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REVIEWS AND ANALYSES

Fate and transport in environmental quality

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Abstract

Changes in pollutant concentrations in environmental media occur both from pollutant transport in water or air and from local processes, such as adsorption, degradation, precipitation, straining, and so on. The terms "fate and transport" and "transport and fate" reflect the coupling of moving with the carrier media and biogeochemical processes describing local transformations or interactions. The *Journal of Environmental Quality* (JEQ) was one of the first to publish papers on fate and transport (F&T). This paper is a minireview written to commemorate the 50th anniversary of JEQ and show how the research interests, methodology, and public attention have been reflected in fate and transport publications in JEQ during the last 40 years. We report the statistics showing how the representation of different pollutant groups in papers changed with time. Major focus areas have included the effect of solution composition on F&T and concurrent F&T, the role of organic matter, and the relative role of different F&T pathways. The role of temporal and spatial heterogeneity has been studied at different scales. The value of long-term F&T studies and developments in

Abbreviations: F&T, fate and transport; LULC, land use and land cover.

modeling as the F&T research approach was amply demonstrated. Fate and transport studies have been an essential part of conservation measure evaluation and comparison and ecological risk assessment. For 50 years, JEQ has delivered new insights, methods, and applications related to F&T science. The importance of its service to society is recognized, and we look forward to new generations of F&T researchers presenting their contributions in JEQ.

1 | INTRODUCTION

Changes in contaminant concentrations in environmental media occur both due to transport in water or air and due to local processes, such as sorption, degradation, precipitation, or straining. The terms "fate and transport" and "transport and fate" reflect the tight coupling of contaminant movement with the carrier media and physical and biogeochemical processes controlling local transformations or interactions. Usage of these terms underscores the need and intent to characterize the results of such coupling. Fate and transport (F&T) first appeared in publications in 1964 (Horsak et al., 1964) and became actively used by the end of the 1970s. Fate and transport research reports have been omnipresent in the *Journal of Environmental Quality* (JEQ). This journal was the third after *Chemosphere* (Paris et al., 1978) and *Science* (Shnoor, 1981) journals to mention F&T and was the first journal to publish a paper on F&T in runoff (Lorber & Mulkey, 1982). This minireview shows how research interests, methodology, and public attention have been reflected in F&T publications in JEQ during the last 40 years.

For this review we selected papers listed in the SCOPUS database with the term "fate and transport" mentioned in the title, abstract, or keywords. We realize that many JEQ papers do not use the F&T term but de facto report F&T research. We assumed that our paper selection would allow an incomplete but representative view of F&T research as reflected by JEQ. Percentages of publications, including F&T terms for major contaminant groups, are shown in Figure 1, where the time periods used cover approximately equal numbers of papers. Pesticides initially dominated F&T publications in JEQ but encountered a steady decrease. In contrast, F&T of microorganisms—viruses, bacteria, and protozoa—was first mentioned rarely (Jin et al., 2000; Reneau et al., 1989) but recently became dominant. Microbial F&T paper frequency of about 40% is comparable to that of nutrients and pharmaceuticals together.

Water was the dominant carrier of contaminants in the F&T studies referenced in Figure 1, with air as a carrier found only in 5% of papers. The F&T media in order of frequency of studies were soils > unsaturated zone > aquifers > runoff > streams > reservoirs or lakes. Fate and transport research was carried out at lateral scales of field, stream reach, and watershed and at vertical scales of soil horizon, soil profile, vadose zone, and aquifer. The largest number of citations received timely reviews on the F&T of emerging contaminants (Chee-Sanford et al., 2009; Lin et al., 2010).

2 | MAJOR FOCUS AREAS

2.1 | Solution composition and concurrent F&T

Studies showed suspended colloidal particles to be an important F&T factor. Colloids facilitated virus transport through porous media (Jin et al., 2000). Manure dramatically affected *Escherichia coli* attachment to soil (Guber et al., 2005). Complexation of chemical contaminants with bacteria was observed to control F&T in column experiments with cadmium (Li et al., 2019). And F&T in runoff was strongly affected by the attachment to suspended colloids for bacteria and a pesticide (Kennedy et al., 2001; Soupir et al., 2010).

