An Approach to an Unified Process-Based Regional Emission Flux Modeling Platform

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ABSTRACT

The trend towards episodic modeling of environmentally-dependent emissions is increasing, with models available or under development for dust, ammonia, biogenic volatile organic compounds, soil nitrous oxide, pesticides, sea salt and chloride, mercury, and wild fire emissions. These emissions are estimated as hourly values using numerical modeling from physical principles, resulting in more realistic values than the historical approach of using national annual air quality inventories with temporal and spatial disaggregation factors. The basis of many of these new modeling tools is a surface flux model, either one-way or bi-directional, underpinned by similar surface boundary physics, with modifications or parameters to treat the flux of a specific emission compound or class of emissions. These developments will result in closely-related emission modeling tools with overlapping input data requirements. The emission flux models will need to be installed in or coupled to an emission modeling system, such as the Sparse Matrix Operator Kernel Emission (SMOKE) system. To maintain a unified one-atmosphere approach to air quality modeling, and to ensure a consistent scientific basis and computational efficiency, a unified emission flux modeling approach capable of estimating all or most of the environmentally-dependent emissions is recommended.. This can be accomplished by establishing a model platform containing representations of the basic chemical and physical mechanisms for mass fluxes of gaseous and particulate emissions. The modeled emissions will be merged by SMOKE with reported emission data from an inventory and supplied to the Community Multiscale Air Quality (CMAQ) model, a regional Eulerian grid model. In some instances, modeling of bi-directional fluxes will be necessary, which may require a closer coupling with CMAQ to accommodate reinitialization of the concentration field at each time step.

INTRODUCTION

There is a growing recognition that a substantial amount of emissions to the air are influenced by the environment, principally meteorological and land cover conditions. Because of the need to characterize these emissions for air quality modeling at increasingly fine spatial and temporal scales, there are ongoing efforts to model the emissions on an hourly basis as input to regional air quality models.[1] The purpose of this paper is to explore an approach to unifying the many possible hourly emission models. Existing emission processors estimate values from a combination of emission inventories and emission process models. Emission rate factors are applied to activity data for reported inventories; and other factors are modified by meteorological processes characterized by temperature, humidity, and radiation activity and applied to activity data or land cover information. The modeled emission values are provided to an Eulerian air quality model where they are inputs to transport and atmospheric chemistry computations. Modeling of biogenic emissions is an example of a current operational process model example. There are plans for process modeling of other environmentally-influenced emissions, including blowing dust,[2] emissions from wild fires and prescribed burning, and ammonia emissions.[3] In order to use modeled emission data in a regional air quality model, it is

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necessary to prepare the data using the same meteorology and same Eulerian grid description as used for the air quality model; and to merge the modeled data with similarly gridded data for other emission sources. The processing and merging are typically accomplished using an emission modeling system, such as the Sparse Matrix Operator Kernel Emission (SMOKE) model addressed in this paper.[4] The core functions of current emission modeling systems are designed to import and manipulate existing annual emission inventory data and process them so that they are in a form acceptable to the air quality model. SMOKE imports county specific-based gridded data (gridded by an external tool) from annual emission inventories, applies factors from temporal and chemical speciation profiles contained in the system, growth and/or control factors from a source category-specific file, and performs a range of quality control checks on the data. Biogenic emissions are modeled by algorithms which have been coupled to SMOKE for efficiency of processing and to ensure consistency in the grid definitions and chemical speciation profiles used.[5] In addition, SMOKE imports and applies multiplicative factors representing gridded land cover for these models. As hourly models capable of representing episodic events for other kinds of environmentally-influenced emissions are developed, they are also likely to be coupled to a central emission modeling system, also for reasons of consistency and operating efficiency. Embedding several subsidiary emission models in a central emission modeling system is likely to result in some loss of operating efficiency and an increasingly repetitive and cumbersome system, particularly for processes which use some of the same input data at ever finer scales of spatial and temporal resolution.

