentrations at the target sites and that over half of all hours during the simulation period were affected either by anthropogenic or natural emissions from the U.S. sources, indicating the importance of U.S. sources for air quality across Ontario.

1. Introduction

Anthropogenic emissions of chemically active species are altering the composition of the atmosphere and will become increasingly important over the next decades (Brasseur et al., 1999; Crutzen and Ramanathan, 2000). Biomass burning from land use change has been accelerating (Setzer et al., 1994), releasing large quantities of NO_x ($NO + NO_2$) and volatile organic compounds (VOC) to the atmosphere (Chatfield and Delany, 1990). In the developing world, regional scale air pollution will accelerate from urbanization and industrialization, leading to human health problems and crop damage (Chameides et al., 1994, 1999). After being emitted, such pollutants are mixed in the atmosphere and transported across borders (Brankov et al., 2003), resulting in regional scale pollution that can be examined quantitatively only with models that account for chemistry and transport at the appropriate scales. In developed nations, the adverse effects of air pollution on human health continue to be observed—e.g., increased respiratory hospitalization in Windsor, Ontario (Luginaah et al., 2005; Malig and Ostro, 2009). Such human health concerns have led to increasingly stringent controls on air quality by the U.S. EPA (1997) and the Canadian Council of Ministers of the Environment (2000)

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that necessitate careful balances between health benefits and mitigation costs (Pandey and Nathwani, 2003; Russell, 1988). Societal concerns regarding the anthropogenic impact on atmospheric chemistry and air quality call for improvements to modeling and analysis of regional scale atmospheric chemistry (Russell and Dennis, 2000).

A variety of numerical air quality models have been developed since the 1970s. Such models are broadly classified into two types according to whether they adopt Lagrangian or Eulerian coordinate systems. Eulerian models calculate the pollutant's fate and transport everywhere in the modeling domain using a fixed coordinate system. Lagrangian models calculate the trajectories of air parcels and follow them as they move through the model domain (Lin et al., 2011).

Eulerian models are powerful tools for elucidating the chemical and physical mechanisms in the atmosphere. Current generation atmospheric chemistry models generally adopt an Eulerian approach. However, Eulerian models calculate chemical reactions usually based on pollutant concentrations diluted over entire gridcells. The artificial dilution likely results in under-prediction of concentrations (Gillani and Pleim, 1996). Numerical diffusion introduced by space discretization in Eulerian models also imposes artificial mixing of pollutants (Jacobson, 1998; Odman, 1997). Advances in regional chemical modeling require further improvements in incorporating atmospheric transport processes other than mixing. Processes such as turbulent fluctuations in tracer concentrations (Fitzjarrald and Lenschow, 1983; Georgopoulos and Seinfeld, 1986), boundary-layer top entrainment (Davis et al., 1997), and convective transport (Thompson et al., 1994) remain difficult to represent in the sub-grid scale eddy diffusion coefficient approach adopted by most Eulerian models. Moreover, the grid-averaged concentrations prognosed by gridded models are difficult to compare with point observations.

Lagrangian models have the key advantage of being subject to minimal numerical diffusion (Seibert, 2004). Backward-time Lagrangian approaches are also computationally cheap, because Lagrangian air parcel trajectories running backward from the receptor site isolate the upwind influences on the receptor. Various Lagrangian approaches have been adopted for photochemical modeling in an attempt to complement and address limitations in Eulerian methods. The simplest of these Lagrangian models simulates pollutants within boxes that are advected along mean wind trajectories (Eliassen et al., 1982; Simpson, 1993). However, the idealized box representation cannot readily incorporate detailed transport processes. Alternatively, puff models such as CALPUFF (Scire et al., 2000) represent pollutant emissions with Gaussian puffs that attempt to simulate dispersion effects. However, puff models have difficulties capturing the interaction between turbulence and wind shear which distort plumes into non-Gaussian shapes, potentially introducing large biases in the peak concentrations and the plume area, thereby requiring ad hoc parameterizations such as puff splitting (Walcek, 2002; Draxler and Taylor, 1982).

