

Comments from Referee #2

Eq. (24) is considered as the major contribution in this paper. However, it is a frequency domain solution and requires numerical inverse transform to simulate concentrations as a function of time. Since this is pure closed-form solution, the authors may switch the focus on the effect of rate-limited sorption on predicted concentrations. I agree that this is a progress in hydrology community. The authors have done a good review on the topic. From sequential reaction without sorption, to that with linear sorption, to rate-limited sorption, this paper brings analytical models closer to reality. Even though it is semi-analytical, it will benefit the community of biogradation of chlorinated solvents.

I also suggest the authors to move ahead on solutions in 2D and 3D and possibly improve BIOCHLOR using rate-limited sorption.

Author's response

The authors sincerely thank the anonymous reviewer for his helpful comments and suggestions which will substantially improve the quality of the paper. This paper attempts to develop an analytical model for multispecies transport subject to rate-limited sorption. Indeed, we obtain the analytical solution in the Laplace domain and the analytical inverse Laplace transform for Eq. (24) cannot be carried out to yield the solution in exact fashion at present. Alternatively, the numerical method is instead to complete the inverse Laplace transform to yield the solution. Generally, the solution that is obtained in the Laplace domain and the real concentration is achieved by using numerical Laplace inverse transform is referred to as a semi-analytical solution. It is noted that the semi-analytical solution is exact in the space domain but approximated in time domain. Researchers have evaluated computation efficiency of semi-analytical solutions. For instance, Huang and Goltz (1999) derived an exact analytical solution to

equations describing rate-limited soil vapor extraction of contaminants in the vadose zone. Their solution contained a summation of the infinite series expansion. Huang and Goltz (1999) compared their exact solution against the semi-analytical solution with the aid of numerical Laplace inversion presented by Goltz and Oxley (1994). The numerical Laplace inversion solution is useful for Peclet number (Pe) up to 1600, while the exact solution is only useful at values of Pe less than 12 because of problems accurately evaluating high-order Bessel functions of the first and second kind using double precision accuracy. Despite that the semi-analytical solution generally have better computational efficiency, there is still a need to obtain the exact solution. We will pursue the exact solution in the near future. We fully accept the suggestion that the focus should be put out on the effect of rate-limited sorption on predicted concentration. The work is a pioneering study to bring the rate-limited sorption into the problem of multi-species transport and the newly developed model will have more real world applications. Moreover, the concept presented in this study can be extended to derive analytical models in multi-dimensional media subjected to sequential decay chain reactions and rate-limited sorption, for which analytical solutions are not currently available. Additionally, we expect that we can improve BIOCHLOR by replacing the equilibrium-controlled sorption with the rate-limited sorption in the near future.

References

- Goltz, M.N., Oxley, M.E., 1994. An analytical solution to equations describing rate-limited soil vapor extraction of contaminants in the vadose zone. *Water Resources Research*, 30(10), 2691-2698.
- Huang, J., Goltz, M.N., 1999. Solutions to equations incorporating the effect of rate-limited contaminant mass transfer on vadose zone remediation by soil vapor extraction.

Water Resources Research, 35(3), 879-883.

Author's response

Author's changes in manuscript.

1. We have provided a discussion on the numerical Laplace inversion of Eq. (24) in the revised manuscript.
2. We have added a discussion on the advancement of 1-D model to 2/3 D model in the revised manuscript.