

**New Directions:** Time for a new approach to modeling surface-atmosphere exchanges in air quality models?

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## Introduction

Just as the exchange of heat, moisture and momentum between the Earth's surface and the atmosphere are critical components of meteorological and climate models, the surface-atmosphere exchange of many trace gases and aerosol particles is a vitally important process in air quality (AQ) models. Current state-of-the-art AQ models treat the emission and deposition of most gases and particles as separate model parameterizations, even though evidence has accumulated over time that the emission and deposition processes of many constituents are often two sides of the same coin, with the upward (emission) or downward (deposition) flux over a landscape depending on a range of environmental, seasonal and biological variables. In this note we argue that the time has come to integrate the treatment of these processes in AQ models to provide biological, physical and chemical consistency and improved predictions of trace gases and particles.

## Separate Processes or Integrated Whole?

The majority of current state-of-the-art AQ models are structured as shown in Figure 1, although some variation exists. Data required by an AQ model include meteorological variables, including wind, temperature, pressure, humidity and turbulent transport fields, land use data and surface or elevated emissions of gases and particles. Common practice over the last 30+ years has been to create meteorological data fields and emissions data files independently of and prior to the use of the AQ model itself. Typically, a meteorological (i.e., physics-only) model has been run for the simulation time period of interest with the relevant data fields captured and saved for ingestion and use by a subsequent AQ model simulation. In more recent years, this practice has begun to change, with AQ models being developed from meteorological models (e.g., WRF-Chem; Grell et al., 2005), or by directly linking (in either one-way or two-way modes of operation) a meteorological model with an AQ model (e.g., WRF-CMAQ, Wong et al., 2012).

Likewise, emissions data files are usually created separately from the AQ model, although typically employing outputs of a meteorological simulation to modulate emissions of some species that are influenced by environmental conditions. This too has

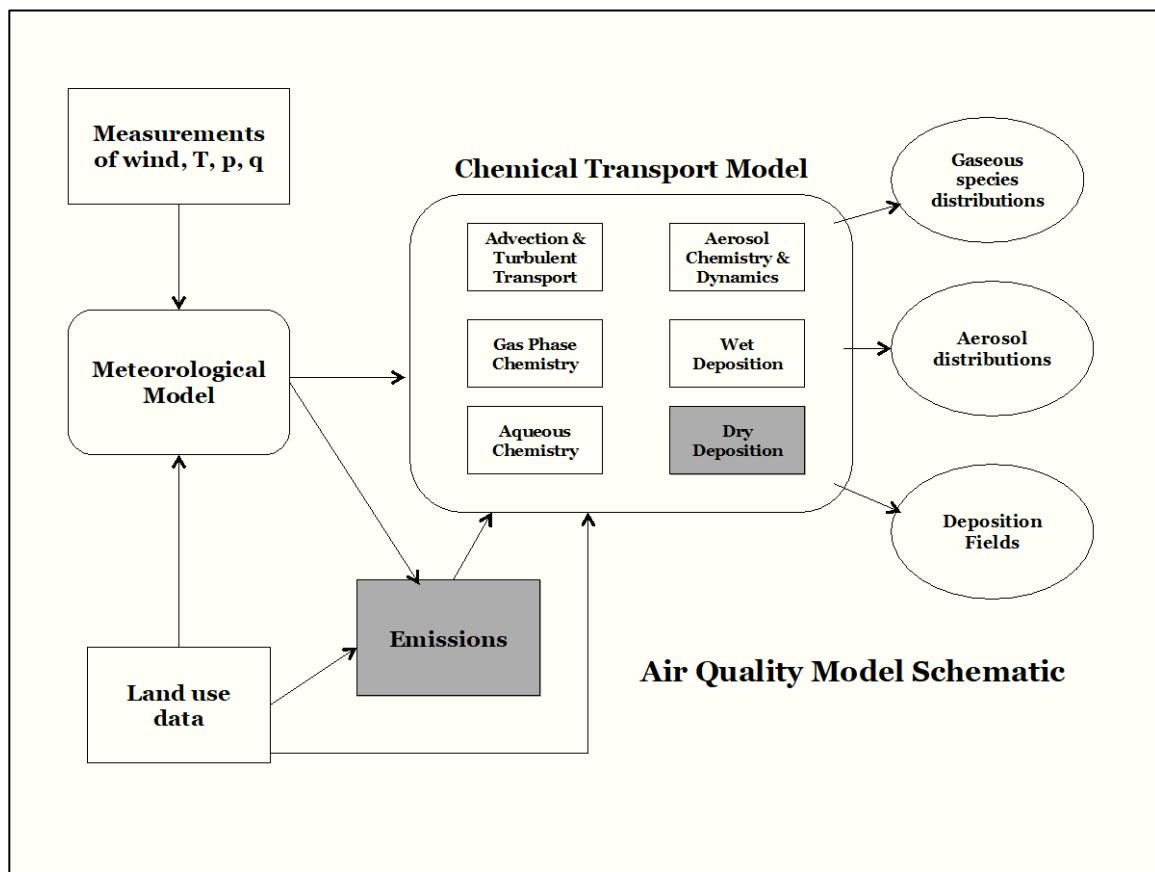
recently begun to change, in particular with emissions of biogenic species (e.g., isoprene, mono- and sesqui-terpenes and others) being calculated from highly empirical parameterizations (e.g., MEGAN, Guenther et al., 1995, 2006, 2012) online during the AQ simulation as a function of ambient temperature and insolation data. However, almost all modern AQ models treat emissions of all gases and particles (either anthropogenic or biogenic in nature) as a completely independent process from the removal of these substances through dry deposition pathways.