Studying concurrent F&T of several substances appeared to be valuable, as in the research of the nitrate-inhibiting effect on selenate (SeO_4) reduction (Bailey et al., 2012) or phosphate effect on the adsorption–desorption kinetics of vanadium (Sun et al., 2019). In a uranium F&T study, acidic water

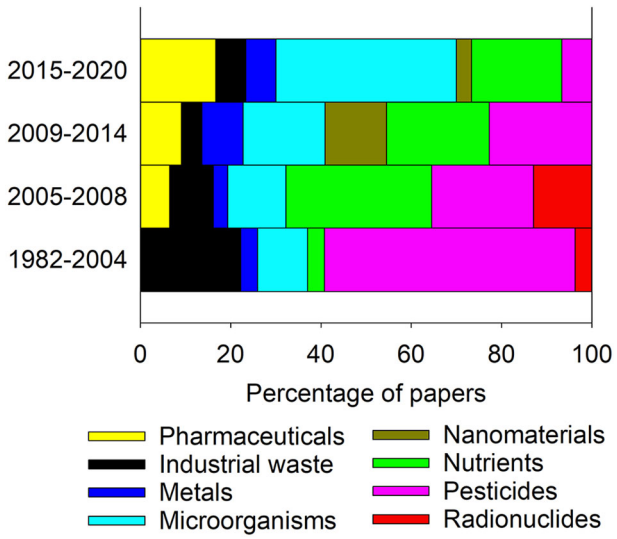

FIGURE 1 Frequency of fate and transport papers for major pollutant groups

created secondary precipitants that cemented quartz grains together and modified pore geometry in the center of the experimental column. This implied that wastes that leaked from buried tanks in the past likely did not migrate vertically but rather in the lateral pathways due to the precipitation of secondary precipitate phases (Um et al., 2005).

Another reason to consider concurrent F&T is that changes in the microbial community caused by the adaptation to contaminants affect the F&T of other solution components. This was the case, for example, when nitrate reduction occurred much sooner than perchlorate reduction in soils that had not been previously exposed to perchlorate, but nitrate and perchlorate were simultaneously reduced in soils previously exposed to perchlorate (Tipton et al., 2003).

Core Ideas

- JEQ was one of the first journals to accept the "fate and transport" terminology.
- Representation of different pollutant groups in papers changed with time.
- The role of temporal and spatial heterogeneity was studied at different scales.
- The value of long-term studies and modeling as the research approach was amply demonstrated.
- Fate and transport studies have been an essential part of conservation measure evaluation.

2.2 | Role of organic matter

The presence of organic substances dissolved and/or suspended in solutions is often an essential factor affecting F&T of contaminants. However, the nature of organic material is of importance. The poultry litter-derived dissolved organic matter was not likely to enhance inorganic phosphorus (P) transport, contradicting the assumption that dissolved organic matter released from organic wastes increases plant-available P when organic amendments and fertilizer P are co-applied (Goyne et al., 2008).

Organic matter is a strong F&T factor affecting the ecological service of soil as a filter. Filipović et al. (2020) suggested that retention of a pharmaceutical carbamazepine in topsoil was enhanced by increasing soil organic carbon. Rich in carbon, biochar was found to be an efficient amendment to prevent hormone transport (Duncan et al., 2015; Mina et al., 2016). Injection of manure showed promise in reducing the potential for off-site losses of hormones from manure-amended soils (Mina et al., 2016). And organic-matter-rich riparian zone soils showed promise in mitigating losses of urea from neighboring fields (Fisher et al., 2016). Tang et al. (2011) noted that carbonaceous adsorbents not only function as a superb sink for organic contaminants but also allow them to be slowly degraded while being trapped.

2.3 | Relative role of different F&T pathways

The relative role of different F&T pathways has been a recurrent subject of studies (Chendorain et al., 1998). At the field scale, different pathways were dominant for different microorganism groups (Rieke et al., 2018). Rieke and colleagues found that surface water *Enterobacter* concentrations were influenced mainly by artificial drainage, whereas *Clostridium sensu stricto* was primarily transported to surface waters by runoff events. Gilfillan et al. (2018) noted distinct ecological drivers of impairment depending on the fecal indicator organism used in their work. *Escherichia coli* survival was driven by increased hardness and heterotrophic activity, whereas bacteriophage detection was inhibited by high levels of coliforms in sediment. Both indicators were influenced by organic matter and P limitation. The form in which contaminants moved was the important F&T pathway role factor. For example, particulate P losses were greater than dissolved P losses in the study of Brendel et al. (2019). These authors noted that "understanding relationships between flow, precipitation, transport pathway, and P fraction at the catchment scale is needed for effective conservation practice implementation" (p. 117).

3 | SCALES AND HETEROGENEITY

3.1 | Spatial and temporal heterogeneity

The role of pore spaces on the mobility of soil water was recognized in early F&T studies. Preferential flow occurring in soil columns was well described using a two-region, mobile–immobile water model, as in the study of bentazon F&T by Gaston et al. (1996) or atrazine F&T in fractured till (Helmke et al., 2005). The two-region model provided satisfactory results for pharmaceutical F&T (Casey et al., 2005; Fan et al., 2007). When flow was facilitated primarily via macropore channeling and actinide was present in the form of oxide particles, the relatively short residence time precluded a continuous interaction between the soil and the flowing water, which minimized the movement of pollutant in the soil (Litaor et al., 1996). In field-scale studies, the hydraulic conductivity of paddy soils, and consequently, heterogeneity of the velocity field, substantially influenced pesticide fate (Miao et al., 2003).

Layering strongly affected F&T of soil constituents because of its potential to increase biodegradation or slow the transport rate (Hansen et al., 2011). Hansen and colleagues

concluded that quantifying coupled hydrologic–biogeochemical processes occurring at small-scale soil interfaces is critical to accurately describing and predicting chemical changes at the larger system scale. Studies of nitrate and pesticide F&T in fractured till suggested that aquifers and surface waters associated with thin, fractured till units may be vulnerable to contamination. Yet these materials may protect deeper aquifers due to increased residence time (Helmke et al., 2005). The controlling role of the residence time in virus F&T was clearly demonstrated in a study using sand-packed columns (Sasidharan et al., 2018).

Temporal heterogeneity could result in estimates of F&T different from that expected based on average values. For example, frequent, small storms had the potential to result in extreme losses of contaminants (Brendel et al., 2019). Accounting for time-scale dependence in F&T drivers improves F&T forecasts.

3.2 | Land use and land cover

Land use and land cover (LULC) was shown to affect F&T at field and watershed scales. Kalin et al. (2010) successfully included LULC in the list of artificial neural networks' inputs to predict water quality in unmonitored watersheds. Gregory et al. (2019) showed that LULC differences could significantly affect background pollutant load transport during runoff events. Differences in root systems of species have affected the F&T of herbicides (Lin et al., 2003). M. Kim et al. (2018) suggested measures to improve our understanding of the impacts of land use change on fecal indicator bacteria in tropical watersheds. Trends in microbial water quality can be driven by land use modifications, including those caused by climate change.

3.3 | Long-term studies

Long-term observations provided insights in possible changes in F&T pathways, processes, and parameters with time. Changes in degradation rates of organic contaminants were observed as the microbial adaptation occurred (Krutz et al, 2008, 2010). Phylogenetic analyses indicated that antibiotic resistance genes have evolved, although some genes have been maintained in bacteria since before the modern antibiotic era (Chee-Sanford et al., 2009). Biosorption was shown to play a role in contaminant transport and to change as microbial populations evolved (Qu et al., 2008). Phillips et al. (2006) provided an example of uranium (U) contamination of interbedded shale and sandstone, where the acidic U-enriched groundwater weathered away calcite veins, resulting in greater porosity, higher hydraulic conductivity, and higher U contamination. The importance of long-term studies of slowly occurring F&T can hardly be overestimated.

Long-term studies have also been instrumental for evaluating transport rates in the vadose zone. Boyle (1989) noted that because chlorinated aromatic hydrocarbons were rare in the environment until a few decades ago, they provide a unique opportunity to study the evolution of multistep catabolic pathways for the degradation of compounds with novel chemical structures. A relationship between groundwater age and the frequency of pesticide detection in the U.S. Midwest showed that pre-1953 water is less likely to contain pesticides because it tends to predate the use of pesticides (Kolpin et al., 1995). Research of pesticide occurrence in groundwater in southern Georgia found a limited number of wells with increases in pesticide concentrations and suggested that groundwater sources of these compounds were not increasing in concentration over the 12-yr time span (Dalton et al., 2008).

