A Coherent Signature of Anthropogenic Nitrogen Deposition to Remote Watersheds of the Northern Hemisphere

Gordon W. Holtgrieve, *et al.*

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An attractive consequence of charge transfer from the ME states is that it may increase the effective MEG yield. Consider the quantum resonance in Eq. 2. Shabaev *et al.* (*18*) showed that when there are other relaxation channels that can move the two states out of resonance with each other, the effective MEG yield is given by Eq. 3:

$$\frac{N_{ME}}{N_{S_1}} = P_{1\rightarrow 2}\frac{\gamma_2}{\gamma_1} \quad (3)$$

where N_{S_1} is the population of the single exciton state; N_{ME} is the total population of MEs and multiple excitons (including those that have successfully undergone multiple electron transfer); $P_{1\rightarrow 2}$ is the probability of forming the ME from the single exciton; and γ_2 and γ_1 are overall relaxation rates from the ME and the single exciton, respectively. Here, γ_1 can include energy relaxation (cooling) of the initial exciton, radiative or nonradiative recombination channels, and (in the case of pentacene) decay into charge transfer excitons (*33*); γ_2 accounts for energy relaxation or localization to form two triplets here or to form band-edge excitons in nanomaterials. When the additional channel of direct and ultrafast multiple charge transfer from the ME states opens, the total γ_2 increases. This effectively shifts the equilibrium to the ME state and increases the MEG yield, which is akin to Le Châtelier's principle for chemical equilibrium.

The discoveries of the coherent superposition of singlet and ME states and associated ultrafast electron transfer from the ME states in pentacene suggest a distinct design principle in solar energy conversion: the harvesting of multiple charge carriers from the ME state. Such an approach may minimize competing Auger recombination processes and maximize MEG yield.

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Supporting Online Material

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A Coherent Signature of Anthropogenic Nitrogen Deposition to Remote Watersheds of the Northern Hemisphere

Gordon W. Holtgrieve,[1]* Daniel E. Schindler,[1] William O. Hobbs,[2] Peter R. Leavitt,[3] Eric J. Ward,[4] Lynda Bunting,[3] Guangjie Chen,[5,6] Bruce P. Finney,[7] Irene Gregory-Eaves,[5] Sofia Holmgren,[8] Mark J. Lisac,[9] Peter J. Lisi,[1] Koren Nydick,[10] Lauren A. Rogers,[1] Jasmine E. Saros,[11] Daniel T. Selbie,[12] Mark D. Shapley,[7] Patrick B. Walsh,[9] Alexander P. Wolfe[13]

Humans have more than doubled the amount of reactive nitrogen (Nr) added to the biosphere, yet most of what is known about its accumulation and ecological effects is derived from studies of heavily populated regions. Nitrogen (N) stable isotope ratios (^{15}N:^{14}N) in dated sediments from 25 remote Northern Hemisphere lakes show a coherent signal of an isotopically distinct source of N to ecosystems beginning in 1895 ± 10 years (±1 standard deviation). Initial shifts in N isotope composition recorded in lake sediments coincide with anthropogenic CO_2 emissions but accelerate with widespread industrial Nr production during the past half century. Although current atmospheric Nr deposition rates in remote regions are relatively low, anthropogenic N has probably influenced watershed N budgets across the Northern Hemisphere for over a century.

Anthropogenic changes to the global nitrogen (N) cycle and the effects of atmospheric reactive nitrogen (Nr) deposition on ecosystems have been appreciated for decades but generally assumed to be confined to areas surrounding population centers and restricted to the latter half of the 20th century (*1*–*3*). The result of increasing amounts of Nr in the atmosphere, primarily as NH_3, NO, NO_2, or peroxyacetyl nitrates, is the long-range transport and deposition on continents and oceans as NH_4^+, HNO_3, or NO_3^- (*3, 4*), even in the most remote ecosystems (*5*–*8*). Human activity is therefore generating a new source of atmospheric Nr that is deposited on otherwise pristine ecosystems and could have important effects on primary producers and food webs (*9, 10*).

