

Contrasting cesium dynamics in neighboring deep and shallow warm-water reservoirs

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ABSTRACT

To measure the long term retention and seasonal dynamics of an initial 4 kg addition of ¹³³Cs into an 11.4-ha, 157,000 m³ reservoir (Pond 4, near Aiken, South Carolina, USA), the concentrations and inventories of ¹³³Cs in the water column were measured at periodical intervals for 522 days following the 1 August, 1999 release. After rapid declines in concentrations and inventories during the first 90 days, the ¹³³Cs concentrations in the water column declined at an average proportional rate of 0.004 d⁻¹. However, there were periods of less rapid and more rapid rates of declines, and these were correlated with periods of increasing and decreasing K concentrations in the water column. The decline rates were less and the K concentrations greater in the winter than in the summer. In the deeper, neighboring monomictic reservoirs of Par Pond and Pond B, a yearly cycle of increasing and decreasing ¹³⁷Cs concentrations in the water column is driven by anoxic remobilization of Cs from the sediments into a persistent summer hypolimnion. In Pond 4, whose mean depth of 1.6 m is too shallow to support a persistent anoxic hypolimnion, the pattern of yearly dynamics for K and Cs appear to be related to the accumulation and release of these elements from the extensive, seasonal macrophyte communities. The contrasting results between Pond 4 and Pond B suggest that a full appreciation of the relative importance of 1) anoxic remobilization and 2) accumulation and release by macrophytes in these systems remains to be established.

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1. Introduction

Before the Chernobyl accident, many of the detailed analyses of radionuclide behaviors in temperate lake environments were based on studies of weapons fallout (e.g., Preston et al., 1967; Whicker et al., 1972; Alberts and Wahlgren, 1981; Alberts and Muller, 1979; Wan et al., 1987), experimental releases into more controlled lakes (e.g., Kolehmainen et al., 1967; Hakonson and Whicker, 1975; Hesslein et al., 1980; Hesslein, 1987; Bird et al., 1998), or releases from nuclear weapons facilities (e.g., Alberts et al., 1979; Paine and Johnson, 1975; Evans et al., 1983; Voshell et al., 1985; Veska and Tracy, 1986; Clulow et al., 1991). Some of the analyses that used releases from weapons facilities involved the complex of canals, ponds and reservoirs comprising the cooling systems for P and R reactors (Fig. 1A) at the U. S. Department of Energy's Savannah River

Site (SRS) near Aiken, South Carolina, USA. Two of the larger reservoirs in this system, the 1000-ha, 17-m deep Par Pond and the 87-ha, 12-m deep Pond B (Alberts et al., 1987), received >6.3 TBq of ¹³⁷Cs (Ashley and Zeigler, 1980) plus readily detectable amounts of Sr, Pu, Am and Cm isotopes (Alberts et al., 1987; Whicker et al., 1990).

A principal result of the SRS reservoir studies was recognition of the importance of thermal and chemostratification on the remobilization of Cs from sediments into the anoxic hypolimnion of the water column (Alberts et al., 1979; Sholkovitz, 1985). Despite a K_D of 2.8×10^4 for pond sediments (Stephens et al., 1998), production of NH_4^+ in the sandy, kaolinite-dominated sediments during anoxia displaced Cs, K, Ca and Mg from reversible binding sites (Evans et al., 1983; Stephens et al., 1998). Anoxia also resulted in increased iron (Fe) concentrations in the anoxic hypolimnion due to the reduction of Fe(III) to Fe(II). Similar demonstrations of the remobilization of Cs from aquatic sediments by NH_4^+ were reported from other systems, with estimates of the fraction of the Cs inventory being remobilized annually ranging from <1% to 5% (Comans et al., 1989; Dominik and Span, 1992; Davidson et al., 1993; Davison et al., 1993; Smith and Comans, 1996). Although estimates of <1% may seem trivial, perhaps the best measure of remobilization's importance is its

