



A stable isotope model for combined source apportionment and degradation quantification of environmental pollutants

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Natural attenuation can represent a complementary or alternative approach to engineered remediation of polluted sites. In this context, compound specific stable isotope analysis (CSIA) has proven a useful tool, as it can provide evidence of natural attenuation and assess the extent of in-situ degradation based on changes in isotope ratios of pollutants. Moreover, CSIA can allow for source identification and apportionment, which might help to identify major emission sources in complex contamination scenarios. However, degradation and mixing processes in aquifers can lead to changes in isotopic compositions, such that their simultaneous occurrence might complicate combined source apportionment (SA) and assessment of the extent of degradation (ED). We developed a mathematical model (stable isotope sources and sinks model; SISS model) based on the linear stable isotope mixing model and the Rayleigh equation that allows for simultaneous SA and quantification of the ED in a scenario of two emission sources and degradation via one reaction pathway. It was shown that the SISS model with CSIA of at least two elements contained in the pollutant (e.g., C and H in benzene) allows for unequivocal SA even in the presence of degradation-induced isotope fractionation. In addition, the model enables precise quantification of the ED provided degradation follows instantaneous mixing of two sources. If mixing occurs after two sources have degraded separately, the model can still yield a conservative estimate of the overall extent of degradation.

The SISS model was validated against virtual data from a two-dimensional reactive transport model. The model results for SA and ED were in good agreement with the simulation results. The application of the SISS model to field data of benzene contamination was, however, challenged by large uncertainties in measured isotope data. Nonetheless, the use of the SISS model provided a better insight into the interplay of mixing and degradation processes at the field site, as it revealed the prevailing contribution of one emission source and a low overall ED.

The model can be extended to a larger number of sources and sinks. It may aid in forensics and natural attenuation assessment of soil, groundwater, surface water, or atmospheric pollution.