[1]School of Aquatic and Fishery Sciences, University of Washington, Seattle, WA, USA. [2]St. Croix Watershed Research Station, Science Museum of Minnesota, Marine on St. Croix, MN, USA. [3]Limnology Laboratory, Department of Biology, University of Regina, Regina, Saskatchewan, Canada. [4]Northwest Fisheries Science Center, National Marine Fisheries Service, National Oceanic and Atmospheric Administration, Seattle, WA, USA. [5]Department of Biology, McGill University, Montreal, Quebec, Canada. [6]Key Laboratory of Plateau Lake Ecology and Global Change, School of Tourism and Geography, Yunnan Normal University, Kunming, Yunnan, China. [7]Departments of Geosciences and Biological Sciences, Idaho State University, Pocatello, ID, USA. [8]Department of Earth and Ecosystem Sciences, Division of Geology, Lund University, Sweden. [9]U.S. Fish and Wildlife Service, Togiak National Wildlife Refuge, Dillingham, AK, USA. [10]Mountain Studies Institute, Silverton, CO, USA. [11]Climate Change Institute, University of Maine, Orono, ME, USA. [12]Fisheries and Oceans Canada, Pacific Region, Science Branch, Cultus Lake Salmon Research Laboratory, Cultus Lake, British Columbia, Canada. [13]Department of Earth and Atmospheric Sciences, University of Alberta, Edmonton, Alberta, Canada.

