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Colloidal organic matter from wastewater treatment plant effluents: Characterization and role in metal distribution

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ABSTRACT

Colloidal organic matter from wastewater treatment plants was characterized and examined with respect to its role in metal distribution by using tangential flow ultrafiltration, liquid chromatography coupled with organic carbon and UV detectors, and an asymmetrical flow field-flow fractionation (AFlFFF) multidetection platform. Results revealed that a humic-like fraction of low aromaticity with an average molar mass ranging from 1600 to 2600 Da was the main colloidal component. High molar mass fractions (HMM), with molar mass ranges between 20 and 200 kDa, were present in lower proportions. Ag, Cd, Cu, Cr, Mn and Zn were found mainly in the dissolved phase (<0.45 μm) and their distribution between colloidal and truly dissolved fractions was strongly influenced by the distribution of dissolved organic carbon. AFlFFF coupled to ICP-MS showed that Ag, Cd, Cu, Cr, Mn and Zn associate to the low molar mass fraction of the colloidal pool, whereas Al, Fe and Pb were equally bound to low and high molar mass fractions.

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1. Introduction

Colloids are macromolecules or assemblages, with sizes operationally defined as ranging from 1 nm to 1 μm (Lead and Wilkinson, 2006). The colloidal pool in natural systems consists of a heterogeneous mixture of polydisperse inorganic and organic components. Major inorganic colloids are often dominated by iron oxides, whereas the colloidal organic matter (COM) is mainly composed of humic substances (HS) and extracellular polymeric substances. Colloidal stability is controlled by coagulation and flocculation processes, depending on the size, the shape and the relative concentration of colloidal components (Buffle et al., 1998).Within the dissolved phase, colloids can be transported through natural waters and for long distances, as is the case for the truly dissolved species (Gustafsson and Gschwend, 1997). COM is highly reactive regarding metal complexation due to its polyfunctional character and large binding site density. Consequently, COM is recognized to control metal distribution, speciation and bioavailability in freshwater systems (Warren and Haack, 2001; Town and Filella, 2002) and has been shown to be important in processes such as metal mobilization and

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Nomenclature

COM	colloidal organic matter
C_{diss}	organic carbon concentration in the dissolved (0.45 μm filterable) fraction
C_{isol}	organic carbon concentration in the colloidal isolates
C_{td}	organic carbon concentration in the truly dissolved (1 kDa filterable) fraction
C_{col}	organic carbon concentration in the colloidal fraction (between 1 kDa and 0.45 μm)
EfCOM	effluent colloidal organic matter
EfOM	effluent organic matter
HMM	high molar mass fraction
LMM	low molar mass fraction
$[M]_{diss}$	total dissolved metal (0.45 μm filterable) concentration
$[M]_{td}$	truly dissolved metal concentration (1 kDa filterable)
$[M]_{isol}$	metal concentration in the isolate
$[M]_{col}$	metal concentration in the colloidal fraction (between 1 kDa and 0.45 μm)
$[M]_{p}$	metal concentrations in the particulate phase
$[M]_{raw}$	metal concentration of the raw samples
M_p	molar mass corresponding to the peak maximum
M_n	number-average molar mass
M_w	weight-average molar mass

dispersion (Weng et al., 2002; Schafer et al., 2003; Ren and Packman, 2004, 2005; Pedrot et al., 2008).

COM behavior depends on its nature (Buffle et al., 1998). In the case of humic substances both aromaticity and metal binding capacity have been shown to depend on molar mass or size fractions (Cabaniss et al., 2000). Recent technological advances, with the coupling of size fractionation techniques such as size-exclusion chromatography (SEC) or flow field-flow fractionation (FlFFF) to highly sensitive metal detectors such as inductively coupled plasma-mass spectrometry (ICP-MS), have confirmed the differences in the molar mass and size distribution of HS–metal complexes (Hassellov et al., 1999; Schmitt et al., 2001; Wu et al., 2004; Bolea et al., 2006; Dubascoux et al., 2008). In freshwater systems, evidence for the preferential binding of some metals with inorganic colloids of larger size, mainly composed of iron or manganese with a low carbon content, suggests that both HS and mineral oxides of colloidal size could be potential carriers for trace metal dispersion (Lyven et al., 2003; Stolpe et al., 2005; Baalousha et al., 2006; Stolpe and Hassellov, 2007).

Although wastewater treatment plant (WWTP) effluents discharged in surface waters rise in volume continuously and are considered as one of major sources of organic matter in urbanized areas (Dignac et al., 2000; Imai et al., 2002; Shon et al., 2006a), little is known about the influence of wastewater treatment plant COM on the distribution and dispersion of metals. In contrast to the considerable work on the characterization of the composition of organic matter from effluents (EfOM), which is important for wastewater treatment optimization (Dignac et al., 2000; Shon et al., 2004, 2006b), its role in the distribution of metals in effluents, and in their potential transfer through the receiving ecosystem, is not well understood.

The present study therefore aims to improve our understanding of the role of the colloidal organic matter from wastewater treatment plant effluents in trace metal transfer and distribution. More specifically, it has an emphasis on the characterization of the composition, chemical properties and molar mass distribution of the effluent colloidal organic matter (EfCOM) isolates and associated metals. Results obtained for three Swiss municipal WWTP effluents are compared with those found for Lake Geneva water or standard fulvic and humic acids. The influence of chemical properties and mass distributions of EfCOM on metal transport, and the implications for the receiving waters, are discussed.

2. Materials and methods

2.1. Sampling and isolation of colloidal components

The effluents of Duebendorf (DB), Hinwil (HW) and Zurich (ZH) wastewater treatment plants were sampled in February 2007 and January 2008. The treatment processes of each WWTP include gravitational sedimentation and activated sludge treatments, followed by chemical treatment with iron addition for phosphate removal. Samples were collected in 20 L plastic bottles that had previously been washed with 0.1 M suprapure HNO_3 (Baker) and rinsed with MilliQ water to avoid metal contamination. Samples were transported to the laboratory and filtered through 0.45 μm pore size cellulose acetate membrane (Millipore). A tangential flow ultrafiltration system (TFF; Pellicon, Millipore), holding a regenerated cellulose membrane with 1 kDa nominal molar mass cut-off was used to concentrate the colloidal matter of each sample, in a sampling mode, in which the retentate flow was directed back to the feed bottle. The permeate flow was directed to a container to be collected. The colloidal isolate was obtained after 5–7 h of circulation, which corresponded to a volumic concentration factor (cf) varying between 12.6 and 20. The colloidal fraction from Lake Geneva water (Vidy Bay (VD) near Lausanne) was isolated in January 2008 using the same procedure as for WWTP effluents. Vidy Bay is known to be impacted by the main WWTP of Lausanne (Pote et al., 2008) especially near the outlet that is located at 127 m offshore at 30 m below the surface. Standard humic and fulvic acids from Suwannee River (SRFA and SRHA), used for comparative purposes, were obtained from the International Humic Substances Society (St. Paul, MN).

2.2. Organic carbon and metals distributions

Organic carbon (OC) concentrations were measured, using a Shimadzu TOC-V analyzer after filtration through 0.45 μm pore size membranes (dissolved, C_{diss}), in the colloidal isolate (C_{isol}) and in the TFF permeate (truly dissolved, C_{td}). The colloidal organic carbon content in the collected water samples was estimated by Eq. (1):

$$C_{col} = (C_{isol} - C_{td})/cf \quad (1)$$

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