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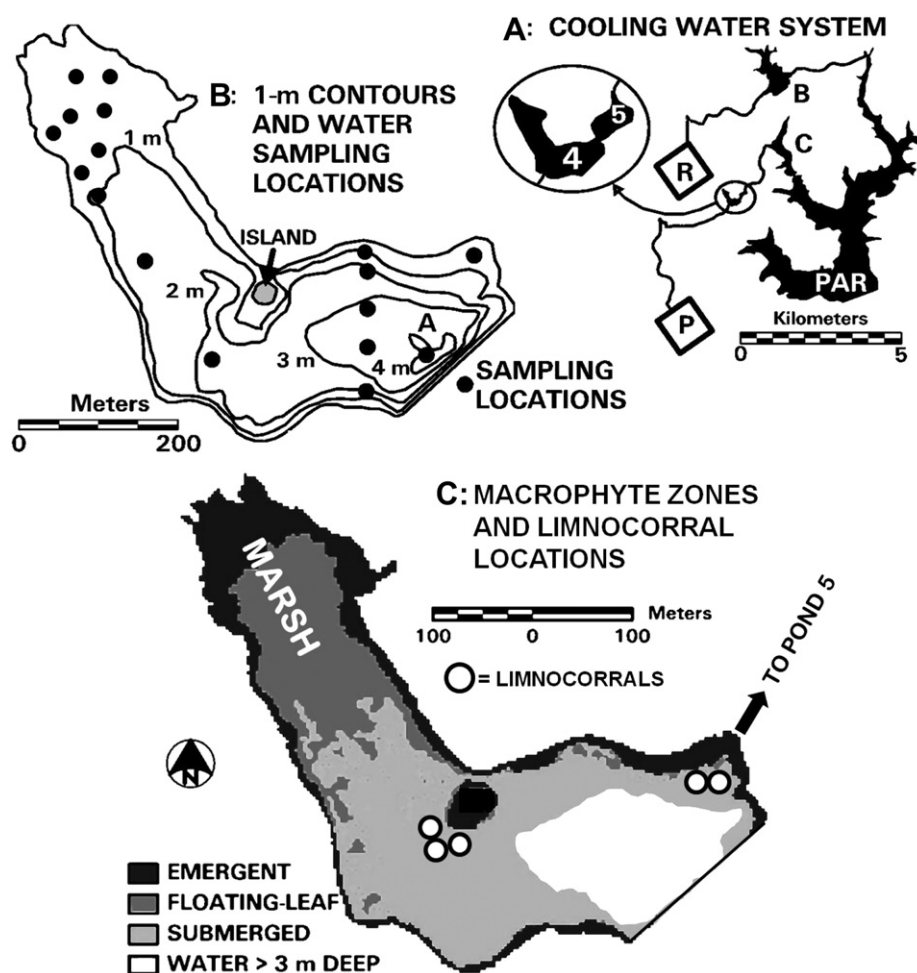


Fig. 1. Maps of Pond 4 including: A) an overview of the canals and ponds of the cooling-water systems for P and R reactors showing the locations of Pond B and Par Pond and Ponds 4 and 5 in an enlarged area; B) the 1-m water depth contours for Pond 4 with water sampling station A and the other 16 water sampling stations; and C) the distribution of emergent, floating-leaf and submerged macrophytes, the locations of the 5 limnocorrals, and the point of water flow from Pond 4 to Pond 5. Limnocorrals are not drawn to scale and only had a diameter of 0.9 m.

effect on Cs concentrations in the water column and subsequently in the biota.

For Par Pond and Pond B, the effect of this remobilization resulted in $1.5\times$ – $3\times$ increases in Cs concentrations in the water column. These increases begin as temperature gradients between surface and deep waters increase to $>15^\circ\text{C}$, and anoxia (defined as O_2 concentrations $<1\text{ mg L}^{-1}$) develops in the hypolimnion during April and May (Chimney et al., 1985; Alberts et al., 1987; Pinder et al., 1992). By June, both reservoirs have anoxic hypolimnions beginning at depths of 6 m. As these anoxic waters develop, ^{137}Cs and Fe concentrations increase. For Pond B, ^{137}Cs concentrations in both the surface waters and the hypolimnion increase by a factor of $2\times$ (Alberts et al., 1979, 1987), and the Fe inventories in the hypolimnion increase by a factor of $>15\times$ (Pinder et al., 1992). The increases in the surface waters reflect mixing across the thermocline, and has been estimated to involve approximately 0.2% of the hypolimnion per day (Hinton and Pinder, 2001). For Par Pond, ^{137}Cs concentrations at 5 m and 15 m are initially similar in late winter, but as anoxia develops concentrations in the hypolimnion at 15-m deep increase by a factor of $3\times$ by September while concentrations at 5 m increase by a factor of approximately $1.5\times$ over the same interval (Alberts et al., 1979). Both ponds are monomictic with a single mixing of surface and deep waters when temperature gradients decline as winter approaches. During this mixing, the Fe

is rapidly precipitated out of the water column (Evans et al., 1983; Pinder et al., 1992), but surface Cs concentrations remain elevated until late winter (Alberts et al., 1979, 1987). A pattern of increases and decreases in concentrations similar to that for ^{137}Cs also occurs for K (Chimney et al., 1985).

Numerous studies of ^{137}Cs in biota were also performed in both Par Pond and Pond B (Domby et al., 1978; Newman and Brisbin, 1990; Whicker et al., 1990; Kelly and Pinder, 1996; McCreedy et al., 1997), and some seasonal patterns in the ^{137}Cs concentrations in biota appeared to be related to the seasonal cycle of concentrations in the water column (Holloman et al., 1997; Peles et al., 2000). Because these studies were conducted >20 years after the main release of ^{137}Cs into the lakes (from 1961 through 1964; Ashley and Zeigler, 1980), they were largely limited to assessments of concentration ratios for systems already at or near steady state. There was limited ability to assess the rate at which the processes of uptake and elimination by the biota were occurring.

Therefore, as part of an effort to measure the short-term kinetics of Cs in southeastern US reservoirs, stable cesium (^{133}Cs) was added to the water column of Pond 4, an 11.4-ha, 4.4-m deep reservoir located on the thermal canal between P reactor and Par Pond (Fig. 1A). Previously reported studies of this experimental Cs addition have documented the foliar absorption of Cs from the

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