A First-Order Closure for Covariances and Fluxes of Reactive Species in the Convective Boundary Layer

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ABSTRACT

Covariances and fluxes of reactive species in the clear convective atmospheric boundary layer (CABL) are studied and parameterized. The covariances result from correlations between reactive species. These covariances may have a considerable influence on the modeled reaction rates in atmospheric chemistry models, but usually are neglected. To facilitate the representation of covariance effects in large-scale atmospheric chemistry models, the authors have developed a new first-order closure for covariances. The closure is based on top-hat distributions, as is common in mass-flux schemes. In addition, the authors utilize an existing nonlocal first-order closure expression for the flux, which represents the combined effects of gradient mixing and nonlocal convective mixing. The authors show how the latter also includes the impact of chemistry on the nonlocal flux contribution. The impact of the closures is illustrated first for artificial, simple chemistry cases. The results are evaluated using large-eddy simulation (LES). By comparing results for the entraining and solid-lid CABL it is established that the covariance closure works satisfactorily away from the inversion. Subsequently, the closures are evaluated against LES for a photochemical case with 10 reactions involving six modeled species. The accuracy of the modeled covariances is found to be within a factor of 2, which is sufficient to improve the modeled concentrations.

1. Introduction

The turbulent mixing of fast-reacting chemical species in the clear convective atmospheric boundary layer (CABL) (here ''fast'' is taken relative to the turbulent timescale) has been known for quite some time now to require special treatment in large-scale atmospheric chemistry models (e.g., Lamb 1973). An important aspect of the turbulent transport–chemistry problem is the fact that reactive species are not always well mixed because of the short chemical timescales associated with certain important reactions—shorter than or comparable to the convective mixing timescale [see Vilà-Guerau de Arellano and Lelieveld (1998) for a precise definition of chemical timescales].

Reacting species concentrations can be (anti)correlated. The correlations are represented in the expressions for the mean chemical reaction rates by covariance terms and can have a considerable impact on these rates (e.g., Donaldson and Hilst 1972; Bilger 1978; Schumann 1989; Krol et al. 1999). Covariance effects related to convective boundary layer mixing can have impacts on large-scale species budgets. However, at present no estimate of large-scale covariance effects exists. This lack is caused mostly by the fact that there is no simple covariance parameterization available that readily can be included in large-scale models. The current paper aims to fill this gap. Future studies can use the covariance parameterization proposed here to perform large-scale assessments.

We develop a first-order closure for the covariance, which is supported with an argument based on the study of mass-flux characteristics of reactive species in the CABL by Petersen et al. (1999). The closure must be simple, that is, first order, since for more complex higher-order closures (e.g., Sykes et al. 1994; Verver et al. 1997) too many prognostic variables would have to be

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