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Reactive Gas transport in soil: Kinetics versus Local Equilibrium Approach

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Gas transport through the unsaturated soil zone was studied using an analytical solution of the gas transport model that is mathematically equivalent to the Two-Region model. The gas transport model includes diffusive and convective gas fluxes, interphase mass transfer between the gas and water phase, and biodegradation. The influence of non-equilibrium phenomena, spatially variable initial conditions, and transient boundary conditions are studied. The objective of this paper is to compare the kinetic approach for interphase mass transfer with the standard local equilibrium approach and to find conditions and time-scales under which the local equilibrium approach is justified. The time-scale of investigation was limited to the day-scale, because this is the relevant scale for understanding gas emission from the soil zone with transient water saturation. For the first time a generalized mass transfer coefficient is proposed that justifies the often used steady-state Thin-Film mass transfer coefficient for small and medium water-saturated aggregates of about 10 mm.

The main conclusion from this study is that non-equilibrium mass transfer depends strongly on the temporal and small-scale spatial distribution of water within the unsaturated soil zone. For regions with low water saturation and small water-saturated aggregates (radius about 1 mm) the local equilibrium approach can be used as a first approximation for diffusive gas transport. For higher water saturation and medium radii of water-saturated aggregates (radius about 10 mm) and for convective gas transport, the non-equilibrium effect becomes more and more important if the hydraulic residence time and the Damköhler number decrease. Relative errors can range up to 100% and more. While for medium radii the local equilibrium approach describes the main features both of the spatial concentration profile and the time-dependence of the emission rate, it fails completely for larger aggregates (radius about 100 mm).

From the comparative study of relevant scenarios with and without biodegradation it can be concluded that, under realistic field conditions, biodegradation within the immobile water phase is often mass-transfer limited and the local equilibrium approach assuming instantaneous mass transfer becomes rather questionable.

References

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