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ES&T Guest Comment: Celebrating Bidleman's 1988 "Atmospheric Processes"

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ES&T Guest Comment
Celebrating Bidleman's 1988 "Atmospheric Processes"

Since its 1988 appearance in *ES&T*, Terry F. Bidleman's article, "Atmospheric processes: wet and dry deposition of organic compounds are controlled by their vapor-particle partitioning", has had a notable impact on the field of contaminant science. The paper has been cited in over 600 journal articles published by authors from every continent. Far from fading into obscurity, the paper's influence has been remarkably consistent. Citations over the last year match the annual average attained since publication.

"Atmospheric processes" is an impressive example of scientific synthesis whereby implicitly linked concepts are brought together in explicit form. Bidleman was one of the first environmental scientists to realize that the atmosphere was an important contributor to persistent organic pollutants in aquatic systems. Not only did he strengthen the link between air and water, he laid out the relevant processes in sufficient algebraic detail for the reader to understand them without being overwhelmed.

The paper opened with a succinct presentation of real-world observations regarding the atmosphere as an important pathway in the fate of organic chemicals. The foundation for the rest of the article was then laid by introducing the processes of partitioning and deposition and by describing the size and composition of atmospheric particles.

When "Atmospheric processes" was published, knowledge of the relationship between partitioning and volatility was in its infancy and prevailing opinion implicated adsorption as the responsible mechanism. However, studies identifying conceptual and technical challenges to understanding partitioning were becoming available. Bidleman described those challenges and thereby helped set the stage for much future research.

Bidleman communicated the experimental difficulties associated with separate but concurrent gas- and particle-phase sampling that are necessary to understanding partitioning. He described the predominant filter-sorbent method and the artifacts that arise during its use. The use of diffusion denuders that showed potential for overcoming those artifacts has remained uncommon whereas passive sampling has grown in popularity despite its lack of phase separation and low temporal resolution. Sampling methods that yield accurate phase distributions in ambient air are still needed.

Though partitioning and volatility were known to be linked when "Atmospheric processes" appeared, the appropriate measure of volatility had yet to be definitively identified. Bidleman presented a discussion of saturated vapor pressures and emphasized the use of appropriate sub-cooled liquid values. Furthermore, his observations led him and others to conduct studies using an alternative volatility descriptor, the octanol-air partition coefficient or K_{OA} , which is now used routinely in studies of partitioning between air and several organic compartments in the environment. K_{OA} has been integral to subsequent studies that have expanded the partitioning adsorption paradigm to consider two sorbing particulate phases, one adsorptive (e.g., soot carbon) and the other absorptive (e.g., amorphous or liquid-like organic carbon).

Though Bidleman's 1988 paper devoted substantial attention to gas-particle partitioning, its most important contribution was the linking of partitioning and atmospheric deposition. "Atmospheric processes" included detailed descriptions of wet and dry deposition and the manner in which each is affected by the fractions of semivolatile organic that are in the gas and particle phases.

Bidleman integrated the results of multiple studies to generate conclusions about the effects of gas-particle and gas-water partitioning on wet deposition. Gas-phase washout was expected to follow Henry's Law but the correspondence between measured and predicted values was not always satisfactory. Bidleman urged further work to measure Henry's Law constants for semivolatile organics since the incorporation of their temperature dependence appeared to improve gas-phase washout predictions. For the particle fraction, a wide range of washout ratios had been reported for semivolatile organics. Bidleman showed that compounds with lower volatilities tended to have greater fractions removed by particle washout, an observation that is aligned with expectations based on their gas-particle partitioning.

When examining dry deposition, Bidleman noted that flux measurements implicated both gas and particle deposition as important processes. He recommended further work to improve dry deposition measurements since surrogate surfaces that were intended to collect only depositing particles were likely capturing gas-phase compounds. His observations motivated several studies using novel approaches in semivolatile organic research such as flux towers, micrometeorological methods, and passive samplers. Better knowledge of partitioning thus helped improve our understanding of deposition.

The paper's scientific influence has extended beyond shaping our understanding of partitioning and deposition. Research on the reactivity of polycyclic aromatic hydrocarbons and other semivolatile organics was greatly improved by incorporating knowledge of partitioning and its relationship to vapor pressure. Our grasp of other environmental processes such as secondary organic aerosol formation and the biogeochemical cycling of organic matter and nutrients has also been influenced. Though progress has been made in reducing uncertainties associated with Henry's Law constants, the determination of accurate physico-chemical property data for organic contaminants continues to be a research priority due in part to the observations laid out in the paper.

Bidleman's 1988 contribution led the scientific community and policymakers to consider the substantial role played by the atmosphere in contaminating aquatic ecosystems. By extension, the paper has been influential in understanding the global distribution of semivolatile organics because partitioning and deposition are fundamental processes underlying atmospheric long-range transport. This has had implications beyond the scientific discourse. The potential for long-range transport became a key criterion for inclusion of contaminants in various international agreements such as the Stockholm Convention on Persistent Organic Pollutants.

On a personal note, Terry Bidleman is widely regarded with respect and admiration. His deep understanding of both analytical and environmental chemistry is renowned. He has shown great generosity in discussing his work with colleagues, motivated by improved understanding for all and unhindered by professional competition. Colleagues and students have delighted in his infectious enthusiasm and strong social conscience and have additionally enjoyed his disregard for official dress codes and his penchant for spicy food.

"Atmospheric processes" was viewed as a revelation when it was first published, and it has since become a standard reference in the study of semivolatile organic compounds in the environment. It is included in university course reading lists and has primed countless graduate students for their research on the role of the atmosphere in the fate of organic contaminants. Bidleman's influence is felt by all who study semivolatile organic compounds as the legacy of "Atmospheric processes" continues to unfold.

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Reference

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