4 | MODELING IN F&T RESEARCH

Modeling appears to be an indispensable methodology for studying F&T and integrating results of different studies. More than 30% of F&T papers in JEQ included modeling. Popular models such as CREAMS, RZWQM, SWAT, LEACHM, HYDRUS1D, and HYDRUS2/3D, as well as ad hoc developed models, were used. Whereas mechanistic models dominated the fine scales, empirical models (based on multisite and multiyear observations) demonstrated, to the surprise of authors, strong performances at coarser scales, such as in the case of phosphorus F&T at the watershed scale (Kleinman et al., 2017). Averaging over time the fine-scale dynamics of F&T controls, such as precipitation, could become an impeding factor for mechanistic model applications (Truman et al., 2007). Case studies of mechanistic models at the watershed scale were also reported. For example, Larose et al. (2007) concluded that the model is an effective tool for capturing the dynamics of streamflow and atrazine concentrations in a large-scale agricultural watershed in the midwestern United States. Safaie et al. (2020) demonstrated that the coliphage F&T model for lakes could be used to test hypotheses about potential sources and their behavior and for predictive modeling.

Substantial improvements in watershed-scale models arose from better conceptualization of transport mechanisms, such as for P (Collick et al., 2016; White et al., 2014) and *E. coli* (Hall et al., 2014; Liao et al., 2015). At this scale, models appear to be fairly complex; how much complexity is required in models was a question posed and addressed by Liao et al. (2015). The mismatch between parameters measured in batch experiments and estimated from model calibration has been observed by several authors, for example, Dontsova et al. (2006). Bradford et al. (2014) noted that many model parameters have to be optimized to simulate a diversity of observed transport, retention, and survival behavior even at the

laboratory scale. These authors concluded that improved theory and models are needed to predict the fate of microorganisms in natural subsurface systems that are highly dynamic and heterogeneous.

Modeling was especially beneficial when more than one pathway was involved in F&T, as in the case of simultaneous transport of 1,3-dichloropropen in both liquid and gaseous phases (Wang et al., 2000). A modeling study with multiple scenarios was found to be a useful instrument to evaluate the effect of climate change on F&T in waste management systems (Morales et al., 2015). Kalin et al. (2010) reported the first application of machine learning to estimate water quality across watersheds. These studies show that modeling is a valuable research methodology for F&T resulting from complex interactions.

5 | APPLICATIONS: CONSERVATION AND REMEDIATION MEASURES

Fate and transport studies are an essential part of evaluating and comparing conservation measures (Fisher et al., 2016) as well as ecological risk assessment. Examples in JEQ encompass various topic areas, from risks related to septic tanks (Reneau et al., 1989) and sustainability studies of long-term land application of Class B biosolids (Pepper et al., 2008) to the comparison of managing F&T and input reduction as pollution prevention strategies (Ator et al., 2020). Controlling fate processes can improve best management practices (Kulesza et al., 2016; Mina et al., 2016). Vidon et al. (2019) advocated F&T research to improve our understanding of how riparian water and air quality functions have been affected by various practices, including stream restoration, subsurface drainage, two-stage ditches, beaver dam analogues, denitrification bioreactors and permeable reactive barriers, artificial wetlands, and short-rotation forestry crops. Environmental quality management usually relies on F&T control.

6 | CONCLUSION

Fate and transport of contaminants is an essential component of environmental evolution affecting human and ecosystem health and resilience. The role of F&T research continues to grow, as does the complexity and amount of contaminants released. For 50 years, JEQ has delivered new insights, methods, and applications related to F&T science. The importance of its service to society is recognized, and as editors, we encourage new generations of F&T researchers to present their contributions to JEQ. We anticipate that JEQ will continue to publish F&T papers exploring new experimental techniques, cross-disciplinary approaches, novel modeling tools, artificial intelligence applications, and other innovative advances in the environmental science of our changing world.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

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