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# Sediment-to-Water Partition Coefficients: the Influence of Physicochemical and Seasonal Factors in Eastern Ontario

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## INTRODUCTION

Sediments often represent an important reservoir for contaminants, such as radionuclides, in aquatic ecosystems (Rowan *et al.*, 1995; Yankovich, 2002). Consequently, lake, stream and river sediments may contribute significantly to the radiological doses received by wildlife, making it important to determine contaminant concentrations in sediments.

Despite the complexity and the dynamic nature of sediments in general, for practical purposes, in environmental impact assessments (EIAs), it is often assumed that radionuclide activity concentrations in different compartments (e.g., water and sediments) are at steady state with respect to one another. Therefore, ratios can be used to estimate concentrations in one compartment given a known concentration in another. For example, sediment-to-water partition coefficients ( $K_d$ ) are often applied to estimate the contaminant concentration sorbed to particulate matter relative to the concentration measured in filtered water. However,  $K_d$  values often range by several orders of magnitude between sampling locations (IAEA, 1994) due to site-specific differences in the nature of the water body, physicochemical conditions in surface waters, seasonal factors, as well as differences in sediment attributes that can affect contaminant partitioning between the dissolved and particulate phases. Consequently, in conducting EIAs, it becomes necessary to either apply generic  $K_d$  values that ensure contaminant concentrations in sediments to which biota are exposed are not under-estimated or to take site-specific measurements of surface waters and sediments to facilitate quantification of  $K_d$  values for application on a site-by-site basis.

To gain a better understanding of the site-specific factors that could influence the variability of  $K_d$ , systematic sampling of surface waters and bed sediments was carried out at 119 locations in Eastern Ontario between 2001 and 2006. The results of this sampling programme are discussed in this paper.

## MATERIALS AND METHODS

Sampling of surface water and sediments was conducted over the ice-free season between 2001 and 2006 to quantify contaminant concentrations in sediments relative to surface waters in Renfrew County, Canada. Surface water and sediment samples were collected at a total of 27 locations on Atomic Energy of Canada Limited (AECL)'s Chalk River Laboratories (CRL) site and at 92 off-site locations to facilitate the characterization of sediment-to-water partition coefficients ( $K_d$ ) for application in EIA.

Sampling locations were selected to cover four categories that had been identified to represent near-field and regional (or baseline) locations in the Ottawa River and in inland surface waters, including: 'inland' waters (on-site of CRL); inland waters (regional or baseline);

Ottawa River (near-field); and Ottawa River (regional or baseline). Inland waters were subdivided into three categories, including lakes, ponds and streams, whereas the Ottawa River locations were characterized as upstream of CRL, adjacent to CRL and downstream of CRL. Where possible, on-site sampling locations were selected to complement sites sampled routinely as part of CRL's Radiological Monitoring Program.

Sampling locations were selected at off-site locations to assess regional environmental quality in areas surrounding the CRL site that are not thought to be influenced by CRL's routine operations. Off-site locations were selected based on the following criteria: minimal anthropogenic contributions of contaminants; availability of information on a given water body; size (i.e., surface area, and where possible, bathymetry); expected sensitivity to acid precipitation (which will influence contaminant speciation and bioavailability); presence of predatory fish species (which can influence foodweb transfer of contaminants); composition of the fish community, where possible (which can influence contaminant transfer and exposure pathways); the presence of major roads in the catchment (which can introduce salts, heavy metals and other contaminants into surface waters); and accessibility.

