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Lagrangian models of solute transport in the hydrologic response: a theoretical framework for basin-scale applications

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Basin-scale solute transport within the hydrologic response originates by chemical sorption processes occurring between the hydrologic carrier and immobile phases of the hillslopes where solutes are stored, and is thus controlled by the transport dynamics driving the water carrier. We address the Lagrangian formulations of reactive solute transport at catchment scale, revised in the framework of the travel time distributions, by means of two different approaches known from the literature: the Lagrangian stochastic advective reactive model (LaSAR) and the Mass Response Functions (MRFs). Both models adopt a spatially distributed framework where the catchment is represented by a set of geomorphic units subjected to different initial and boundary conditions, the major difference between them relying in the chemical description of the sorption processes: within the MRF approach, in fact, mass exchange between fixed and mobile phases are described by neglecting the spatial gradients of the immobile concentration within each geomorphic state of the catchment (locally well-mixed assumption). A more refined (and quite elegant) representation of the undergoing chemical processes, capable of retaining the spatial distribution of the solute mass sorbed in the immobile phase, is achieved by the LaSAR model. It suffers, however, from the limitations stemming from the prescribed steadiness of the flow field driving the water flow. A comparison between the MRF and LaSAR models within single geomorphic states characterized by different travel time distributions has allowed the assessment of the range of conditions under which the MRF's well-mixed

assumption is reasonable, suggesting that in many practical cases of interest the spatial distribution of the immobile concentration of solute within the geomorphic units of the catchment leads to a negligible effect on the resulting solute fluxes and loads. Accordingly, the results also evidence the effectiveness of the catchment mixing in smoothing chemical heterogeneities operating at the smaller spatial scales. The major operational advantages of the MRF approach are related to its capability of including hydrologic unsteady flow forcing, and are here shown through the application of the MRFs to model the nitrate release from diffuse sources during intense rainfall events in an agricultural basin of North-Eastern Italy. A discussion on implications on spatial patterns and analysis of water quality modelling will be also presented.