*To whom correspondence should be addressed. E-mail: gholt@uw.edu

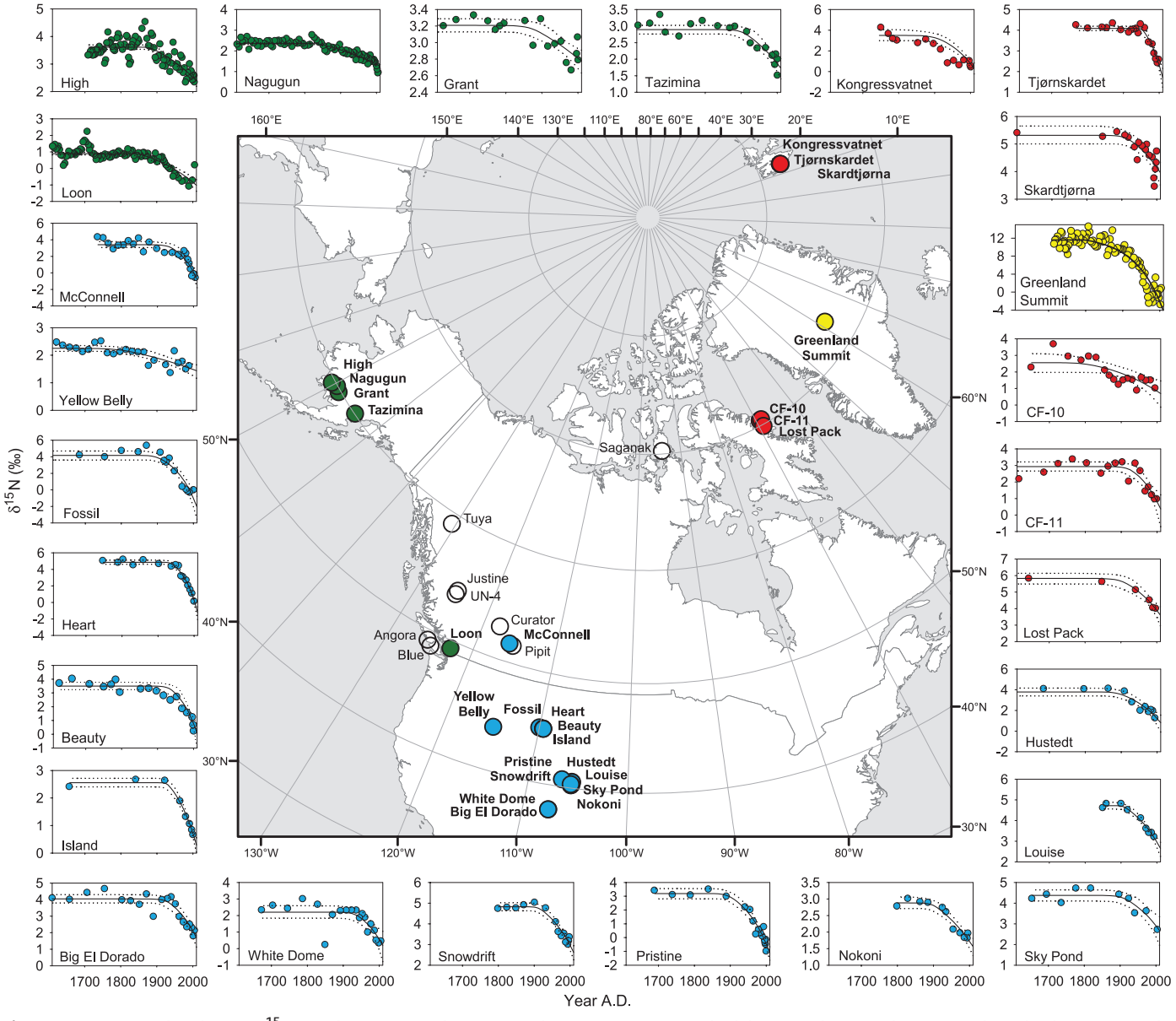

Fig. 1. Locations and sediment $\delta^{15}N$ profiles from Northern Hemisphere lakes grouped by ecotype: temperate/boreal (green circles), alpine (blue circles), and arctic (red circles). Twenty-five of the original 33 lakes were included in the hierarchical model of N isotopic depletion (solid circles). The remaining eight lakes are shown with open circles. The location for the Greenland Summit ice core is indicated with a yellow circle. The solid lines are the median posterior fits to the observed data using the most parsimonious model. The dotted lines are the 2.5 and 97.5% credible limits.

Improved understanding of the spatial and temporal extent of human impacts on atmospheric Nr pools and deposition patterns is challenging because monitoring stations are sparsely distributed, concentrated in areas with intense atmospheric pollution, and have records covering only the past few decades. Determining the mechanisms that drive changes in Nr deposition is also complicated by the numerous Nr sources, including lightning, biomass burning, fossil fuel emissions, fertilizers, and soil emissions. Atmospheric Nr deposited on catchments has multiple fates, including terrestrial uptake, microbial transformations including emission to the atmosphere, or transport in surface and groundwater to lakes and oceans where N can be assimilated by phytoplankton and deposited in sediments. Changes in the N isotopic composition of atmospheric Nr can indicate historic variation in the contributions from human-derived sources, because Nr produced through fossil fuel burning and the production and use of fertilizers has depleted ratios relative to catchment and preindustrial atmospheric N sources (*11, 12*). Isotopic measurements of nitrate-N from the summit of the Greenland Ice Sheet demonstrate a greater than 12 per mil depletion in $\delta^{15}N\text{-}NO_3^-$ deposited over the past 150 years, coincident with increases in NO_3^- concentration (Figs. 1 and 2 and fig. S1) (*11*). Whether this record tracks broader patterns of Nr incorporation in the biosphere remains unresolved.

To evaluate spatial and temporal patterns of Nr deposition in the Northern Hemisphere, we analyzed the N isotopic composition of highly resolved and well-dated lake sediment records from 25 oligotrophic lakes within a hierarchical Bayesian modeling framework (Fig. 1) (*13*). Sediment $\delta^{15}N$ data were fit to a power function model of the form $\delta^{15}N(T_i) = C_0 - C_0^{kT_i} + 1$, where C_0 is the average preindustrial $\delta^{15}N$ and k is the rate constant describing annual change over time. Time (T_i) is the difference between a given year in the historical record (Y_i; expressed in years before 2010) and a critical time threshold when the effects of human contributions to N deposition are estimated to have begun (Y_0) (*14*).

