



Dissolved and particulate aluminum in the Columbia River and coastal waters of Oregon and Washington: Behavior in near-field and far-field plumes

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ABSTRACT

The distribution of dissolved (soluble and total) and particulate (leachable and total) aluminum was examined in the Columbia River and estuary, in near-field and far-field river plumes, and in adjacent coastal waters of Washington and Oregon during the River Influence on Shelf Ecosystems (RISE) cruise of May/June 2006. Dissolved and particulate aluminum (Al) concentrations were significantly greater in the river than in the coastal waters that mixed to form the plume. Dissolved Al concentrations in the Columbia River (~80 nM) were low relative to other major rivers. Leachable and total particulate Al concentrations within the river reached concentrations greater than 1000 nM and 18,000 nM, respectively. Dissolved Al within the Columbia River estuary showed a significant removal (~60%) at salinities between 0 and 10 with salt-induced flocculation of colloidal Al complexes and enhanced particle scavenging being probable explanations for aluminum removal. Dissolved and particulate Al concentrations were significantly greater in near-field plumes relative to surrounding coastal waters. As the plume advected from near-field to far-field away from the river mouth, dilution of the plume with lower dissolved Al surface waters as well as particle scavenging along the flow path appeared to be controlling dissolved Al distributions. Particle settling as well as dilution with lower particle-load waters led to observed decreases in particulate Al as the plume moved from near-field to far-field. However, the percent-leachable particulate aluminum in both near-field and far-field plumes was remarkably constant at ~7%. Dissolved and particulate Al in a far-field plume over 100 km southwest of the Columbia River mouth were over an order-of-magnitude greater than surrounding waters, illustrating the importance of the Columbia River plume as a mechanism for transporting Al offshore. Aluminum could be used to trace the input of biologically-required elements such as iron into waters off the shelf.

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1. Introduction

The Columbia River, originating some 820 m above sea level in British Columbia, is the largest river entering the northeast Pacific ocean, draining an area of approximately 665,000 km² with an average discharge of 7300 m³ s⁻¹ and annual river discharge of 260 km³ year⁻¹ (Barnes et al., 1972; Jay and Smith, 1990; Thomas and Weatherbee, 2006). A sustained maximum river flow generally occurs in May–June from interior snowmelt while shorter maxima occur during winter from storm events in the coastal basins. Minimum river flow occurs during August–September (Neal, 1972; Thomas and Weatherbee, 2006). During the late summer, the Columbia River accounts for ~90% of the freshwater entering the sea between the Strait of Juan de Fuca and San Francisco Bay (Barnes et al., 1972).

The Columbia River plume forms as coastal seawater intrudes and mixes with river water at or near the mouth of the Columbia River estuary (Barnes et al., 1972). The Columbia River plume is a dominant hydrographic feature of the California Current system off the Washington and Oregon coasts and is observed as a shallow (~2–20 m) surface lens of low-salinity water (Hickey et al., 1998). Because the chemistry of the river water and coastal seawater that mix to form the plume varies depending on time of year, tidal phase, and oceanographic conditions, the chemistry of the plume itself is expected to vary according to these changing conditions (Bruland et al., 2008). The Columbia River plume also plays a key role in the delivery of both macro- and micro-nutrients to offshore waters of Washington and Oregon (Hill and Wheeler, 2002; Lohan and Bruland, 2006; Aguilar-Islas and Bruland, 2006).

The movement of the plume is strongly influenced by seasonal variations in the local wind forcing. When dominant alongshore wind stress is to the south (upwelling conditions), Ekman transport is offshore and surface currents are to the southwest. Therefore, the plume separates from the coast and advects offshore and southward

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from the Columbia River mouth (Hickey et al., 1998). When dominant alongshore wind stress is to the north (downwelling conditions), Ekman transport is onshore and surface currents are to the north. This acts to keep the plume confined tightly along the Washington coast during strong northward wind events and more over the mid-Washington shelf during weaker wind events (Thomas and Weatherbee, 2006). Reversal of the wind forcing can lead to sudden changes between these two modes. Recent studies show that episodic relaxation or reversal of the northerly winds during summer can allow the Columbia River plume to strongly influence the Washington and Oregon coast with plumes moving north and south of the mouth of the Columbia River simultaneously (Garcia Berdeal et al., 2002).

Sources of aluminum (Al) to the world oceans include riverine inputs, atmospheric inputs, and, to a much lesser degree, dissolution from sediments while scavenging onto particle surfaces is regarded as the predominant mechanism for removal of Al. Oceanic levels of dissolved Al are found at trace concentrations, ranging from less than 0.50 nM in intermediate/deep waters of the North Pacific to >25 nM in surface and deep waters of the eastern North Atlantic where high eolian dust input from the Saharan desert is observed (Hydes, 1979; Orians and Bruland, 1986; Kramer et al., 2004; Measures et al., 2008). Higher surface water Al concentrations have been observed in the oligotrophic subtropical gyres of the ocean basins while lower concentrations have been observed in the more productive boundary regions (Orians and Bruland, 1986; Johnson et al., 2003; Measures et al., 2005). Concentrations of dissolved Al in riverine waters are highly variable, ranging from ~50 nM to >1 μM (Hydes and Liss, 1977; Mackin and Aller, 1984a; Morris et al., 1986; Upadhyay and Sen Gupta, 1995; Takayanagi and Gobeil, 2000).