The exception to the separation between emissions and deposition in current AQ models is the treatment of the bi-directional nature of ammonia ( $\text{NH}_3$ ). For some time it has been recognized that the interaction and exchange of atmospheric  $\text{NH}_3$  with vegetation canopies and soils can result in either net deposition or emission (Sutton et al., 2007; Sutton et al., 2009) depending upon a complex and temporally varying set of environmental, soil and vegetative conditions. Modules have been developed to account for the bi-directional exchange of  $\text{NH}_3$ , either as stand-alone analysis tools (Nemitz et al., 2001; Bash et al., 2010) or as sub-modules of AQ models (Zhang et al., 2010; Pleim et al., 2013). Although significant challenges remain to be resolved in modeling bi-directional  $\text{NH}_3$  exchange in AQ models, improved simulation of  $\text{NH}_3$  and related species has already been demonstrated using these approaches (Bash et al., 2013; Zhu et al., 2015).

As techniques have advanced for measuring abundances and fluxes of organic compounds, evidence has accumulated that many gaseous biogenic hydrocarbon species also exhibit bi-directional exchange between vegetative canopies and the atmosphere. In a recent review article, Niinemets et al. (2014) catalogue a variety of studies, from leaf-scale laboratory experiments (Kesselmeier, 2001; Rottenberger et al., 2004, 2005; Karl et al., 2005; Kuhn et al., 2002) to canopy-scale flux measurements (Karl et al., 2010; Jardine et al., 2008, 2011) that demonstrate that bi-directional exchange of biogenic volatile organic compounds (BVOCs) and their oxidation products between vegetation and the atmosphere is “the rule rather than the exception.” Moreover, as Park et al. (2013, 2014) demonstrated in measurements above an orange grove in California’s Central Valley, the majority of measured BVOC compounds (484 out of 555) exhibited bi-directional fluxes, most with pronounced diurnal cycles, suggesting a close linkage to environmental conditions and biogeochemical processes. Likewise, Wohlfahrt et al. (2015) synthesized ecosystem-scale methanol flux measurements from eight locations with a variety of dominant plant functional types and concluded that bi-directional exchange occurs routinely at all sites included in their analysis. Based on their investigation, Wohlfahrt et al. (2015) call for a new generation of models in which methanol emission and deposition are integrated into a unified framework.

At the same time that the bi-directional nature of gaseous BVOC fluxes is becoming more accepted, it is also being recognized that above-canopy fluxes of atmospheric fine particles can exhibit either net deposition or net emission. Recurrent upward fluxes of atmospheric particles above vegetative canopies have been measured for many years (e.g., Hicks et al., 1982 and Hicks et al., 1989). More recent studies, using several flux measurement methods over a variety of surfaces, routinely find both upward and