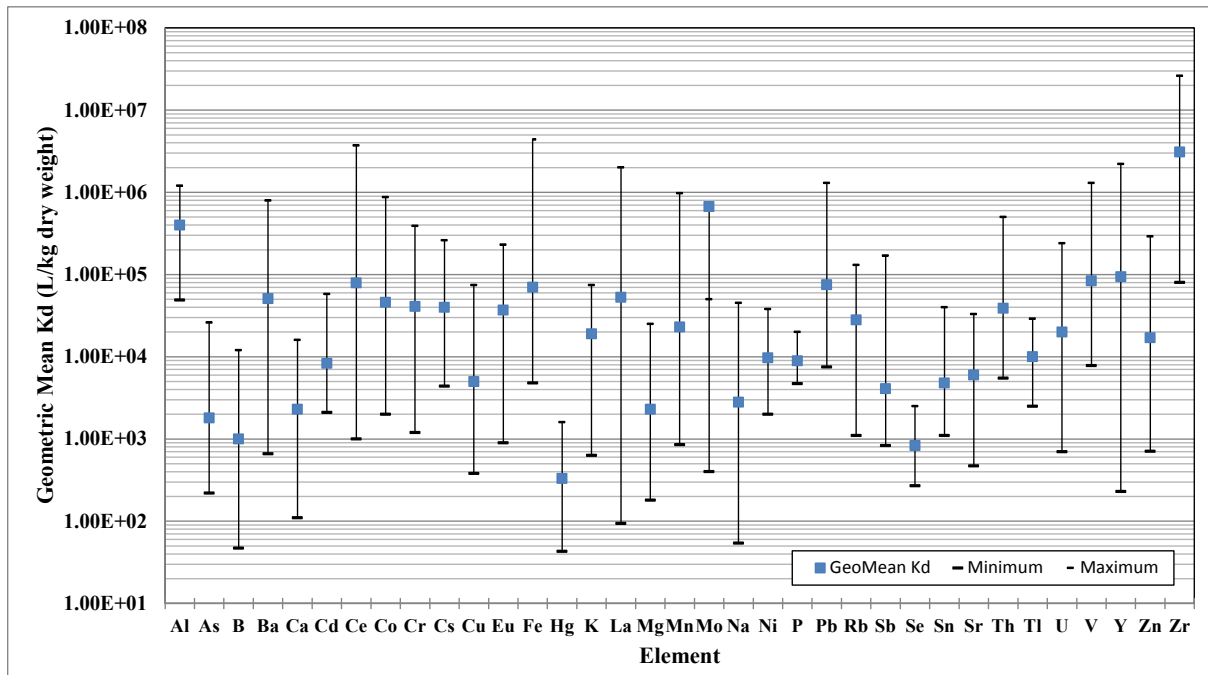
Water was sampled, filtered through a 0.45- $\mu\text{m}$  cellulose acetate filter, and decanted into a trace element-grade, pre-cleaned, polyethylene bottle (Series 300). Filtered samples were then immediately acidified in the field to 1 % using ultra-trace grade, Seastar nitric acid ( $\text{HNO}_3$ ) and maintained at 4 °C until analysis. Sediments were collected in the littoral zone of water bodies from the surface to depths of 5 to 10 cm, placed into 20-mL, plastic bioassay cups with lids, and stored at 4 °C until processing. During processing, sediments were dried at 70 °C until no change in sample mass could be detected, disaggregated using a mortar and pestle, and sieved through a 2-mm sieve. The finer fraction was sub-divided using a mechanical splitter (Soiltest Model EI23-3360) to obtain representative sub-samples of approximately 0.1 to 0.15 g for analysis. Homogeneous sediment sub-samples were acid digested, by placing them into microwave vessels to which concentrated ultra-trace grade, Seastar nitric acid ( $\text{HNO}_3$ ), hydrofluoric acid (HF) and hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) were added. Vessels were then heated under pressure for 33 minutes in the Milestone MLS1200 microwave unit. Once cooled, the supernatant was transferred to clean centrifuge tubes and the residue was washed into small, glassy carbon crucibles. The residue was dried,  $\text{Na}_2\text{O}_2$  beads added and fused at approximately 900 °C for 5 minutes. The cool melt was dissolved in dilute  $\text{HNO}_3$ , and the microwave and fusion fractions were analyzed separately.

A total of 35 elements were analyzed in water and sediments by Inductively Coupled Plasma-Mass Spectroscopy (ICP-MS) and ICP-atomic emission spectroscopy (AES) (Figure 1). In cases where a given element was detectable in both sediments and water, the  $K_d$  values were estimated.

## RESULTS AND CONCLUSIONS

In general, for the elements covered in this study,  $K_d$ s ranged over several orders of magnitude between sampling locations, particularly when data from all locations were pooled (Figure 1); however, it is important to note that in many cases,  $K_d$  values showed a tighter range when categorized by location (i.e., off-site versus on-site) and by type of water body (i.e., lake, pond, upstream of CRL in the Ottawa River, etc.). In addition, water bodies with larger volumes and correspondingly larger capacities to dilute contaminant inputs, such as lakes and the Ottawa River, generally tended to show lower inter-site variability than did smaller water bodies, such as streams and ponds (e.g., Figure 2), possibly due to their greater ability to dampen the magnitude of seasonal changes in dissolved contaminant concentrations

that may be related to seasonal changes in the water budget. Therefore, it may be appropriate to distinguish water bodies by type and location for application in EIAs (Environment Canada, 1997).