Conomos and Gross (1972) investigated the physical, geochemical, and biological processes governing suspended particulate material in the Columbia River estuary and nearby coastal ocean. Concentrations of riverine suspended particulate material varied from 8 to 40 mg L^{-1} , of which 85–95% consisted of lithogenous particulate material. Assuming 8.2% Al by weight (Taylor, 1964) in crustal material, this equates to greater than 20 μM particulate Al. This suspended lithogenous particulate material consisted largely of feldspar and quartz with the aluminosilicates biotite [$(\text{K}(\text{Mg},\text{Fe})_3\text{Al-Si}_3\text{O}_{10}(\text{OH})_2$] and muscovite [$\text{Al}_2\text{Si}_4\text{O}_{10}(\text{OH})_2$] dominating the larger particle size fractions (Conomos and Gross, 1972). Being that the high suspended particulate load of the river is dominated by aluminosilicates, it follows that the Columbia River is likely a major source of dissolved and particulate Al as well as silicic acid to coastal waters of Washington and Oregon. In contrast, California Current waters are low in both dissolved Al (<1 nM Al; Orians and Bruland, 1986) and silicic acid (~2–4 μM ; Hill and Wheeler, 2002; Aguilar-Islas and Bruland, 2006).

This study provided a unique opportunity to investigate dissolved and particulate Al and silicic acid distributions during both upwelling and downwelling conditions in May/June 2006 in the Columbia River, the river plume, and the adjacent coastal waters of Oregon and Washington. This paper describes the distributions of dissolved and particulate Al and silicic acid both within the Columbia River and estuary as well as in near- and far-field plumes both north and south of the Columbia River. The behavior of Al and silicic acid in river–ocean mixing within the estuary and during advection of the plume away from the river mouth is also discussed.

2. Methods

2.1. Sample collection and filtration

Seawater samples were collected aboard the R/V *Wecoma* off the Oregon and Washington coasts from May 21, 2006 to June 21,

2006 during the RISE-4W cruise, the last of five cruises associated with the RISE (River Influence on Shelf Ecosystems) program (see Bruland et al., 2008, for more detail). Surface (~1 m) sampling was conducted using an underway clean surface pump “fish” system described in detail elsewhere (Bruland et al., 2005; Aguilar-Islas and Bruland, 2006; Lohan and Bruland, 2006). Briefly, this system utilizes an all PTFE Teflon™ diaphragm pump and PFA Teflon™ tubing mounted to a PVC depressor vane 1 m above a 20-kg PVC fish, allowing for clean surface water sampling while underway at speeds of 4–7 knots. Concomitant underway surface salinity and temperature measurements were obtained using a YSI CTD Sonde attached to the PVC fish. Underway fluorescence and turbidity data were obtained through the ship’s flow-through seawater system. Vertical profiles down to approximately 20 meters were also collected with the fish system while deeper samples were collected using 30-L GoFlo bottles (General Oceanics) deployed on Kevlar line (Bruland et al., 1979). Samples for dissolved Al collected from the “fish” system were filtered in-line through acid-cleaned 0.45 μm Teflon™ capsule filters (GE Osmonics Capsule filters) while dissolved Al samples collected from the Go-Flo’s were filtered through acid-cleaned 0.4 μm Nuclepore polycarbonate filters.

For particulate Al samples, an unfiltered water sample was collected from the surface fish system or 30-L GoFlo bottle directly into a 2-L acid-cleaned low-density polyethylene (LDPE) bottle. All unfiltered samples were kept cold and dark prior to filtration, which occurred within a few hours after sample collection. Unfiltered water samples were filtered under trace metal clean conditions in a class 100 clean bench using an in-line filtration apparatus as described in Berger et al. (2008). Briefly, unfiltered samples passed through acid-cleaned 47 mm, 10 μm filters followed by 47 mm, 0.4 μm filters (Nuclepore™ polycarbonate track-etched membrane filters) mounted in polypropylene filter sandwiches (Millipore™). This filtration technique captured 0.4–10 μm and >10 μm particulate material utilized in both leachable particulate Al and refractory particulate Al analyses. Filters were folded into eighths and placed in acid-cleaned 2 mL high density polyethylene (HDPE) vials and stored frozen. Total sample volume filtered ranged from 225 mL to 2095 mL. Filter blanks were processed as described above with the exception that no sample passed through the filters.

Soluble (<0.03 μm) Al samples were collected by passing an unfiltered sample first through a 47 mm, 10 μm filter as mentioned above followed by an in-line acid-cleaned 0.03 μm (200 kDa) polyethylene hollow fiber flow-through filter (Sterapore™, Mitsubishi-Rayon, Tokyo, Japan). More detail for this procedure is given in Hurst and Bruland (2007) while protocols for filter maintenance are outlined in Nishioka et al. (2001).

2.2. Analytical methods

Macronutrients (nitrate + nitrite, [referred to herein as nitrate], phosphate, and silicic acid) were determined underway directly from the outflow of the fish system. Measurements were taken every two minutes on a Lachat QuikChem 8000 Flow Injection Analysis system using standard colorimetric methods (Parsons et al., 1984).

Filtered samples for dissolved and soluble Al analyses were acidified on-board ship to pH 1.7–1.8 using sub-boiled quartz distilled 6 N hydrochloric acid (4 mL 6 N HCl per L of sample, ~0.024 M HCl added to the sample). Dissolved and soluble Al were determined at the University of California Santa Cruz (UCSC) under trace metal clean conditions in a class 100 clean space using the flow injection method of Brown and Bruland (2008) which utilizes the chelation of dissolved and/or soluble Al onto a commercially-available iminodiacetate (IDA) resin. Depending on the Al concentration, samples were preconcentrated onto the IDA resin for 30 s to

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