We assessed the strength of support for seven alternative model structures that expressed how trends in sediment $\delta^{15}N$ were related among lakes. Under a hierarchical design, lake-specific estimates of C_0, k, and Y_0 were based on measured

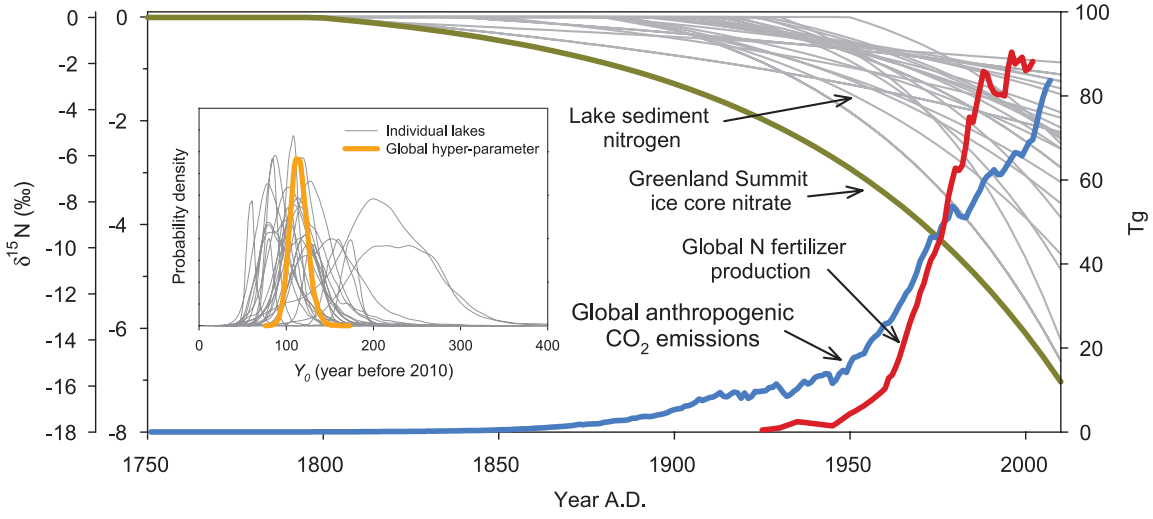

Fig. 2. Relative change in sediment $\delta^{15}N$ from 25 Northern Hemisphere lakes over the past 260 years (inner left ordinate), standardized to a common asymptotic $\delta^{15}N$ (C_0) of zero. Also shown is the relative change in $\delta^{15}N\text{-}NO_3^-$ from the Greenland Summit ice core (outer left ordinate) (*11*), Haber-Bosch N fertilizer production (right ordinate) (*25*), and global anthropogenic CO_2 (right ordinate, data rescaled by 1/100 for plotting purposes) (*26*). (**Inset**) Posterior probability distributions of Y_0, describing the timing of onset of declining sediment $\delta^{15}N$.

historical changes in sediment $\delta^{15}N$ and how parameters were shared among lakes. Models included combinations of global hyperparameters (parameters for each lake were drawn from a distribution with a global mean for Y_0, k, or both Y_0 and k) or ecotype hyperparameters (parameters for each lake being drawn from distributions with ecotype means for Y_0, k, or both Y_0 and k), where lakes were classified as alpine, arctic, or temperate/boreal based on previous analyses and geographic similarities (Fig. 1) (*7, 15*). Last, a fixed-effects model with no hyperparameters was included, in which each lake was described by its own fully independent set of parameters.

Since the beginning of the 20th century, there has been a clear and continuous trend toward isotopically depleted $\delta^{15}N$ in lake sediments, a trend that parallels the record of Nr deposition preserved in the Greenland Ice Sheet (Fig. 1) (*11*). Historical $\delta^{15}N$ time series data were described best by a model that included a global hyperparameter for the critical time threshold ($Y_{0,global}$) identifying the time of onset of a novel N source to these lakes with a depleted isotopic signature (table S1). The posterior probability distribution of $Y_{0,global}$ had a median value of 115 ± 10 (±1SD) years before 2010 (Fig. 2, inset). This timing of departure from baseline N isotope characteristics was earlier than the onset of industrial N fixation but remarkably similar to trends in Nr deposition identified from Greenland (Fig. 2). Both sets of records are coincident with increases in human-caused CO_2 emissions from biomass and fossil fuel combustion, which also emits considerable Nr to the atmosphere. The timing of Nr deposition resulting in depleted $\delta^{15}N$ of sediments was highly coherent among these geographically widespread and ecologically diverse ecosystems, suggesting a common source with a hemisphere-wide footprint throughout even the most remote areas of the Northern Hemisphere. Although initial effects of Nr deposition appeared in the sediment record near the beginning of the 20th century, the most pronounced changes have occurred over the past 40 to 50 years (Fig. 3), during which there has been an expansion of industrial N fixation through the Haber-Bosch process as well as increased fossil fuel emissions from both mobile and stationary sources (Fig. 2) (*1*).