**Figure 1:** Geometric mean sediment-to-water partition coefficients ( $K_d$ ), with minimum and maximum values for pooled data from Renfrew County ( $n= 2$  to 96).

In many cases, sites draining CRL Waste Management Areas (WMAs) tended to show relatively lower  $K_d$  values than those measured at off-site locations, possibly due to relatively higher contaminant concentrations in the water (which represented the denominator in the equation used to estimate  $K_d$ ). As expected, such trends seemed to be pronounced in water bodies, such as inland ponds and streams, which typically have relatively small volumes, and therefore, tend to respond significantly to seasonal changes in the water budget (e.g., due to dilution during snowmelt).

There are two main observations from this study: (i) If regional  $K_d$  values were applied to surface waters on the CRL site, in some cases, contaminant concentrations could be *over-estimated* in the sediments, particularly in small surface waters draining WMAs. Therefore, in such cases, it may be useful to apply the site-specific  $K_d$  values in areas impacted by site activities for EIA. (ii) In general, geometric mean  $K_d$  values and  $K_d$  ranges estimated based on regional sediment and water data that have been collected in Renfrew County tended to be higher than  $K_d$  values recommended in the scientific literature (e.g., IAEA 1994 and 2001). Application of recommended literature  $K_d$  values may not be appropriate for Renfrew County, which represents a Canadian Shield environment, consisting of soft waters with poor buffering capacities, since it could lead to an *under-estimation* of contaminant concentrations in the sediments (equivalently, an *over-estimation* of water concentrations if applying literature  $K_d$  to measured sediment concentrations).

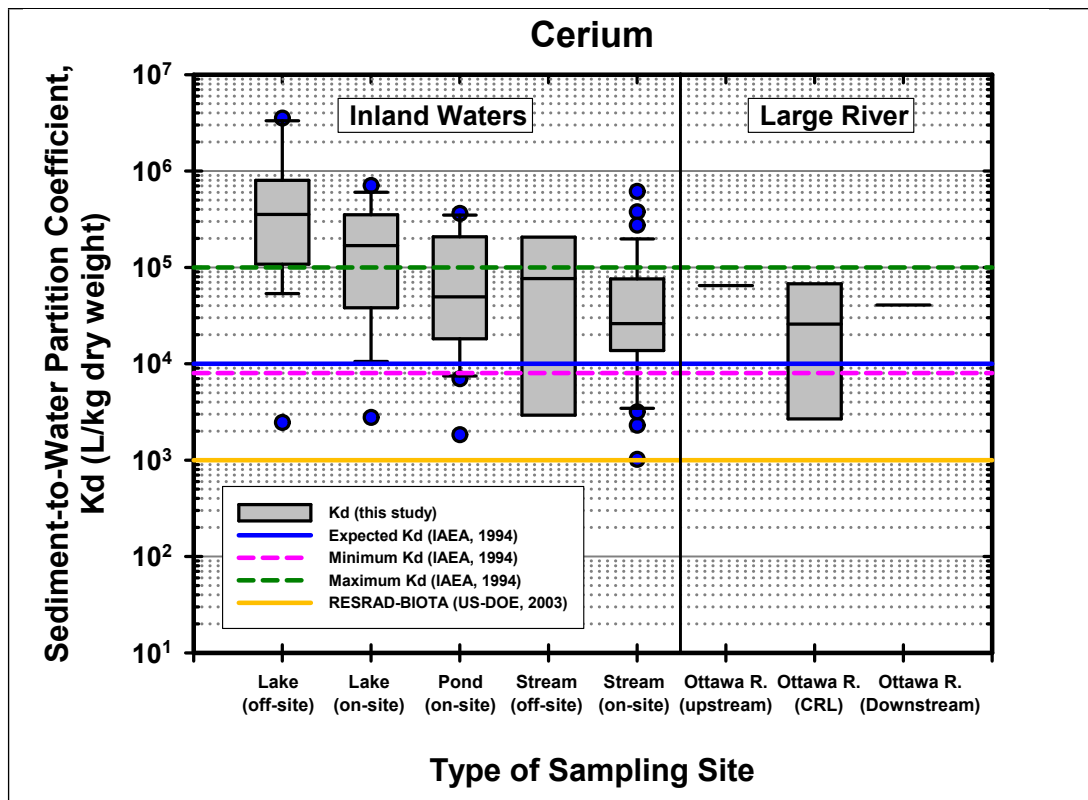


Figure 2: Comparison of Ce  $K_d$  values for different types of water bodies.

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