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Editorial

Climate change and coupling of macronutrient cycles along the atmospheric, terrestrial, freshwater and estuarine continuum

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ABSTRACT

This paper provides an introduction to the Special Issue on "Climate Change and Coupling of Macronutrient Cycles along the Atmospheric, Terrestrial, Freshwater and Estuarine Continuum", dedicated to Colin Neal on his retirement. It is not intended to be a review of this vast subject, but an attempt to synthesize some of the major findings from the 22 contributions to the Special Issue in the context of what is already known. The major research challenges involved in understanding coupled macronutrient cycles in these environmental media are highlighted, and the difficulties of making credible predictions of the effects of climate change are discussed. Of particular concern is the possibility of interactions which will enhance greenhouse gas concentrations and provide positive feedback to global warming.

1. Introduction

Biogeochemical cycles are fundamental to sustaining life on Earth, and support the productivity and diversity of both natural ecosystems and those managed for food production. Recently, concern has been growing that anthropogenic disruptions to these cycles, especially the macronutrient cycles of carbon (C), nitrogen (N) and phosphorus (P), are damaging these support functions and associated ecosystem services (e.g. Finzi et al., 2011a). Human activities have considerably increased flux rates in all three cycles—the N cycle through inputs from the Haber–Bosch process, and also from combustion of fossil fuels (Erisman et al., 2008); the P cycle from mining P deposits (Cordell et al., 2009); and the C cycle from fossil fuel combustion and increased biomass burning (IPCC, 2007). These increases have made our current industrial civilisation possible—the enhanced N and P inputs are needed to feed the current and future human population (Godfray et al., 2010), and the C from fossil fuels provides most of the energy needed to drive human activity. However, the side effects of these inputs are becoming increasingly troublesome: the increase of CO_2 in the atmosphere is generating global warming (IPCC, 2007) and leakage of N and P from agricultural systems and human waste disposal systems is leading to a range of problems in all environmental media and ultimately posing a threat to human health. These problems include nutrient enrichment of soils; eutrophication of water bodies; reduced drinking water quality; loss of biodiversity; impaired air quality and general degradation of the biosphere support systems which can be characterised as ecosystem services (e.g. Chapin et al., 2010). With these problems in mind, it is clear that our understanding of macronutrient cycles, and particularly how they respond to human perturbations and how they are linked together, needs to be considerably enhanced in order to predict future problems and suggest remedies. To this end, various research programmes are under way, such as the UK Natural Environment Research Council (NERC) Macronutrient Cycles Programme, described by Whitehead and Crossman (2012-this issue). This Special Issue of *Science of the Total Environment* entitled "Climate Change and Coupling of Macronutrient Cycles along the Atmospheric, Terrestrial, Freshwater and Estuarine Continuum" is a contribution to this research effort, and is dedicated to Colin Neal, whose 300 or so peer-reviewed publications span all these environmental media, and have made a huge contribution to the evidence base for understanding these issues.

Better understanding of macronutrient cycles might help decrease the tension between several important global agendas. One is food security: given the projections of population growth and improving living standards, food production will need to increase in coming years, requiring even greater inputs of N and P fertilizers (Godfray et al., 2010). Meeting the population's food requirements whilst preserving our natural resources and capital and decreasing the impacts of nutrients on ecosystem services, biodiversity and human health is now a priority encompassed in the term 'sustainable intensification' (Royal Society, 2009; Foley et al., 2011). There are problems other than excessive supply of nutrients. The P cycle is a sedimentary cycle in which the major global pools of P are geological deposits with a residence time of c.100 Myr (Schlesinger, 1997), turning

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them effectively into a non-renewable resource on human timescales. The rapid mining of currently economic reserves of P has led to fears of a P crisis (short supplies, high prices)—P fertilizer prices have already increased beyond the means of farmers in some poorer nations (Elser and Bennett, 2011).

Understanding the influence of climate change and increasing CO_2 concentrations on macronutrient cycles at all scales will be essential for accurate predictions of future global warming and avoidance of nasty surprises in the future. Research into macronutrient cycles represents a major challenge because it requires the integration of disparate science disciplines, and needs to capture processes and fluxes operating at many different spatial and temporal scales. Changes in ecosystems in response to environmental drivers or mitigation measures may not manifest themselves for decades, making them difficult to estimate. Nevertheless this research challenge is a necessary one and the papers in this issue seek to contribute to current understanding of macronutrient cycling rates and processes; the interactions between C, N and P at different scales; and flux exchanges and transport processes at the air–land–freshwater–estuarine interfaces, and to assess the implications for productivity and eutrophication against a backdrop of climate change.

2. Coupled macronutrient cycles

It has long been realised (e.g. Bolin and Cook, 1983; Schlesinger, 1997) that biogeochemical cycles are not independent of each other, but are coupled (i) through common chemical and biological reactions; (ii) with other elements e.g. Fe, Mo, Mn, S, via microbial metabolism (Burgin et al., 2011); and (iii) with ecological dynamics (Halliday et al., 2012-this issue; Neal et al., 2012-this issue; Palmer-Felgate et al., 2011). As an example, the enzyme nitrogenase which is responsible for N fixation requires both iron (Fe) and molybdenum (Mo) as co-factors (see Finzi et al., 2011b). Where Fe and Mo are in short supply, their availability may limit the rate of N fixation, and where N availability controls primary productivity, which is a widespread situation in terrestrial, freshwater and estuarine environments, C assimilation and P uptake may be limited too. Thus the C, N and P cycles are coupled to the less-studied Fe and Mo cycles.