Advanced emission models are also likely to require modeling of emission processes at sub-grid spatial scales in order to determine the portion of emissions available to the air quality modeling gridded domain, rather than simply by using factors to make an aggregate amount of emissions available to the air quality model. Emissions of blowing dust, which are dependent on local soil and vegetation characteristics, are an example. Multiple emission process models will likely duplicate some basic sub-grid computations, such as soil moisture flux, which could introduce discrepancies in common computed parameter values and specific chemical and physical properties of the emissions modeled in each case. Historically, regional modeling of the fluxes of many substances, particularly volatile and semi-volatile compounds, has assumed a one-way upward flux rather than bi-directional behavior, with deposition accounted for after chemical reaction and transport by the air quality model. This treatment is unrealistic for substances like ammonia, for which direction frequently varies according to a temperature compensation point affected by vegetation. New emission models including bi-directional flux processes (for example ammonia, sea salt, and chlorine) are in the beginning stages of development and may require initial and step-wise (hourly) determination of the concentration field of the emitted substance, as well as the rate and direction of flux to or from the atmosphere.[6] Their use will require reinitialization of modeled concentration fields from the air quality model at each time step. To date, bi-directional emission fluxes, complicated by local soil properties, vegetation cover and phenology, have been directly addressed mainly at a detailed field scale[7]. Reintialization at each modeling time step might most efficiently be accomplished dynamically within CMAQ (after initial spatial gridding and initial computation in the emission model). The need for dynamic reinitialization implies a more closely coupled operation of SMOKE and CMAQ than has previously existed.

EMISSION MODEL STRUCTURE

The prospect of multiple advanced emission process models operating simultaneously suggests that they might be standardized and/or combined. Combining common aspects of the emission process models will result in emission modeling more consistent with the goal of unified “one-atmosphere” air quality modeling. As more is learned about emission processes, additional specific physics and chemistry can be added to the computational procedures. Figure 1 illustrates the conceptual unified structure and information flow for modeling of process-based environmental emissions.

The input data will go to a Emission Process Flux Module (Figure 2), which will contain the general and specific computations required. Computations in the Module will be for all grid cells, using matrices to the extent possible, to avoid imposing non-multiplicative computations within the sparse matrix structure of SMOKE, which would interfere with SMOKE's design efficiency. The Module will contain general algorithms applicable for computation of volatilization of gaseous emissions and wind suspension of particulate emissions. Interaction with the soil surface and vegetation canopy will be sub-models within the system to produce at least an initial emission field. It is possible that operating efficiency will require that versions of the soil and vegetation canopy submodels be containing in CMAQ to allow dynamic reinitialization of concentration fields during modeling. Using meteorological data adjusted for scale, these algorithms will be applied to each grid cell, for different emitted substances, such as dust and ammonia emissions. A hypothetical common sub-grid computational scale is 100 meters. This scale approximates that of many field experiments which are the basis for detailed emission flux information. The results may need to be adjusted for more specific local conditions. Next, the more pollutant-specific chemical-physical parameters and gridded input data will be applied to obtain the range of gridded modeled emission fluxes (Figure 3).

After computation of the gridded emission fluxes for the different substances, the data will be passed to the main emission model system (SMOKE) for speciation according to the lumped species chemistry schema being used in CMAQ,[8,9] and merging with gridded, speciated emission data from an inventory. The resulting combined gridded emission data will be provided to CMAQ. The requirement for reinitialization of emission concentration fields at selected time steps implies that CMAQ chemistry must be capable of modeling an output concentration field of the modeled emission compound(s) (as opposed to standard lumped species compounds) at each time step, as well as making use of the compound(s) in its internal chemical computations. This implies expanded chemistry and a more closely coupled operation of SMOKE and CMAQ than is the current practice.

EXAMPLES

Hourly biogenic emissions are modeled in the SMOKE system using the Biogenic Emission Inventory System (BEIS Version 3.12) and a detailed 1km resolution land cover data base for North America (BELD3).[10] BEIS3 uses plant-species and chemical compound-specific emission factors in conjunction with the gridded meteorological data from the Mesoscale Meteorological Model (MM5) (also used by CMAQ) to obtain gridded emission values.

