SCIENCE OF THE TOTAL ENVIRONMENT 400 (2008) 290–314

available at www.sciencedirect.com

ScienceDirect

www.elsevier.com/locate/scitotenv

The effects of land use on fluvial sediment chemistry for the conterminous U.S. — Results from the first cycle of the NAWQA Program: Trace and major elements, phosphorus, carbon, and sulfur

Arthur J. Horowitz[a,*], *Verlin C. Stephens*[b]

[a]*U.S. Geological Survey, Peachtree Business Center, Suite 130, 3039 Amwiler Road, Atlanta, Georgia 30360, USA*
[b]*U.S. Geological Survey, Denver Federal Center, Building 53, MS 415, Lakewood, Colorado 80225, USA*

A R T I C L E I N F O

Article history:
Received 19 March 2008
Received in revised form
21 April 2008
Accepted 21 April 2008
Available online 12 June 2008

Keywords:
Sediment chemistry
Land use
Trace elements
Major elements
Carbon
NAWQA Program

A B S T R A C T

In 1991, the U.S. Geological Survey (USGS) began the first cycle of its National Water Quality Assessment (NAWQA) Program. The Program encompassed 51 river basins that collectively accounted for more than 70% of the total water use (excluding power generation), and 50% of the drinking water supply in the U.S. The basins represented a variety of hydrologic settings, rock types (geology), land-use categories, and population densities. One aspect of the first cycle included bed sediment sampling; sites were chosen to represent baseline and important land-use categories (e.g., agriculture, urban) in each basin. In total, over 1200 bed sediment samples were collected. All samples were size-limited (<63 μm) to facilitate spatial and/or temporal comparisons, and subsequently analyzed for a variety of chemical constituents including major (e.g., Fe, Al,) and trace elements (e.g., Cu, Zn, Cd), nutrients (e.g., P), and carbon. The analyses yielded total (≥95% of the concentrations present), rather than total-recoverable chemical data.

Land-use percentages, upstream underlying geology, and population density were determined for each site and evaluated to asses their relative influence on sediment chemistry. Baseline concentrations for the entire U.S. also were generated from a subset of all the samples, and are based on material collected from low population (≤27 p km^{-2}) density, low percent urban (≤5%), agricultural or undeveloped areas. The NAWQA baseline values are similar to those found in other national and global datasets. Further, it appears that upstream/underlying rock type has only a limited effect (mostly major elements) on sediment chemistry. The only land-use category that appears to substantially affect sediment chemistry is percent urban, and this result is mirrored by population density; in fact, the latter appears more consistent than the former.

Published by Elsevier B.V.

1. Introduction

Since the publication of the Hawkes and Webb (1962) treatise on geochemical exploration, as well as the subsequent publication of a number of geochemical atlases (e.g., Webb et al., 1978; Fauth et al., 1985; Ottesen, et al., 2000), there is a widely accepted perception that land use as well as local mineralogy/petrology (geology) can exert a substantial influence on fluvial and/or lacustrine sediment geochemistry. There are numerous studies describing the effects of ore deposits as well as

* *Corresponding author.*
E-mail address: horowitz@usgs.gov (A.J. Horowitz).

0048-9697/$ – see front matter. Published by Elsevier B.V.
doi:10.1016/j.scitotenv.2008.04.027

abandoned and/or ongoing mining/smelting operations on aquatic chemistry (e.g., Moore and Luoma, 1990; Horowitz et al., 1995; Salomons, 1995; Thornton, 1996; Macklin et al., 1997; Allan et al., 2002a,b; Angelo et al., 2007). Evidence for the impact of other types of land use on water quality has come from numerous divergent studies that have attempted to relate land use to surface and groundwater chemistry (e.g., Osborne and Wiley, 1988; Eckhardt and Stackelberg, 1995; Boutt et al., 2001; Honisch et al., 2002; Pip, 2005; Robinson and Ayotte, 2006; Fitzpatrick et al., 2007), sediment/soil chemistry (e.g., Bloemen et al., 1995; Kelly et al., 1996; Rice, 1999; Birch et al., 2000; Liebens, 2001; Owens and Walling, 2002; Ruiz-Cortes et al., 2005; Reimann et al., 2007), and ecological analyses (e.g., Carral et al., 1995; Ometo et al., 2000; Dauer et al., 2000; Meador and Goldstein, 2003; Tang et al., 2005; Venne et al., 2006). The advent, and subsequently successful application of sediment 'fingerprinting' and source ascription to evaluating the chemistry of suspended and bed sediments, as well as floodplain deposits, has provided additional support for the view that land use can exert substantial influence over sediment chemistry (Collins et al., 1997; Collins and Walling, 2002; Carter et al., 2003; Krause et al., 2003; Collins and Walling, 2004; Walling, 2005; Yeager et al., 2005; Foster et al., 2007).