Out of all of the Lagrangian approaches, stochastic particle models are the most sophisticated (Stohl, 1998). These models have the capability to simulate complicated transport effects—e.g., wind shear, convective redistribution, and turbulent dispersion. Of particular importance for stochastic particle models are simulations of transport within the planetary boundary layer (PBL), in the lower troposphere, where strong turbulence renders single deterministic mean wind trajectories highly erroneous (Stohl and Wotawa, 1995). Since ground-based air quality monitoring sites are necessarily located within the PBL, a strong need exists for the Lagrangian particles to be stochastic in nature and run backward in time, to take advantage of the aforementioned computational savings.

Stein et al. (2000) developed a stochastic Lagrangian model that runs forward in time. Recently, Miller et al. (2008) and Wen et al. (2011) described the use of backward-time stochastic trajectories to simulate air quality-relevant species. However, chemistry was neglected or highly-simplified in those studies.

In this paper, we developed a comprehensive Lagrangian air quality model based upon the backward-time stochastic Lagrangian approach. The new model is capable of simulating a wide variety of gas phase species that affect air quality using the Carbon Bond IV (CB4) mechanism (Gery et al., 1989). Lin et al. (2003) have demonstrated that given proper formulation of turbulent transport and mass conserving meteorological fields, backward-time results are identical to their forward-time analogs. In other words, backward-time simulations retrieve the trajectories of all air parcels arriving at the receptor in the forward-time case. This means that the backward-time air parcels—and only these parcels—contribute to variations in chemical tracers at the receptor, and these parcels isolate the region of the model domain needed to be accounted for in the chemical simulations. The chemical calculations can then be conducted forward in time from the regions marked out by these particles and along their trajectories, taking into account surface emissions, chemical transformations, and mixing processes.

This approach opens up several key advantages: 1) exclusive focus upon air parcels that affect the receptor's air quality; 2) the separation of transport processes elucidated by backward-time trajectories from chemical reactions, enabling implications of multiple emission scenarios to be probed ("reusing" transport information to achieve computational efficiency); 3) the potential to incorporate detailed sub-gridscale mixing and transport phenomena that are not tied to Eulerian gridcells.

It bears mentioning that 3) above is only a potential that may be realized in the future with the approach presented in this paper. Currently the particles' concentrations are mixed and averaged over fixed Eulerian grids to simulate chemical transformations, following a "hybrid Lagrangian-Eulerian" approach that has been introduced by others already (Stein et al., 2000; Stevenson et al., 1998).

The model is designed to simulate air quality over scales of 10–1000 km, serving as a crucial bridge at the regional scale, between coarse-scale global models and the fine-scale large-eddy simulations or urban air-shed models. As a test and initial application of the model, it was applied to simulate air concentrations of tracers at eight measurement sites in Ontario, Canada (Sect. 4.2). A comparison with the forward-time approach was also carried out. As an application of the model, the impact of cross-border transport of U.S. emission on Ontario was investigated (Sect. 4.3). This is an important policy and health question for Canada, as a significant fraction of the Canadian population resides near the U.S. border, downwind of numerous cities, power plants, and other large pollution sources (CEC, 2004).

2. Model description

2.1. Overview

The backward-time stochastic Lagrangian air quality model was developed from the Stochastic Time-Inverted Lagrangian Transport Model (STILT; see http://www.stilt-model.org) (Lin et al., 2003). STILT was built from the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Hess, 1997). STILT was originally developed for atmospheric transport simulations of inert tracers, especially greenhouse gases (Gourdji et al., 2010; Zhao et al., 2009; Kort et al., 2008). Recently, efforts have been made to simulate air quality-relevant species using the STILT model (Miller et al., 2008; Wen et al., 2011). However, chemistry was neglected or highly simplified in those studies. This paper represents further development to account for chemical transformations of a wide