downward particle fluxes, often with discernable diurnal variation (Nemitz et al., 2004; Pryor et al., 2008; Vong et al., 2010; Gordon et al., 2011; Lavi et al., 2013; Pryor et al., 2013; Farmer et al., 2013; Deventer et al., 2015; Rannik et al., 2015). Attempts have been made to explain the upward fluxes (Lee and Wesely, 1989; Pryor et al., 2008; Pryor et al., 2013), but no consensus has emerged that accounts for all observations and environmental conditions. Time scales for aerosol dynamics and BVOC chemistry (Pryor and Binkowski, 2004; Saylor 2013) and within- and above-canopy turbulent transport (Foken et al., 2012; Thomas et al., 2013) are similar enough that interpretation of above canopy particle fluxes is challenging. In any case, the inherent separation of emission and deposition, coupled with the coarse vertical grid resolution typical of current AQ models makes adequate simulation of these complex bio-physio-chemical systems practically impossible.



**Figure 1.** Process Schematic of a Traditional Air Quality Model

### Integrated Surface-Atmosphere Exchange

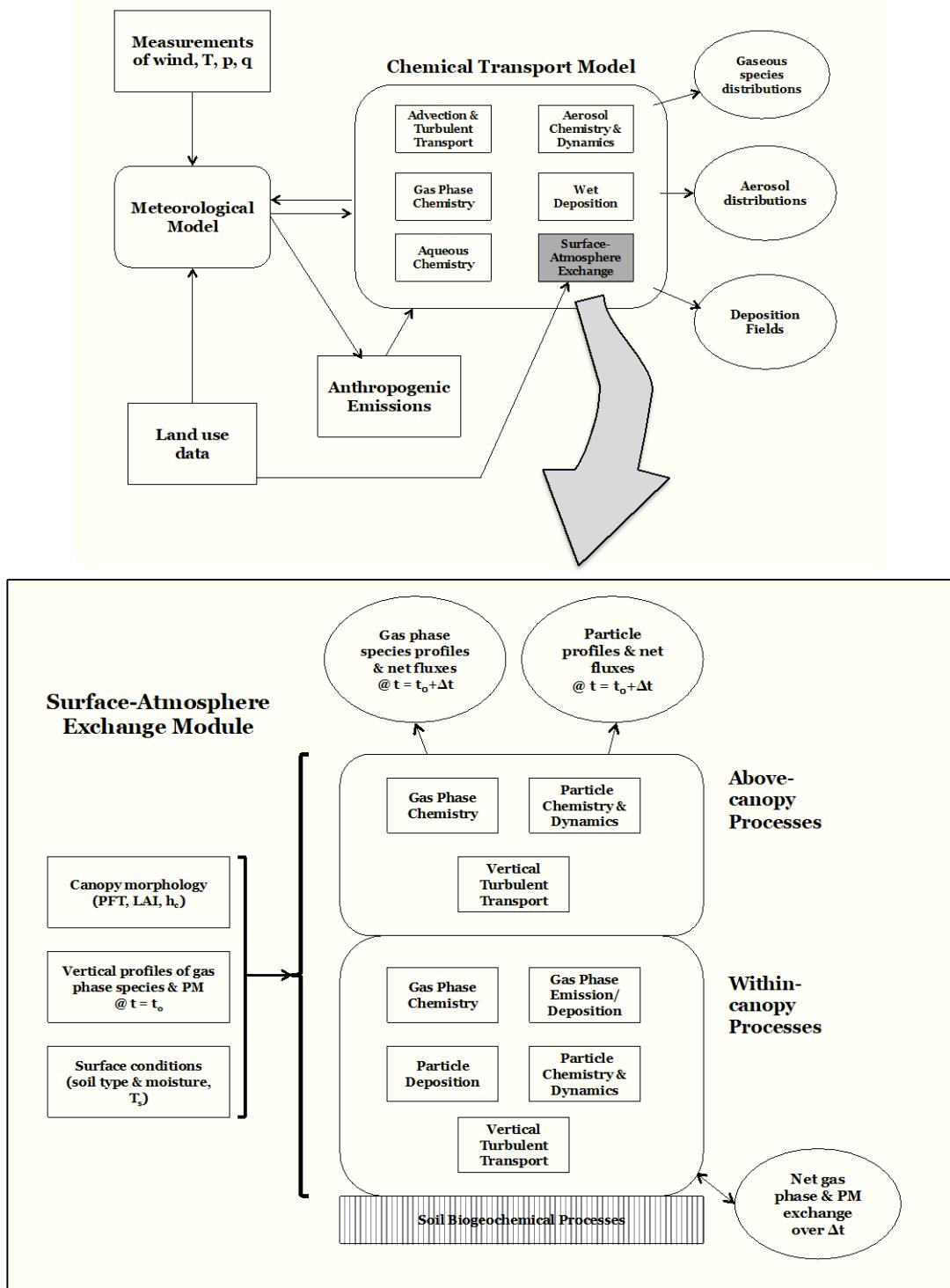
Given the emerging observational picture of the widespread bi-directional nature of surface-atmosphere exchange for many gases and aerosols, it seems clear that the

