CURRENT RESEARCH EXPERTISE

My research focuses on using environmental forensics to identify the impacts of perturbations on biogeochemical cycling at ecosystem interfaces (coastal margins, wetlands, urban atmospheres, lakes, and flooded terrestrial zones). To that end, I have developed analytical and empirical approaches to link field observations to processes of environmental change. In parallel, to address issues of learning and thinking in the environmental sciences, I have incorporated this research into inquiry-based educational models that questions how people learn and take decisions on environmental issues. Please find below a narrative of these research interests with references to publications in each respective field.

Major Orientations in Environmental Research

1) Natural and human-induced fluctuations in material fluxes within watersheds.

Recent research suggests that we need to reassess our conceptual models of watershed function in our working paradigms of elemental cycles, as well as contaminant transport, in these systems. To build more appropriate mass budgets of major elements and contaminants (i.e. carbon and nitrogen, heavy metals and organics) in aquatic systems, we thus need to quantify the role of watershed structure, its alterations (i.e. land use), as well as short- to long-term fluctuations in hydrology on temporal redistribution of natural and anthropogenic materials across land-to-water interfaces[1,2,3,4]. My contributions to this field have focused specifically on quantifying the impact of anthropogenic transformations of drainage basins on the source and fate of terrigenous organic matter (TOM) transferred to aquatic ecosystems. In particular, this work has shown that large-scale land use alterations in tropical to boreal forests lead to substantial remobilization of TOM from soils[5,6,7], which promotes increased carbon sequestration in long term sedimentary reservoirs[5,6,7] and, in some cases, a direct stimulation of primary productivity in receiving aquatic ecosystems[5]. How aquatic environments react does not depend exclusively on the nature and quality of the remobilized TOM but on a combination of factors that range from molecular interactions (i.e. surface sorption of particles), to human activities (urbanization, land use), and large-scale physical processes (i.e. climate-driven shifts in environmental conditions)[5,7,8]. To address these latter processes, a recent NASA-USDA grant was recently awarded to my group[9] to quantify how historical changes in wetland coverage (wetland loss due to urban/sub-urban growth; climate-induced ecosystem replacement of saltmarshes by mangroves) impact carbon sequestration fluxes in ecosystems that lie at the interface between terrestrial and marine as well as pristine and highly-developed environments.

In addition, the extent, density, and configuration of the built environment in urban centers have a direct impact on biogeochemical and hydrological processes. These built environments, with their increased proportion of impervious surface areas (ISA), modify hydrology through the combined influence of peak and base flow alterations and most importantly flashier stream hydrographs. Since urban centers are also substantial sites of pollution *hotspots*, the combination of increased ISA and hydrodynamic pulse events amplifies the rapid and episodic transfer of priority pollutants (toxics, nutrients) to receiving surface and ground water reservoirs[8,10,11]. In particular, the release or remobilization of legacy contaminants from terrestrial and shallow aquatic environments due to continued land and waterway alterations seriously hampers environmental recovery of impaired ecosystems[6,8,11,12]. This in turn can prevent the attainment of environmental standards such as TMDL mandates and amplifies the vulnerability of inland and coastal aquatic resources to oxygen limitations as this process affects the ecological structures of these basins.

2) Pyrogenic by-products in environmental mixtures and their application to combustion monitoring and pollution remediation strategies.

Combustion processes, whether natural (wildfires) or anthropogenic (industrial/urban, agricultural, and vehicular) are major sources of particulate matter (PM), black carbon (BC), and volatile organic carbon to the atmosphere[8,13,14,15,16,17]. Increased emissions of BC-containing aerosols during the 20th century have been implicated in the spinning-down of hydrological cycling at the regional scale, whereas at the global scale they significantly impact the earth's radiative budget. At a more local level, the continued exposure of humans to ultrafine PM and associated contaminants (i.e. polycyclic aromatic hydrocarbons, dioxins) leads to adverse health effects including heightened asthma and respiratory endpoints, and increased risks of cardiovascular disease. Despite our recent understanding on the influence of combustion processes on natural and human systems, large uncertainties still exist in our efforts to characterize spatial and temporal heterogeneities in past and present BC emissions[8,14,18]. Such historical records are paramount to the efforts of climate modelers since they provide datasets against which the wide-scale distribution of estimated BC emissions can be gauged. In short, no model trying to predict the impact of BC on the radiative balance of earth can be derived from fuel utilization and

