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Multidimensional Fluorescence Studies

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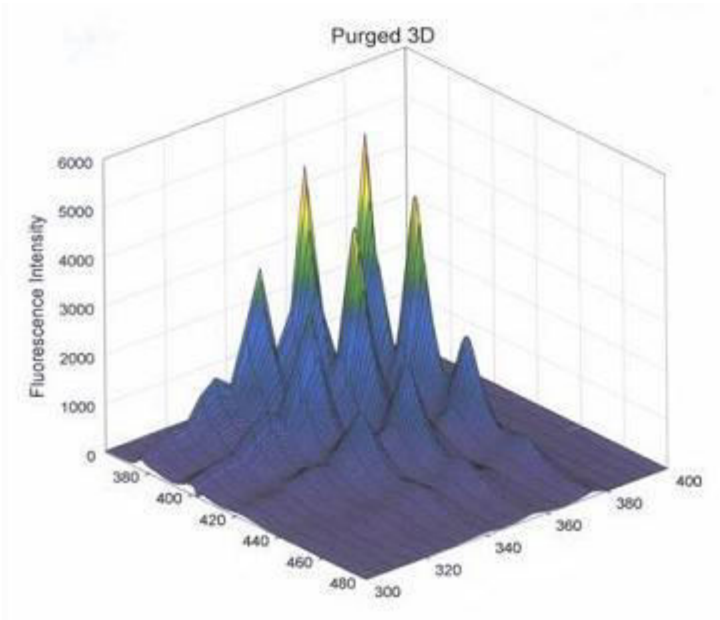
Multidimensional Fluorescence Spectroscopy



Because of its intrinsic dependence on a large number of parameters (excitation wavelength, emission wavelength, polarization, concentration, decay time, sample location, etc.), fluorescence intensity is multidimensional and its measurement has become an important analytical tool in environmental chemistry. In addition, fluorescence measurements may often be performed in situ, offering additional benefits of minimal or no sample preparation, generation of chemical waste, or exposure of workers to hazardous substances. In recent years, the excitation-emission matrix, or EEM, a matrix of fluorescence intensities which can also be plotted as a three-dimensional "fingerprint" of a sample, has gained favor over individual emission spectra or synchro-scanned excitation-emission spectra because of its high information content.

Chemometric Analysis

Extraction of the information in EEMs has been hampered until recently by the possible contributions to the signal from substances not in the investigator's spectral library. That limitation has been transcended, at least apparently, by PARAFAC, an acronym for PARAllel FACtor analysis, a form of multi-way analysis recently introduced into the chemical community. The rotational ambiguity in decomposing spectral data using conventional chemometric tools such as principal component analysis is removed by the additional dimension in the data set. In the case of EEMs, which may be viewed as two-dimensional rectangular arrays of intensities, this may be achieved simply by stacking a set of individual EEMs along an axis to form a three-dimensional "data cube". This third axis may corresponding to sample number; it may also represent a more meaningful dimension such as sample date, pH, etc. (Additional axes may be added to permit the use of PARAFAC and related multi-way analysis methods.)

Subject to certain limitations, PARAFAC decomposes a data cube (or hypercube, in the case of additional axes) into a set of component spectra (excitation and emission) along with a set of scores indicating relative concentration of each component in each sample. The intensity at the jth excitation wavelength and kth emission wavelength of the ith sample is modeled as a sum of contributions from R fluorophores.

The conditions required, namely, that the fluorescence contribution of each component at each excitation emission wavelength pair be trilinear, i.e., a product of an extinction coefficient, a fluorescence quantum yield, and the concentration of the component, are met by solutions of sufficient diluteness, with no interactions among the fluorophores.

Determination of Complexation or Distribution Constants

"Application of PARAFAC to determination of distribution constants and spectra of fluorescent solutes in micellar solutions," Hao Chen and Jonathan E. Kenny, Analyst, 2010, 135, 1704–1710.

Photophysics of Aromatic Molecules: Oxygen Quenching Studies

Evidence from fluorescence quenching experiments suggests that, in addition to the dominant dynamic quenching of naphthalene and pyrene by oxygen, there is a small static component. An understanding of oxygen interactions with aromatic compounds is important for several reasons. Oxygen quenches the fluorescence of most polynuclear aromatic hydrocarbons. Polynuclear aromatic hydrocarbons (PAH) are by-products of petroleum combustion and common environmental pollutants that can be analyzed by fluorescence methods. By removing dissolved oxygen from solution, PAH concentrations can be determined with higher sensitivity.

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Spectroscopic Investigations of Dissolved Organic Carbon

The largest reservoir in the global carbon cycle is represented by the humic substances, the partially decomposed organic matter from dead plants and animals. These complex materials are a key factor in determining soil fertility and metal bioavailability. When present in natural waters as DOM, they can affect drinking water quality, both by fouling membranes used in water treatment and by providing the chemical precursors that become toxic disinfection byproducts such as trihalomethanes (THMs). There is evidence that both the amount and nature of DOM are likely changing as a result of global climate change.

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"Study of pH Effects on Humic Substances using Chemometric Analysis of Excitation-Emission Matrices," H. Chen and J. E. Kenny, Annals of Env. Science 1, 1 (2007).

Fingerprinting of organisms and water samples by location of origin, species, etc.

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Extension of PARAFAC to Interacting Fluorophores

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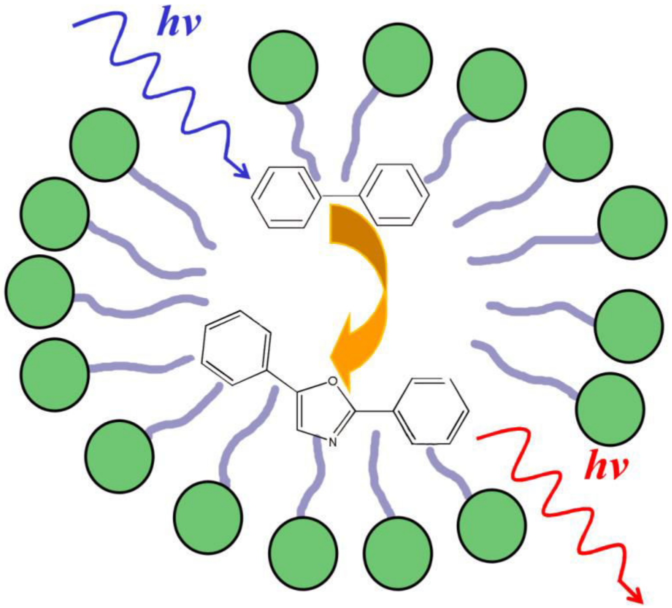

Spectroscopic Methods and Instrumentation

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"Nitrogen Gas Purging for the Deoxygenation of Polyaromatic Hydrocarbon Solutions in Cyclohexane for Routine Fluorescence Analysis," T. Pagano, A. J. Biacchi, and J.E. Kenny, Appl. Spectrosc. 62, 333-336 (2008).

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