programmatic practice of separating emissions and deposition in AQ model simulations needs to change. We contend that the time has come to begin to incorporate surface-atmosphere exchange modules into AQ models that integrate emissions and deposition of all gases and particles into biologically, physically and chemically consistent systems as conceptualized in Figure 2. By accounting for interactions of biology, micrometeorology, chemistry, canopy/surface dynamics and multi-phase physics, AQ models would be in a better position to more accurately simulate the effective bi-directional fluxes of gases and particles and replicate observed surface-atmosphere exchange fluxes. Moreover, such bio-physio-chemical modules would necessarily be more responsive to changes in land use, surface conditions and meteorological variables, resulting in AQ models that are more adept at simulating impacts of changing land use patterns and climate change. Our discussion here addresses mostly the terrestrial situation, which dominates regional- or continental-scale AQ simulations. However, for global-scale simulations oceanic air-surface exchange becomes a dominant process and likewise should be treated consistently as bi-directional exchange and not as separate emission and deposition processes (e.g., O'Dowd and de Leeuw, 2007; Sinha et al., 2007). Our intent with this note is not to offer a complete solution to this model shortcoming, but only to call attention to it and thereby initiate a rigorous discussion among modelers and field measurement researchers on the path forward.

In the past, when AQ models used horizontal grid resolutions of 12 km, 20 km or even 40 km, separate emissions and deposition modules gave adequate approximate results for grid cells consisting of a patchwork of land use types and vegetative canopy structures. As horizontal grid resolutions approach smaller and smaller scales (4 km being typical currently to 1 km or less in the not so distant future), land use types and canopies become more uniform across grid cells, allowing for (maybe requiring?) a more highly surface-specific treatment of surface-atmosphere exchange. Of course, incorporating integrated surface-atmosphere exchange modules into AQ models will likely increase the overall computational burden of each simulation. However, recent supercomputer architectures are increasingly taking advantage of the superior processing ability of graphics processing unit (GPU) accelerators to provide fine grain parallelism in large-scale simulations (Brock et al., 2015; Michalakes and Vachharajani, 2008). Techniques to exploit GPUs may prove useful in ameliorating the added computational burden of integrated surface-atmosphere exchange modules in AQ simulations.

The experience with implementation of ammonia bi-directional fluxes into AQ models serves as a revealing lesson. Model developments were guided by a dedicated and revealing experimental program (q.v. Sutton et al., 1993). Although a static compensation point approach has been shown to be a good first-order modification to what otherwise would be a straightforward application of the conventional deposition velocity approach (e.g. Aneja et al. 2001), the simplicity of this methodology fails to lend itself to the BVOCs of major interest here, since even in the case of ammonia there is need to take biological factors into more detailed account (Schjoerring et al., 2000). Doing so often leads to a need for a dynamic compensation point to effectively simulate changing environmental and biological conditions. Specifying even a static

compensation point is fraught with uncertainty in an AQ model simulation over a heterogeneous grid cell; however, this uncertainty may be of lesser magnitude than completely ignoring the bi-directional nature of the exchange. Extending this approach to the case of BVOCs will require substantial effort in both new model structures as well as new field measurements. A concerted effort of modelers and field measurement scientists will thus be required to advance our understanding of these processes for a broader suite of species simulated in AQ models. Nevertheless, the effort required to design and implement sub-grid-scale integrated surface-atmosphere exchange modules seems a worthwhile investment to improve the way in which atmosphere-surface exchange is described in AQ models. As a starting point, we propose that the appropriate modeling and measurement communities convene a workshop to identify research needs and next steps to initiate this effort.



**Figure 2.** Potential Air Quality Model with Surface-Atmosphere Exchange Module

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