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emission factors alone. Present theoretical models trying to predict the impact of BC on the radiative balance of earth tend to be derived from fuel utilization and emission factors alone. However, such consumption data are susceptible to the large ranges and non-regional nature of fuel consumption and BC emission factors as well as geographical heterogeneities in particle production and distribution. I highlighted this uncertainty in a series of recent studies, which showed a discrepancy between modeled estimates of soot-BC emissions for the 20th century in the U.S. *vs*. actual reconstructions from historical sedimentary archives in the northeast and northwest[8,14]. In addition to soot-BC, which is predominantly emitted from vehicular and industrial activities, recent research in my lab has shown the link, in the last century, between climate cyclicity, wildfire incidence, and transfer of charcoal-BC to aquatic systems[8]. Although such BC components appear as critical tracers of hydrologic fluctuations (drought *vs*. wet conditions influencing fire events), little is known of the influence of wildfires on aquatic productivity. Because deposited charcoals yield large quantities of easily metabolizable dissolved organic matter[19], one could anticipate that large-scale wildfires in watersheds would support increased metabolism in receiving aquatic systems. However, the significance of this process, particularly under changing climatic conditions, is still unknown at this time.

In addition to the useful information that centennial and millennial scales BC histories could provide towards understanding the role of combustion by-products on physical systems, recent reconstructions of combustion sources and subsequent deposition in urban environments[17,20,] are also vital to the purpose of developing effective environmental and health management policies. Historical reconstructions of toxic loadings to urban airsheds, for example, point to a recent increase in combustion-derived contaminants resulting from widespread increases in urban sprawl in the last decades[8]. Moreover, in urban coastal systems, the majority of the atmospheric contaminants are transferred to the aquatic systems through storm water and short-lived hydrological events, illustrating the link between air and water quality[25]. Because inputs of combustion byproducts are episodic in nature (i.e. seasonal inputs of biomass-derived particulate matter during winter heating and summer wildfire seasons) [17,21], air and water quality are highly heterogeneous on both spatial and temporal scales making the management of these systems a difficult task. Targeted seasonal and episodic monitoring as well as modeling efforts may thus provide the necessary information to solve some of these management difficulties. Finally, although BC is generally regarded as a 'contaminant' in aquatic sediments, its strong sorption capacity for hydrophobic organic contaminants may help reduce uptake by organisms by up to two orders of magnitude[12]. This implies that current environmental risk assessments for these contaminants may need revision to incorporate limited mobility and bioavailability when in the presence of strong sorbing phases such as BC and other geosorbents. More importantly, in some cases, its addition can be used as an *in situ* remediation strategy that avoids costly methods such as sediment dredging[22].

3) Chemical markers in environmental media and analytical developments applied to environmental forensics.

Chemical tracers are extremely useful in the field of environmental forensics as low-level concentrations or isotopic signatures of trace elements and molecules can elucidate specific material source inputs (terrigenous *vs*. marine OM, contaminant sources) and/or processes (fate and mobility of OM and contaminants) in large systems (coastal to ocean basins, atmosphere, soils and wetlands, lakes and reservoirs). I started my career by applying chemical biomarkers to characterize anthropogenically-derived environmental change in aquatic ecosystems[6,7,23,24,25]. I have kept active in this field of chemical biomarker geochemistry for the last fifteen years developing and applying methods tracing lignin constituents, a biomarker of vascular plant organic matter, in terrestrial, aquatic, and now atmospheric media[1,1,5,6,7,12,14,26,27,28,29,30,31,32,33,34]. In particular, this work allowed me to complement my work in environmental process reconstruction with an expertise in methodological development to measure such biomarkers, and a suite of other chemical tracers, in ever increasingly diluted matrices and smaller sample sizes.