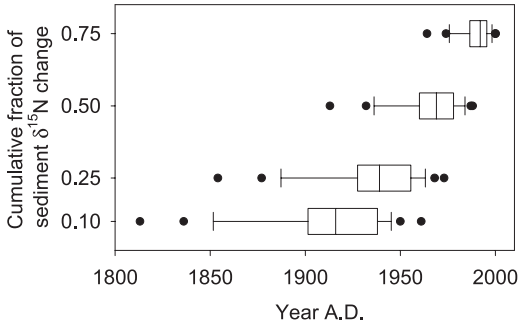

Fig. 3. Timing of cumulative changes in sediment $\delta^{15}N$ based on model results. Data are the year in which 10, 25, 50, and 75% of the total change in $\delta^{15}N$ occurred among the 25 lakes in the analysis.

The rate of change in sediment $\delta^{15}N$ (k) was independent among lakes, and there was variation of lake-specific Y_0 around the global distribution. These findings are not surprising given the range in watershed morphometry, vegetation characteristics, climate, geology, and hydrology of the study sites [Fig. 2, supporting online material (SOM), and tables S3 to S5]. Lake basin and watershed characteristics will affect the sequestration, cycling, and retention of Nr and thus affect how the onset and rate of change are recorded in lake sediments (*3*). We also expected that lake sediment records of Nr deposition would be attenuated relative to the ice core record (Fig. 2), because lake sediments also integrate N from soil, vegetation, and groundwater. Our modeling framework allowed for such variation in these parameters at the scale of individual lakes and identified the best model as describing variation among lakes derived from a common shared effect. The fixed-effects model describing complete independence among lakes was the worst of all models considered. Together, these results imply a common perturbation to the N cycles of lakes distributed across the Northern Hemisphere (table S1).

Several lines of evidence suggest that the coherent timing of changes in sedimentary $\delta^{15}N$ was not derived from common biogeochemical, climatic, or non-Nr anthropogenic causes. Postdepositional mineralization of organic matter (OM) could, in principle, increase the $\delta^{15}N$ of older sediments during burial and contribute to the pattern we have observed. Direct evaluations of the effect of OM mineralization on sediment $\delta^{15}N$ provide few common patterns (*16–18*) (SOM). The magnitude and timing of postdepositional changes should also be a function of the sedimentation rate and be reflected in the C and N stoichiometry of sediments. Sedimentation rates were unrelated to our modeling results, and changes in C:N coincided poorly with changes in $\delta^{15}N$ and were inconsistent across lakes (figs. S1 to S3 and SOM). Compound-specific analysis of sediment algal chlorin $\delta^{15}N$ in one study lake (Sky Pond) showed conclusively that recent changes in $\delta^{15}N$ of algae were the result of anthropogenic Nr and that bulk $\delta^{15}N$ was a conservative estimator of that change (*18*). Climate is unlikely to be the common driver, because the study lakes lie in regions with vastly different patterns of climatic and hydrologic change during the past century,

including warming in northwestern North America and Greenland, increased drought in the western United States, and increased precipitation in the Canadian Arctic (*19*). We can also eliminate the known effects of forest harvest on $\delta^{15}N$ values (*20*), because there is no indication of land-use change in all but one of these watersheds. Finally, although we cannot fully exclude an increased influx of isotopically depleted N from vegetated catchments as temperature increased after the Little Ice Age (*21*) and as glaciers receded (*22*), the broad geographic distribution of our sites suggests that such mechanisms are unlikely to produce the consistent pattern observed. We contend that coherent historical changes in sedimentary $\delta^{15}N$ are explained most parsimoniously by elevated deposition of Nr from atmospheric and, ultimately, human sources.