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Ammonia

The modeling of the emission flux of ammonia is a developmental area which demonstrates the proposed architecture of the emission process flux model.[11] In concept, the model will require gridded meteorology data, detailed vegetation data (vegetation type and morphology), soil type data, and the locations and source strength (amount of ammonia emitted per unit time) from human activities - principally agricultural husbandry operations. Because ammonia is a bi-directional flux, varying over the range of ambient temperatures and vegetation conditions, the existing ambient ammonia concentration is important to the subsequent flux. Consequently reinitialization at each time-step by means of a concentration field output from CMAQ is important. Computation of an initial concentration field using gridded input data would be accomplished in the unified emission model. However, computational efficiency may require that the hourly concentration fields be computed dynamically within CMAQ, using versions of the soil surface and vegetation canopy sub-models within CMAQ.

Dust

Emissions of dust are difficult to estimate as a part of annual emission inventories using traditional emission factor approach. This approach does not directly account for the processes suspending dust. Consequently all dust potentially emitted (suspended) was available for transport by the air quality model. Recent work has demonstrated that localized spatial and temporal variability and interaction of dust sources, soils, vegetation, and meteorological conditions make generalized estimates at the county level problematic. Efforts are underway to develop an episodic dust model which can be applied to model suspension and local transport of blowing dust from the land surface, as well as suspension and local transport of dust from unpaved roads, construction activity, and agricultural tilling.[2] Episodic dust emission models require data at a high spatial resolution in support of efforts to connect suspension and/or transport at the local sub-grid or field scale of measurements to the grid resolution scale of CMAQ. Meteorological data is needed to model the local suspension and transport of dust particles prior to the transport modeling in the context of the gird scale of CMAQ. A tighter coupling between the flux of modeled dust and dust transported by CMAQ might be attained by use of stepwise reinitialization of the dust concentration field within CMAQ.

Fires

Estimates of emissions from fires, both wild and prescribed, have been problematic primarily because of uncertainty in the size and location of sources, and their temporal and spatial variability. Annual and multi-year averages of fire emissions have climatological value, but are of little use in modeling episodic emissions for a specific year and time. Current work by the U.S. Environmental Protection Agency in conjunction with the U.S. Forest Service will substantially improve the ability to episodically model fire emissions in response to improved fuel loading data, fire location information, and fire behavior modeling (including plume behavior), reflecting meteorology.[12] Except for the largest fires, the fire emission source areas are sub-grid phenomena at the grid resolution of CMAQ runs, with some emissions remaining within a grid and some being transported out, depending on the size and location of the fire. The behavior of fire plumes is one of the current research development areas. It is necessary to bridge between the scale at which the fires occur and the grid resolution scale, possibly using a combination of reported fire data and remote sensing imagery.

Sea Salt and Chlorides

Marine aerosol emissions are an important part of the modeling of particulate matter. In particular sea salts and interaction with chlorine are crucial.[13] The emissions are influenced by temperature, winds (affecting wave action), water salinity and the existing concentration of sea salts in surface air boundary layer. The effect of emissions of sea salt in coastal and estuarine waters on atmospheric chemistry have been explored by parameterizations built into an unreleased research version CMAQ. However, spatial and temporal emissions of the concentrations field are generalized. The modeling of sea salt could be improved by developing a model within the context of the unified emission process model, which will simulate the initial boundary layer sea salt concentration at the grid scale specified for a particular application using surface salinity data and gridded meteorological data from MM5. The modeled grid concentration fields could be reinitialized dynamically within CMAQ, provided when CMAQ contains the appropriate chemistry..