Most methodological developments tend to focus on one or a combination of the following approaches: a) improvement to analyte detection (i.e. HR-GCMS, LC/MS, GC/MS–MS)33 in order to optimize sensitivity and selectivity, b) increased throughput using simplified extraction/preparation methods (solid phase extraction, LA-ICPMS, CHN-IRMS)[16,27], and/or c) establishment of analytical protocols to validate extraction recovery and quantitation accuracy[13,14,16]. Although analysis performance is bound to continuously improve and change the landscape of the feasible, a large part of an analyst's work lies in improving the efficiency and speed of sample preparation and demonstrating the consistency of any new method with respect to past analytical efforts. This latter approach is crucial to answer quality assessment/quality control protocols required in environmental monitoring. The development of environmental standards is thus a

critical tool that insures inter-laboratory consistency of the data produced.

Thanks to the present advances in instrumental analysis, chemical constituents can now be more accurately and rapidly determined in a wide range of environmental media (biological tissues, soil and sediment particles, aerosols, dissolved organic matter). The potential for environmental forensics, based on the measurement of such chemical markers in the smallest-sized samples imaginable, has therefore soared and facilitated the multiplication of analyses and their application to the reconstruction of local to global environmental processes. For example, new mass spectrometry systems (MS) now permit routine quantification approaches that only the most expensive instruments could perform a few decades ago. Among these, multiple reaction monitoring (MRM) performed during tandem mass spectrometry (MS-MS) brings an added advantage over other monitoring modes such as full scan (FS) or selective ion monitoring (SIM). Although both SIM and MRM provide increased sensitivity over full scan mode during quantification by decreasing overall noise and focusing on few limited ion(s) per scan time, MRM also increases specificity. Because the product ions are unique fragments of specific ion precursors, tandem mass spectrometry avoids the potential artifact generated by co-eluting extraneous analytes yielding similar isotope mass fragments as the target ion(s). Moreover, tandem mass spectrometry yields mass losses that are indicative of functional groups in the precursor ion, strengthening both identification and structure elucidation of the original molecule. Although the use of tandem mass spectrometry has a few drawbacks with respect to other conventional methods of quantification (time of development, lack of libraries), it is yet another high precision tool in the kit of environmental forensics that permit the source identification of contaminants in difficult samples or the reconstruction of processes from a suite of small environmental samples (aerosols, ice cores, dissolved organic matter)[16,28,33,34].

4) Environmental education.

Multidisciplinary approaches in higher education (whether in practice building or knowledge building programs) are necessary because 1) in an increasingly multicultural world the advancement of knowledge requires cross-linking insights derived from the humanities, social sciences and natural sciences, 2) natural sciences need to address the new social contract, which goals should now include a series of moral and ethical issues for which science can provide enlighten information about the consequences of different choices, and finally 3) we need to build skills in a professional world that now demands flexibility and broad (applied) knowledge. The global reach and impact of human endeavors begs now for a new structure of knowledge that respects its new social contract. During the construction of clear and comprehensible messages in multi- to cross-disciplinary fields, wide open "transaction spaces" are created in which novel, hybrid languages emerge. Environmental issues belong to such highly contextualized transaction spaces in which vastly different stakeholder interests and backgrounds interact and collide. Educating professionals to act in such heterogeneous spaces is an incommensurable challenge if one seeks to cover all domains of interest with the depth usually provided to specialists. Accordingly, a reductionist approach to address such transactions with respect to specific levels of expertise (i.e. scientific, social, economic) is nothing but a selective simplification of the multiple dialogues that take part in these transactions, and as such much is lost in (lack) of translation.

Building practice in multidisciplinary fields thus requires, by necessity, that specific goals are set with respect to distributed accountabilities. Accountability towards disciplinary requirements, accountability towards academic excellence and social relevance, and accountability towards student diversity. And though the terminologies specific to fields as wide ranging as social and natural sciences may never be truly blended, the concept of analysis remains a mutual goal of educational approaches and application outcomes. For example, training multidisciplinary practitioners, as opposed to scholars, may not need to focus on concept generation. Instead, such professionals need to become exceptionally adept at analyzing issues across different fields and take appropriate decisions using such analysis[35,36].

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