Although the effects of Nr deposition on watersheds have been studied extensively in more-populated regions of the northeastern United States and Europe, the effects of low-level chronic Nr deposition have not been given the same attention. Our results show that the sources of N to ecosystems throughout the Northern Hemisphere have changed substantially over the past century; however, the biogeochemical and ecological effects of this widespread disturbance are essentially unexplored. A growing body of literature, primarily from arctic and alpine regions, has identified substantial changes in algal communities over a similar 100- to 150-year period, including accelerated turnover in species composition, enhancement of planktonic communities, and greater abundance of nitrophilous taxa (*7, 15, 23*). These ecological shifts have been previously attributed to climate warming and attendant limnological effects, including prolonged lake stratification and an expansion of algal habitat. Our results point to an important alternate and potentially synergistic mechanism that further explains processes underlying the accelerating pace of global ecosystem change.

Two implications emerge from these findings. First, although most of the detectable change in lake N geochemistry from anthropogenic Nr has happened in recent decades with widespread industrial N fixation, it began with fossil fuel use and industrialization near the beginning of the 20th century. Second, despite relatively low current rates of atmospheric Nr deposition in remote areas (*4, 24*), increases since preindustrial times are large enough to be imprinted coherently in the lake sediment record and thus are likely to permeate the biosphere at the hemispheric scale.

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Supporting Online Material

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Caption for Database S1
Database S1

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High Relatedness Is Necessary and Sufficient to Maintain Multicellularity in *Dictyostelium*

Jennie J. Kuzdzal-Fick,[1,2] Sara A. Fox,[1] Joan E. Strassmann,[1,3] David C. Queller[1,3]*

Most complex multicellular organisms develop clonally from a single cell. This should limit conflicts between cell lineages that could threaten the extensive cooperation of cells within multicellular bodies. Cellular composition can be manipulated in the social amoeba *Dictyostelium discoideum*, which allows us to test and confirm the two key predictions of this theory. Experimental evolution at low relatedness favored cheating mutants that could destroy multicellular development. However, under high relatedness, the forces of mutation and within-individual selection are too small for these destructive cheaters to spread, as shown by a mutation accumulation experiment. Thus, we conclude that the single-cell bottleneck is a powerful stabilizer of cellular cooperation in multicellular organisms.

Many of the major transitions in evolution have resulted from groups of lower-level units cooperating so extensively that they merged into a higher-level unit (*1–7*). When the units belong to the same species, such as cells in multicellular organisms and individuals in eusocial insect colonies, high relatedness is essential (*2–7*). High relatedness reduces conflicts among units and can select for some to sacrifice themselves for others, and these factors yield the reproductive division of labor seen in both kinds of systems (germ-soma and worker-reproductive). High relatedness is nearly universal in these entities; eusocial insects likely evolved only from monogamous families [relatedness (r) = 1/2] (*8*), and multicellular organisms are typically clonal cell groups (r = 1) (*5*). But little direct evidence supports the hypothesis that conflict reduction via clonality is either necessary or sufficient to maintain multicellularity in any eukaryotic system.

Theoretical models show that a combination of sufficiently high mutation rates plus within-individual selection could favor the spread of cheaters in clonal systems (*9–11*). It has been suggested that key steps in metazoan development

[1]Department of Ecology and Evolutionary Biology, Rice University, 6100 Main Street, Houston, TX 77005, USA. [2]Section of Integrative Biology, College of Natural Sciences, The University of Texas at Austin, University Station C0930, Austin, TX 78712, USA. [3]Department of Biology CB1137, Washington University in St. Louis, One Brookings Drive, St. Louis, MO 63130, USA.

*To whom correspondence should be addressed. E-mail: queller@wustl.edu