Compound specific stable isotopes and specific biomarkers to trace sediment origin and connectivity of sediment source areas to freshwater systems: case of the Baldegg Lake catchment (CH)

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Slope destabilization and associated sediment transfer are one of the major causes of aquatic ecosystems and surface waters quality impairment. Through land uses and agricultural practices, human activities modify the soils erosive risk and the catchments sedimentary connectivity, becoming a key factor of sediment dynamics. Hence, restoration and management plans of water bodies can only be efficient if the sediment sources and their respective contributions, and thus the proportion attributable to different land uses and agricultural practices, are identified. **Sediments can be traced, from their source to their deposition zone, using fingerprinting tools.**
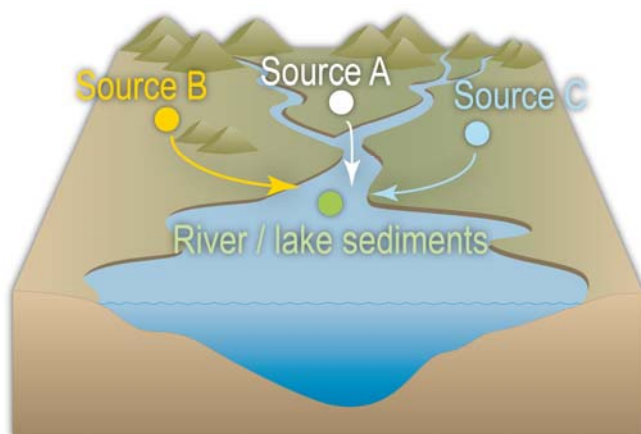

Figure 1 – Fingerprinting concept - Catchment symbol: Courtesy of the Integration and Application Network, University of Maryland Center for Environmental Science (ian.umces.edu/symbols/)

1. Compound Specific Isotope Analysis (CSIA)

Classic fingerprinting techniques (e.g. magnetism, elemental composition, sediment color, radionuclides, $\delta^{15}N_{bulk}$) are not adapted at providing information about the contribution of different land uses to erosion. A new technique, using the compound specific stable isotope (CSSI) signatures of inherent organic biomarkers in the soil (in this project: fatty acids, FAs), is developed since a few years (Gibbs, 2008; Blake et al., 2012; Hancock et Revill, 2013; Cooper et al., 2015; Alewell et al., 2016; Reiffarth et al., 2016). Although all plants produce the same FAs, the carbon stable isotopic signature ($\delta^{13}C$) of those biomarkers is different for each plant species. Because of their polar nature, FAs are easily leached from the plant or the decaying plant material and become tightly bound to soil particles. Coupled to a stable isotope mixing model, CSIA can thus help to discriminate and apportion the source soil contribution from different land-uses.

2. Biomarkers: quantitative approach

To overcome the difficulty in distinguishing some land use types from their isotopic signature (e.g. pastures and some arable lands; Alewell et al., 2016), the CSIA approach can be strengthened by a quantitative approach using biomarkers.

Highly specific biomarkers (HSB), which synthesis is restricted to some plant groups, were studied. Some triterpenes were indeed recently validated as family- or even species-specific (e.g. some triterpenyl acetates for Asteraceae; some sesqui-, di- and triterpenoids for conifers, methoxyserratenes for Pinaceae; pentacyclic triterpenes methyl ethers (PTMEs) for Gramineae, including Cerealia; Lavrieux et al., 2011; Otto and Wilde, 2001; Le Milbeau et al., 2013; Ohmoto et al., 1970; Jacob et al., 2008; respectively). Mostly recently developed for paleo-environmental studies, the strong potential of highly specific biomarkers as sediment fingerprinting tools remains under-exploited.

Besides, we used a linear regression model using the concentrations of *n*-alkanes, *n*-alkanols and FAs. Far more common and more concentrated than HSB, these linear compounds are also far less specific as they are produced by most of the living organisms – the long chains (> C20) being generally produced by higher plants, the shorter ones being more restricted to algae and bacteria. However, the relative concentration of the different homologues is known to vary according to the source organisms.