While there is evidence that both land use and local geology can affect fluvial sediment chemistry, examination of several geochemical atlases, in conjunction with a number of land-use studies, appears to indicate that the relative impact of the former versus the latter, tends to be the result of a combination of geographical scale (e.g., local, regional, national, continental, global) in conjunction with sampling factors [e.g., random versus geostatistical (Bayesian); high versus low density]. Generally, large geographical scale combined with low density sampling is more likely to identify geologic factors whereas limited geographical scale combined with high density or geostatistical sampling is more likely to identify land-use factors (e.g., Garrett, 1983; Keith, 1988; Xie and Yin, 1993; Ramsey et al., 1995; Xie and Cheng, 2001; Reimann, 2005).

In 1991, the U.S. Geological Survey (USGS) began the first cycle of its multidecadal, multidisciplinary, National Water Quality Assessment (NAWQA) Program. The first cycle of the Program encompassed 51 river basins (study units) that collectively accounted for >70% of the total water use (excluding power generation), and 50% of the drinking water supply in the U.S. (Hirsch et al., 1988; Leahy et al., 1990). The selected river basins represented a range of hydrologic settings, rock types (geology), land-use categories, and population densities (Fig. 1). About one-third of the basins were under active study for about 3 years at a time; hence, it took 9 years to complete the first cycle.

NAWQA encompasses a biological component that includes ecological surveys performed in conjunction with the collection of water-quality data, as well as the collection/concatenation of a wide variety of ancillary data covering such factors as land-use percentages, drainage basin area, annual discharge, underlying rock type, and population density. The initial ecological surveys included the simultaneous collection of selected benthic organisms and fish, as well as representative bed sediment samples (Hirsch et al., 1988; Cuffney et al., 1993; Gilliom et al., 1995). The intent was to determine if there were any detectable interrelations between bed sediment and tissue chemistry and/or biological community composition (Gurtz, 1994; Meador and Gurtz, 1994; Gilliom et al., 1995). Sampling sites were selected to represent important land-use categories (e.g., agriculture, forested, mining, urban) in each river basin (Fig. 1). In total, more than 1200 sites were sampled over the 9-year period. To date, despite substantial effort, few if

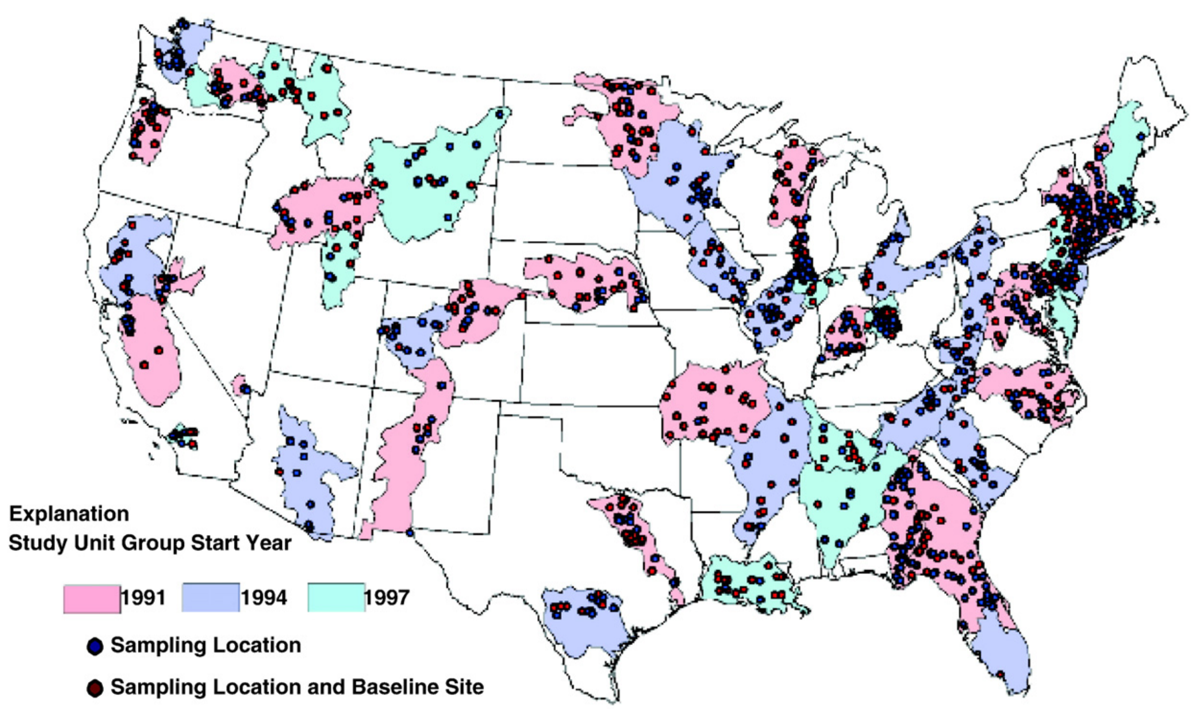

Fig. 1 – A map of the NAWQA study units and the location of all the bed sediment and baseline sampling sites.