Macronutrient cycles are coupled over *time* on various scales (Manzoni and Porporato, 2011). For instance, diurnal cycling of dissolved organic carbon is linked to the changing balance between net heterotrophic and net autotrophic activity in daylight and dark hours, and may affect N and P uptake and release. Event-based dynamics occur where biogeochemical cycles vary between baseflow and stormflow conditions, with control of cycling passing to the wider catchment during stormflow (Manzoni and Porporato, 2011; Halliday et al., 2012-this issue). There are seasonal patterns (Halliday et al., 2012-this issue; Neal et al., 2012-this issue), and even longer (decadal, century) timescales related to the retention and release of macronutrients from ancient stores. The demonstration (Trimmer et al., 2012-this issue) that caddis fly larvae in a British river obtain a significant amount of C from chemoautotrophic bacteria feeding on methane raises the possibility that ancient C derived from groundwater is supporting contemporary food webs and biogeochemical cycles.

Macronutrient cycles are coupled in *space*. The atmospheric, terrestrial, freshwater, estuarine and marine systems form a continuum which is connected physically by fluxes of water, gases and aerosols (Manzoni and Porporato, 2011). However, the hydrological flows are overwhelmingly one-way down the continuum, from atmosphere to ocean, and this has important implications. Dawson et al. (2012-this issue) suggested that particulate organic carbon from upstream in the River Dee, Scotland, became more reactive and cycled faster as it passed downstream into areas influenced by arable agriculture which have higher nutrient concentrations.

Stoichiometry couples the macronutrient cycles and is an important control on productivity down the terrestrial–aquatic estuarine continuum (e.g. Schade et al., 2011). Sterner and Elser (2002) suggested that a study of the stoichiometric ratios of the major elements in organisms and their environment could elucidate many of the factors controlling ecological dynamics. Research attention has focussed on simple inorganic forms of the macronutrients since these are the forms which are normally considered to be taken up by plants and autotrophic microorganisms, although recent evidence suggests plants can take up peptide and amino-acid N at similar rates to inorganic N (Hill et al., 2011). However, it is clear from many of the papers in this issue that nutrient transport down the continuum involves many different forms of the macronutrients—notably those where the elements are incorporated into organic compounds (either particulate or dissolved) and particulate inorganic compounds as well as dissolved inorganic forms. Thus there are a plethora of more-or-less well-defined fractions such as dissolved organic C, N and P (DOC, DON and DOP), particulate organic carbon (POC), a whole array of operationally-defined phosphorus fractions such as soluble or molybdate reactive phosphorus (SRP, MRP), dissolved inorganic or hydrolysable phosphorus (DIP, DHP), dissolved reactive phosphorus (DRP), particulate P (PP) and so on. P cycles between the various inorganic forms fairly readily, and there is active exchange between much of the particulate and dissolved inorganic P (e.g. Statham, 2012-this issue), but the role of dissolved and particulate organic P is poorly understood compared to the other fractions. The same ignorance attaches to particulate and dissolved organic N—indeed many of the papers in this volume emphasise the scale of our ignorance of the organic forms of the nutrients. Dungait et al. (2012-this issue) suggest that tracer technology offers the best potential for advances in our understanding of such C, N and P transformations. The paper by Jarvie et al. (2012-this issue) explores the relative role of colloidal (<1 kDa) and truly dissolved substances in the transport of metals and macronutrients in rivers of various characteristics. Colloids dominated the filtered (<0.45 μm) river water fraction in the upland rivers, whereas truly dissolved fractions were proportionally greater in the lowland rivers, probably linked to sewage effluent inputs. Another nutrient which cannot be neglected is silicon, important as a limiting factor for diatom growth in Spring (e.g. Neal et al., 2005).

Conventional wisdom was that there is a shift from P limitation in headwaters to N limitation downstream (e.g. Vitousek et al., 2010)—this view, however, is based on largely pristine environments. But in much of the world, in most ecosystems, macronutrient cycles are now coupled to anthropogenic factors and decoupled from pristine biogeochemical cycles (e.g. Moss, 2012-this issue) by such factors as downstream increases in both P and N from agricultural and wastewater sources, which disrupt these predicted stoichiometric transitions. One highly contested claim is that exclusive focus on P control in rivers has exacerbated N-limited downstream eutrophication in estuaries and coastal waters (e.g. Paerl, 2009). In contrast, Elser et al. (2007) suggested that N and P limitation was surprisingly similar in the terrestrial, aquatic and marine environments. Worrall et al. (2012-this issue) compared modelled land to water flux of N in the UK (2125 k tonnes N yr^{-1}) with estimates of N fluxes to estuarine and ocean systems at the tidal limit (791 k tonnes N yr^{-1}), suggesting in-channel N losses which are equivalent to a loss of 55 kg N ha^{-1} yr^{-1} in the terrestrial catchment. This large figure implies that there is a lot of N metabolism occurring in river channels which would be susceptible to disruption by environmental change, with consequences which are unknown at present. The decoupling of pristine macronutrient cycling has important implications for a variety of issues such as reductions in nutrient use efficiency, nutrient retention capacity and the "self-cleansing" capacity of aquatic systems (see Chapin et al., 2011 and associated papers).

The papers in this Special Issue indicate that cycling and exchange between different 'pools' with different residence/turnover times along the continuum between atmosphere and ocean result in spatial discontinuities in P, N and C retention and storage. Also clear is the