3. Modelling

The molecular approach was supported by a modified sediment connectivity index (IC) based on the approach by Borselli et al. (2008) and Cavalli et al. (2013). Sediment connectivity is defined as the degree of linkage which controls sediment fluxes throughout landscape, and, in particular, between sediment sources and downstream areas and finally the freshwater system (Cavalli et al., 2013). The identification of connectivity patterns allows for an estimation of the contribution of a given part of the catchment as sediment source, and it defines sediment transfer paths.
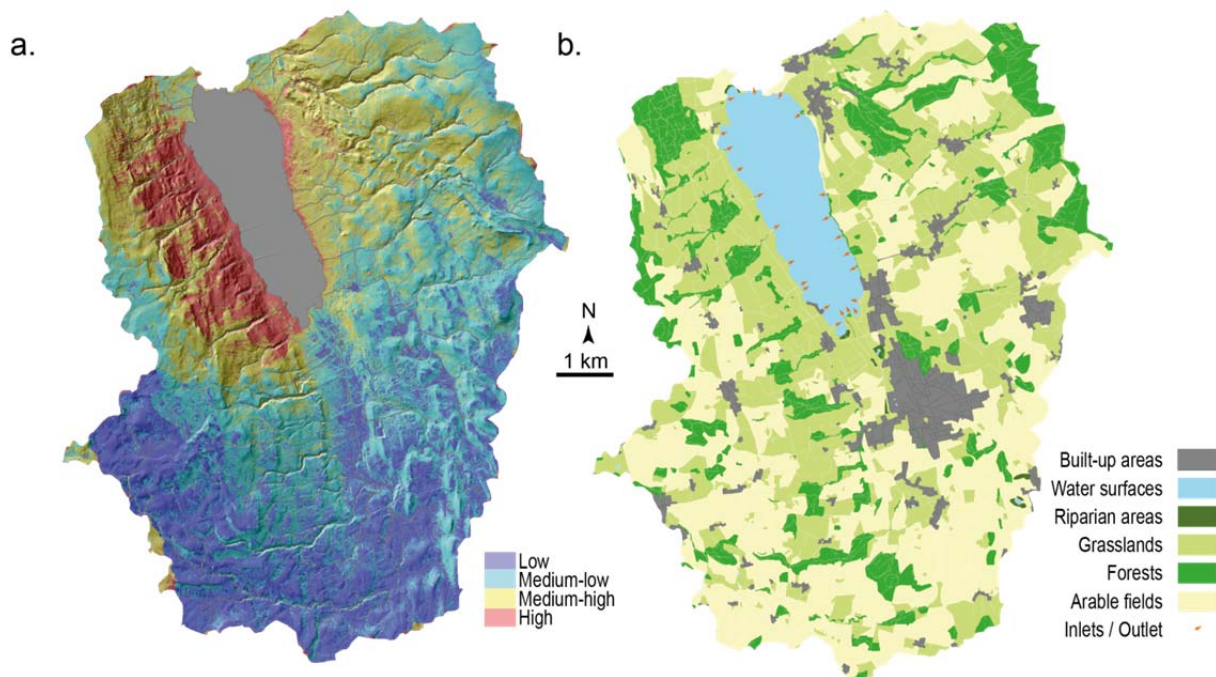
Figure 2 – a. Connectivity map of the Baldegg Lake catchment. b. Land-use map.

4. Study site and sampling

The Baldegg Lake catchment (Canton Lucern, Switzerland) was chosen to apply this combined approach of geochemical fingerprints and sediment connectivity modelling. Almost 80% of the catchment area is used intensively for agriculture, while forests and urban areas cover less than 20%. The lake is eutrophic since the end of the 19th century, despite several attempts to restore the water quality (including an artificial oxygenation system since 1982).

Each land-use geochemical fingerprint was defined from the analysis of plants and soils. The dynamic of sediment input to the freshwater system was assessed on a short and long time scale, using (1) river suspended sediments sampled during high flow conditions and (2) a lake sediment core covering the last 130 years.

5. Main results

The connectivity model was used to define soil sampling strategy. It reveals that the Western part of the catchment has the highest connectivity to the lake. Soils under different land-uses show CSSI signatures distributed along a line which ends are the grasslands and the forests. Orchards and arable lands lie in between. The short-term variability of the sediment export was evaluated with high-flow river suspended sediments. The 5 rivers selected for sampling flow through contrasted dominant land-uses. CSIA shows a high variability in the signatures of each river, covering the whole range of land-use signatures defined in the source soils. A similar trend is present in all the rivers during the vegetation growing season, with values decreasing up to 4 per-mil in 1.5 months. While the CSIA signals of the river sediments are well within the mixing polygon of the endmembers, the CSIA of a 130-yrs long lake sediment core revealed that most of the samples signatures lie out of the source soils mixing line. Other possible sources are the input of fresh vegetal biomass and the contribution of lake autochthonous biomass. The chronological evolution allows defining 4 different units, which match the catchment and lake eutrophication history.

Terrestrial source apportionment was assessed with a biomarker modelling using concentrations of linear compounds (*n*-alkanes, FAs, *n*-alkanols). It reveals the same units as defined in the CSIA, indicating the high response of the lake system to the changes in land-use and agricultural practices.

Our CSIA results emphasize (1) the high seasonal variability of the signatures, as well as (2) the potential major role of sources so far under-evaluated. Biomarker modelling offers promising results with a potential for source-to-sink sediment studies deserving a more in-depth evaluation.