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TITLE: Widespread positive bias in calculated carbon dioxide concentration: causes and implications for large-scale estimates of carbon dioxide efflux from freshwater ecosystems

AUTHORS (FIRST NAME, LAST NAME): Edward Stets[1], Peter A Raymond[2], Gwenaël Abril[3], Alberto Vieira Borges[4], Steven Bouillon[5], Trent Richard Marwick[5], Frederick Tamooh[5, 6], Christian Teodoru[5, 7]

INSTITUTIONS (ALL): 1. National Research Program, U.S. Geological Survey, Boulder, CO, United States.
2. Yale School of Forestry and Environmental Studies, Yale University, New Haven, CT, United States.
3. Environnements et Paléoenvironnements Océaniques et Contenentaux, Université Bordeaux, Bordeaux, France.
4. Unité d'Océanographie Chimique, University de Liége, Liége, Belgium.
5. Department of Earth & Environmental Sciences, Katholieke Universiteit Leuven, Leuven, Belgium.
6. Kenya Wildlife Services, Mombasa, Kenya.
7. Department of Civil and Environmental Engineering, University of Trento, Trento, Italy.

ABSTRACT BODY: Large-scale estimates of carbon dioxide efflux from freshwater aquatic ecosystems provide important insights into the role of these systems in the global carbon cycle. At the largest scales (continental, global), the available data on direct carbon dioxide measurements are not sufficient to provide adequate spatial coverage, and so carbon dioxide is often calculated from pH, alkalinity, and temperature using well-established carbonate equilibrium constants. However, in a globally distributed data set, we find a consistent positive bias in calculated values versus those measured directly. The overall bias is nearly 40 percent, but varies strongly and is most pronounced in with low-pH samples . This finding is particularly important for global and continental carbon dioxide efflux estimates because regions with low pH freshwaters tend to have high carbon dioxide concentrations and so figure prominently in carbon dioxide efflux estimates. We explore several likely causes of the bias in carbon dioxide estimates including dissolved organic carbon contribution to alkalinity in low-pH samples, asymmetric error propagation in pH probes, and improper probe calibration when sampling low-conductivity systems. The outcome of this study will help guide refinements to ongoing efforts to constrain the role of freshwater systems in the global carbon cycle.

KEYWORDS: 0428 BIOGEOSCIENCES Carbon cycling, 1806 HYDROLOGY Chemistry of fresh water, 0414 BIOGEOSCIENCES Biogeochemical cycles, processes, and modeling , 0490 BIOGEOSCIENCES Trace gases.

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Additional Details

Previously Presented Material:

Contact Details

CONTACT (NAME ONLY): Edward Stets

CONTACT (E-MAIL ONLY): estets@usgs.gov

TITLE OF TEAM:
