Can a bulk plume represent the effect of aqueous-phase reactions in shallow cumuli?

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In addition to moist convection's effects on heat, moisture, and momentum, it also produces, redistributes, and removes chemical pollutants and aerosols that are important not only to air quality but also to climate. For example, some of the climatically and chemically important reactions, such as the oxidation of sulfur dioxide, occur in the aqueous phase. Since effects of moist convection on the transport and reaction of species with aqueous-phase reactions are sub-grid scale in global climate models and chemical transport models, such effects must be parameterized in these models.

In this study, we assess how well the widely used bulk plume model can represent tracers with aqueous-phase reactions in the presence of shallow cumuli. A tracer, modeled after sulfur dioxide, is released from surface at a horizontally uniform rate. A second tracer, modeled after hydrogen peroxide, is nudged toward a prescribed vertical profile with a timescale of one day. The surface released tracer is carried upward by sub-cloud layer turbulence and by the cumulus updrafts and reacts with the second tracer in the presence of liquid water to produce the third tracer, which may be viewed as sulfuric acid. The reaction rate is the product of a prescribed reaction constant, concentrations of the first and second tracers, and the amount of liquid water.

We use Large-Eddy Simulations (LES) as a virtual reality, against which results of the bulk plume model are tested, and examine a wide range of reaction scenarios with different reaction rate constants. We derive the entrainment and detrainment rates of the bulk plume model from the LES using conserved thermodynamic variables such as total water and liquid water potential temperature. Doing so allows the bulk plume to perfectly reproduce convective tendencies of heat and moisture generated by the LES and isolates errors introduced by the aqueous reactions. We identify two error sources when using the bulk plume model, both arising from the simplifying assumption made in the bulk plume model that properties are uniform within clouds and within the environment surrounding the clouds. The first source of error is that the effective entrainment and detrainment rates appropriate for the chemical tracers can be substantially different from those for the conserved tracers. The second source of error is that chemical reaction rates of a heterogeneous cloud field, produced by the LES, differ from those of a uniform cloud field assumed in the bulk plume model. In cases with fast aqueous reactions, there are large differences between the effective entrainment and detrainment rates for the chemical tracers and those for conserved tracers such as total water. Chemical tracers are also very heterogeneous within clouds when reactions rates are fast. However, even with these large sources of errors, the fast reaction damps the error strongly as the plume rises,

resulting in moderate overall errors. In the slow reaction regime, the reacting tracers behave similarly as the conserved tracers so that the bulk plume model performs well. Error in total production of the third tracer ("sulfuric acid") integrated over the cloud layer is about 10% for reaction rate constants appropriate for sulfur dioxide and hydrogen peroxide, and exceeds 20% only for reaction rate constants that are two orders of magnitude higher. We conclude that the bulk plume model is generally adequate in reproducing the transport and reaction of tracers with aqueous-phase reactions in the presence of shallow cumulus convection.