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Impact of online/offline calculation of photolysis rates on CTM modeling accuracy in presence of heterogeneous clouds

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Sunlight drives the chemistry of the atmosphere by dissociating some molecules into highly reactive fragments: it's the photolysis process. Photolysis is so one of the most important process in the atmospheric chemistry, especially in polluted areas. The atmospheric oxidation capacity is mainly controlled by the photolysis of major species such as NOx or ozone which produce most of the tropospheric peroxides and radicals. However, calculation of these rates is very expensive in CPU time. Due to this problem, lots of CTM's use offline tabulated actinic fluxes instead of resolving them at each time step and each grid cell. But photolysis rates in the troposphere are greatly affected by the presence of clouds and aerosol layers, surface albedo, temperatures, pressure… and the temporal and spatial variability of these parameters can't be taken into account by offline model, such as TUV (Tropospheric Ultraviolet and Visible radiation model; Madronich et al, 1989). With the evolution of computer capacity, online calculation of photolysis rates can be considered. Some works have been done in this way, with the FAST-J model (Wild et al, 2000). Are these improvements really necessary under cloudy (heterogeneous or homogeneous cover) and clear sky situations in the case of urban pollution peaks?

Another problem to focus on is that both TUV and FAST-J models use the homogeneous and plane-parallel clouds (or aerosol layers) hypothesis. But clouds are not plane-parallel, presenting bumps and lacks. As shown by satellite measurements, cloud and dust optical properties (optical depth, effective radius…) are not homogeneous in space and time. Thus, we can wonder how large the uncertainties introduced by this approximation are.

This study is divided into two parts:

- A comparison of 2 methods for photolysis rates calculation: (1) offline calculation with the TUV model, configured for an “8 stream” resolution of the radiative transfer equation; (2) online calculation with the FAST-J model (which also uses an “8 stream” resolution method), for cloudy and clear sky conditions. The meteorological driver is the RAMS v4.3 mesoscale model (Cotton et al, 2003), coupled online with a condensed version of the MOCA chemistry scheme (Aumont et al, 1998). Data used to validate our simulations come from the ESCOMPTE campaign which took place on the southeast of France during summer 2001.

- An estimation of the error introduced by the plane-parallel approximation by comparison between actinic fluxes for heterogeneous clouds and aerosol plumes and their homogeneous plane-parallel equivalents layers (simulated with the Spherical Harmonic Discrete Ordinate Method, SHDOM [Evans, 1998]).