Pesticides

The chemical and physical composition of pesticides is highly variable, ranging from the highly volatile to the nearly inert. However, many common pesticides used for lawns, gardens, and large scale agriculture (including herbicides, insecticides, and fungicides), have some degree of volatility. Because pesticides are not classified as air pollutants, they have not been included in annual inventories, except to the extent that they are contained within Volatile Organic Compound (VOC) emissions. Some pesticides, or constituents thereof, are classified as toxic substances and are included in inventories of toxic emissions. Others, such as DDT or Toxaphene are banned, and are re-emitted only as fugitive substances. The need to perform regional episodic modeling of air quality emissions may increase with more attention to toxic emissions. The temporal and spatial variability of use of pesticides makes annual regional estimates of emissions difficult, and of questionable value for episodic air quality modeling. In most cases, pesticide emissions are bi-directional, and depend on soil characteristics, vegetation cover, time and mode of pesticide application, meteorology, and the existing pesticide concentration in the boundary layer. Recently, models such as the Pesticide Emission Model have been developed and tested for regional application.[14,15] Again, reinitialization of the pesticide model, using concentrations output by an air quality model such as CMAQ, are needed to establish a better coupling between the emission model and the air quality model.

DATA INPUTS

Data inputs needed by process-based emission models include meteorological variables, surface attributes (land cover, soils, vegetation, etc.), activity data (e.g., agricultural practices such as tilling, crops, fertilizer use; construction activities), and substance and process-specific characteristics. Meteorological variables in gridded form are provided from the Mesoscale Meteorological Model (MM5). For algorithms in an advanced emission model, the meteorological data (usually hourly) may need to be converted to different time steps or units for use by the components of the emission flux model(s). One reason for the establishment of emission modeling systems was to use gridded spatial data in the computation of emission values. These data are gridded to the same spatial resolution and domain defined by the user for the use of the air quality model. The data may be used directly in the computations of emission process flux models, or as surrogates to spatially allocate emission inventory information. As previously mentioned, meteorological data usually come in a consistently gridded form from the same meteorological model used to drive the air quality model. Different emission processes draw on a variety of spatial data sources in addition to meteorological data. The data are usually gridded by a commercial geographic information system (GIS) either external to or embedded in the emission modeling system. A unified emission process module will require access to expandable libraries of geographic information and a gridding tool. It is not necessary that all the spatial data be located on the computer system on which the model is running, since spatial data could be downloaded once for repetitive application This will be accomplished by a scale adjustment converter designed to aggregate or disaggregate data temporally and spatially, and provide the needed values and formats. Some of the conversion work can be accomplished by existing public domain tools, such as the Spatial Allocator for the spatial gridding of information.[16] Spatial scale adjustments by aggregation may be required if modeled emission processes assume a smaller spatial regime than the gridded meteorological data. Scale adjustment will provide modeled information at appropriate temporal scales which specific model components will use as necessary. The modeled emission data will then need to be accommodated to the common (typically hourly) temporal scale and the spatial grid scale being modeled, in order to allow the results to be merged with emission data derived from emission inventories, prior to being used in the air quality model. .

Process-based emission modeling will eventually encompass a wide range of substances, requiring a wide range of chemical-physical characteristics as input data. These data may be effectively organized in an input data library of substance properties. Data within the input library will be organized into chemical and physical classes of characteristics. For example chemical compounds can be organized into volatile, semi-volatile, and non-volatile classes, with sub-groups by properties. Particulate matter can be organized by chemical composition and particle size characteristics. This organization will allow consolidation of computations by class and/or assist the modeler in selection of substance with properties similar to the substance of interest.

CONCLUSIONS

The increasingly sophisticated and detailed capabilities of episodic Eulerian regional air quality models such as CMAQ require corresponding improvements in the form of more environmental-process based emission flux models for those emissions directly affected by environmental conditions. These models often have overlapping data needs and some common processes, including boundary layer momentum and energy balance components which affect suspension of particulate material, evaporation of volatile substances, and transport of the substances. With some reformulation and units conversion these processes may be used for several emission process models, and then supplied to more model-specific computations. Hourly computation of emission fluxes are data intensive, both spatially and temporally. However, computational savings may be realized by sharing common information and calculations, such as soil characteristics, land cover data, vegetation characteristics, and chemical compositions. These data may be processed and gridded consistent to a spatial domain of interest and supplied to the all of the emission flux models. The emission model results then may be supplied in a consistent and efficient manner to a central emission model, such as SMOKE, for merging with inventory-based emissions and speciation according the lumped species modeling scheme being used in CMAQ. In some cases, notably for bi-directional fluxes as for ammonia , sea salt, and pesticides, CMAQ may need to dynamically reinitialize the emission concentration field after each modeling step. Improvements in CMAQ chemistry and soil-surface and vegetation canopy sub-models will be required in these cases.

DISCLAIMER

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KEYWORDS

Emission modeling
Process-based modeling
Bi-directional flux
SMOKE
CMAQ

Figure 1. Conceptual information flow diagram of a unified process -based emission model

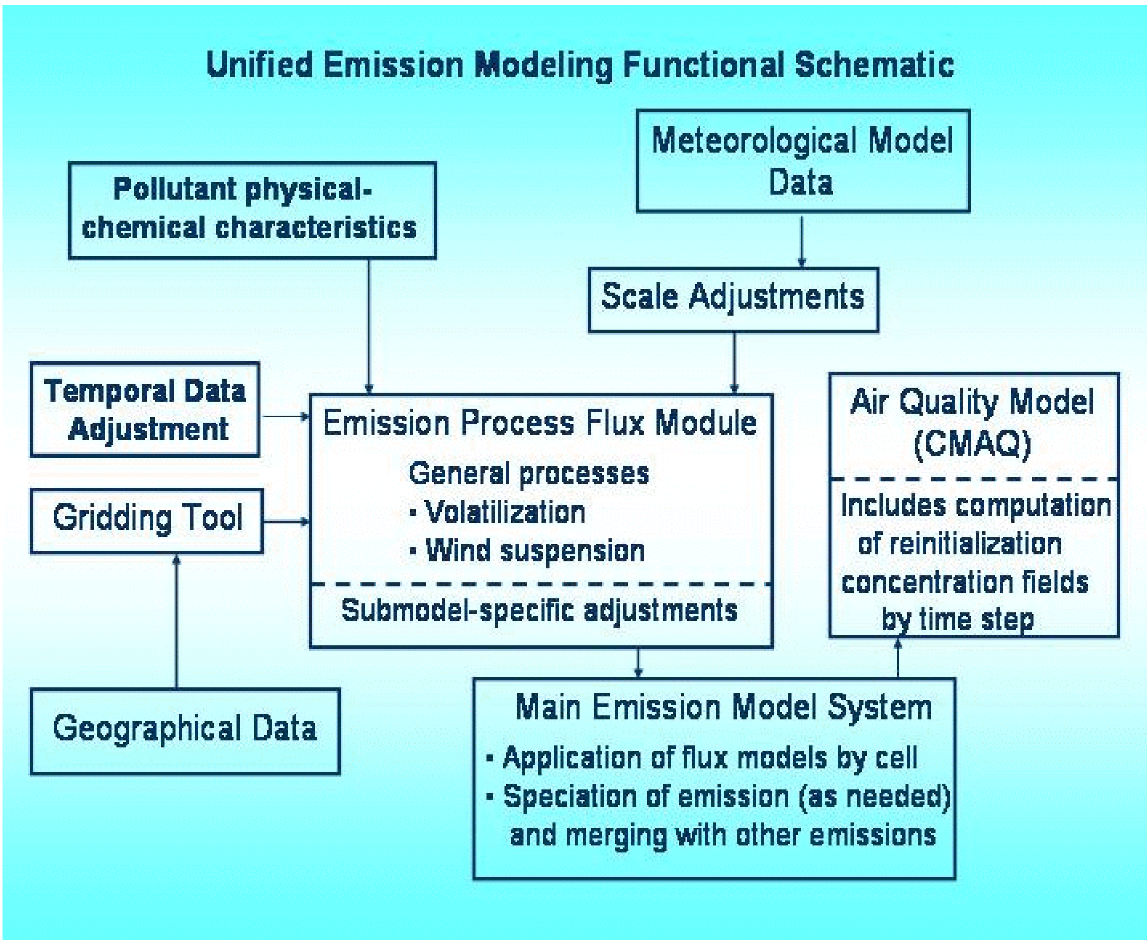

Figure 2. General processes in a unified process-based emission model

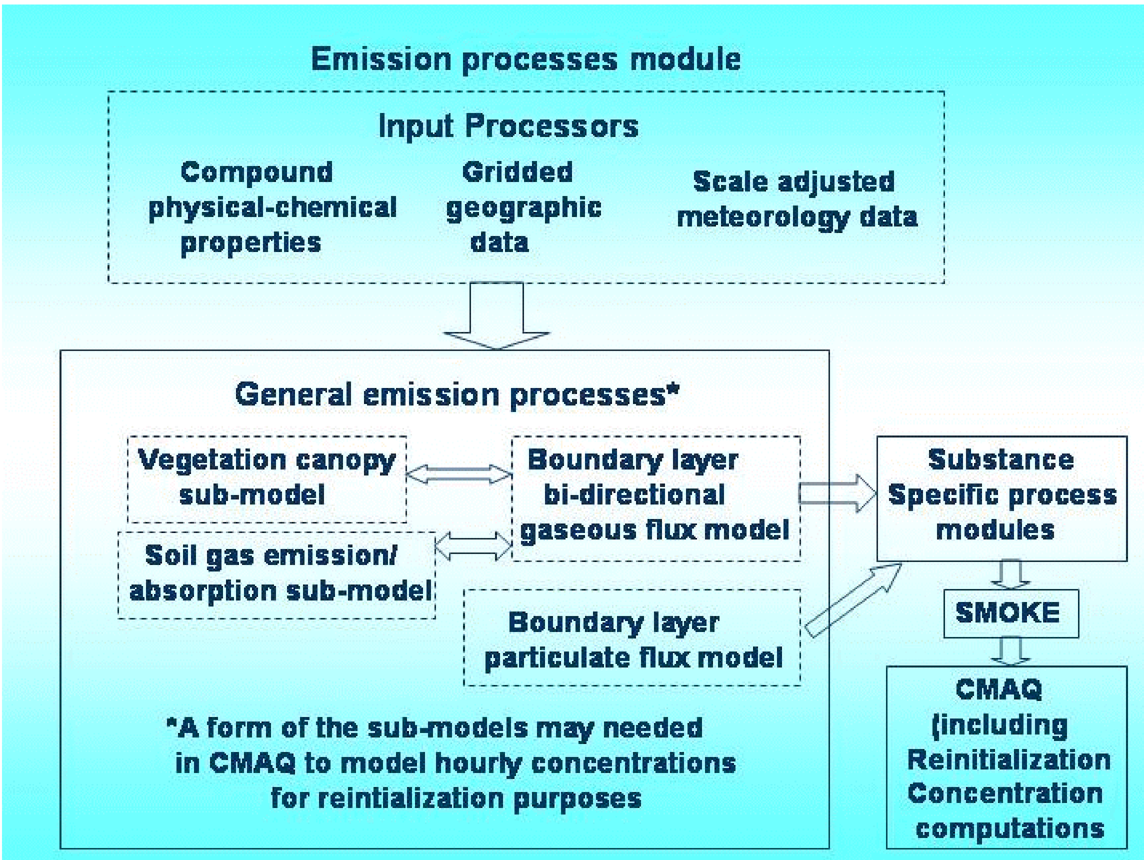

Figure 3. Examples of substance or source type specific computations in sub-modules of a unified process-based emission system

Substance Specific Process Examples
(Sub-modules by substance)

Series of algorithms (developmental) to address substance-specific processes and adjustments to accommodate the physical and chemical specifics of each substance. These may be in the emission module and/or CMAQ as computationally most efficient. Examples of substance-specific process include:

- **Emission tools and one-way fluxes**
  - **Biogenic plant species-specific modeling**
  - **Computations for fuel loading, tree species, and prior fires for wild fire emission modeling**
  - **Adjustments for application, seasonality, and crop type for emissions from pesticides and nitrogen fertilizers**
- **Bi-directional flux related computations, such as:**
  - **Ammonia compensation points**
  - **Semi-volatile phase changes**
  - **Chlorine and sea salt chemistry**