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Diagnosis of the transport of adsorbed material in the Scheldt estuary: A proof of concept

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ABSTRACT

Many contaminants can attach to suspended particles. Their transport differs therefore from the transport of dissolved substances, especially in highly turbid environment like estuaries. In this paper, we show how the Constituent Age and Residence time Theory (CART — www.climate.be/CART) can be adapted to quantify in a rigorous manner the transport rate of contaminants that are present in both the dissolved and adsorbed phases.

On the basis of numerical experiment using a 1D model of the Scheldt estuary, it is shown that the interaction with suspended particles significantly affects the transport of contaminants with partition coefficients larger than 10^3 ml/g. The mean transit time from Ghent to Vlissingen of such contaminants can reach 160 days while it is only 60 days for water and dissolved constituents. This increase of the transit time is mainly due to the fact that adsorbed constituents spend long periods of time on the bottom. Surprisingly, the downstream transport of adsorbed constituents in the water column appears more effective than that of dissolved constituents. This transport affects however a small fraction of the adsorbed constituent and is therefore not sufficient to compensate for the long resting phase on the bottom of the bulk of the constituent.

The concept and methodology introduced in this paper are easily applicable to most model studies and provide powerful and flexible tools for the detailed understanding of the transport of contaminants in estuaries. In particular, the concept of age and modified ages taking into account specifically the time spent in the water column or in the bottom provide new diagnostic tools to understand and quantify the dynamics of contaminants.

1. Introduction

The transport of pollutants in coastal areas and estuaries is a complex process, not only because of the non linear hydrodynamics of such regions, but also because of the variable propensity of many such contaminants to attach to suspended particles.

Heavy metals are known to associate easily with particulates. Baeyens et al. (1998b) report for instance that, because of the high turbidity in the Scheldt estuary, less than 3% of the total Pb burden is present in the dissolved phase. The interaction with suspended particles is also known to play a major part in the regulation of the transport or accumulation of mercury in estuaries (e.g. Lawson et al., 2001; Pato et al., 2010). Similar effects are significant for the other heavy metals, like zinc, that have a large partition coefficient in marine waters (e.g. Balls, 1989; Turner, 1996; Valenta et al., 1986).

The transport of many other contaminants, like many polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs), currently monitored by most environment protection programs (e.g. OSPAR, 2010), is also linked to the dynamics of sediments (e.g. Smith et al., 2009).

The dynamics of some nutrients depends also on that of suspended particles. Many ecosystem models take therefore into account the adsorption of ammonium and phosphate on suspended solids. In addition, detrital particulate organic matter is produced that tends to sink through the water column and is transported in much the same way as sediments and contaminants adsorbed on suspended particles (e.g. Arndt et al., 2011; Gregoire and Beckers, 2004; Lancelot et al., 1987).

Consequently, the transport rates and even the transport routes of many contaminants are likely to differ significantly from the general movement of the water masses. A strong and complex control by the processes responsible for the dynamics of sediments (tidal variations, sedimentation/erosion, river flow, waves, stratification,…) is therefore expected, in addition to the many environmental and chemical parameters (pH, temperature, salinity, composition of suspended matter,…) influencing the process of adsorption/desorption (e.g. Gao et al., 2009; Liu and Lee, 2007; Tappin et al., 2010; Tremblay et al., 2005; Xu and Li, 2009).

Obviously, the settling of suspended particles is the main physical mechanism at the origin of the difference between the transport rates

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of the tracers in dissolved and particulate forms. If turbulence is not strong enough to mix the water column, suspended particles tend to accumulate in the bottom layer and their transport depends therefore mainly on bottom currents (and the interaction with waves). In tidal estuaries, the dynamics is dominated by strong variations of turbulence, oscillating currents and recurrent episodes of settling on the bottom and resuspension. The net movement of adsorbed contaminants depends therefore on the particular sampling of the oscillating velocity field experienced by suspended particles during their vertical movements in the water column, taking into account a possible resting phase on the bottom. Tidal asymmetry (Son and Hsu, 2011; Uncles, 1981; Uncles and Jordan, 1980) and the time-lag effect, associated with the competitive effects of settling and mixing (Christie et al., 1999; de Swart and Zimmerman, 2009), influence therefore the net transport in a complex manner.

The resulting delay of the transport of adsorbed constituents was demonstrated in previous studies on the dynamics of radioactive pollutants in the English Channel. Boust (1999) estimated a transport timescale of particle-bound radionuclides by comparing the isotopic ratio of artificial radionuclides collected in the bottom sediments to the time evolution of the isotopic ratio of the release at the nuclear fuel reprocessing plant of La Hague. They reported apparent transit times of 10–30 years for the Western Channel and of 10–15 years for the Eastern Channel, which are much larger than the correspond timescales for the dissolved constituents estimated to a few months only by Salomon et al. (1995). Similar results are obtained by Perianez and Miro (2009) using a numerical model of the English Channel. These authors demonstrate that the transport time scales increase when the interaction with the solid phase is taken into account; while the transit time from La Hague to Dover is about 70 days for a conservative radionuclide, the transit time of ^{137}Cs is about 2.7 years while that of ^{239,240}Pu reaches 65 years.

The previous studies aiming at the quantification of the transport rate of adsorbed constituents rely on the analysis of the correlation between the time series of the concentration of the target pollutant at the source and at the observation point where the transit time is estimated. The method basically ignores the diffusion process and introduces a systematic bias (Delhez and Deleersnijder, 2008). It is also not appropriate to capture temporal variations of the transit time. In the following, we show how the concept of age can be used to diagnose and quantify in a rigorous way the transport rate of such contaminants.

The age of a particle is defined as the time elapsed since a given origin that can be regarded as the 'birth' of the particle (Bolin and Rhode, 1973; Delhez et al., 1999; Monsen et al., 2002; Takeoka, 1984; Zimmerman, 1976). With an appropriate definition of this time origin, the age can be used to quantify the ventilation rate of the ocean (e.g. Bendtsen et al., 2009; England, 1995; Holzer and Hall, 2000), the transport in the atmosphere (e.g. Hall and Plumb, 1994), the horizontal transport of dissolved contaminants (Deleersnijder et al., 2001; Delhez and Deleersnijder, 2002; Orre et al., 2008; Shen and Lin, 2006), the renewal rate of water masses (e.g. de Brye et al., 2013-this issue; Gourgue et al., 2007), the fluxes of nutrients in an ecosystem model (Delhez et al., 2004b),…. The age of a dissolved constituent can be easily computed using the Constituent Age and Residence time Theory (CART — `www.climate.be/CART`), which is well suited to mathematical or numerical models (Deleersnijder et al., 2001; Delhez and Deleersnijder, 2002; Delhez et al., 1999, 2004a). The approach requires only the resolution of an evolution equation for the so-called 'age concentration'. Since the equation for the age concentration is basically an advection–diffusion equation with a source term introducing a coupling with the concentration of the tracer and accounting for its aging, it is easily implemented in existing numerical models.

As a first extension to CART, Mercier and Delhez (2007) computed different characteristic timescales associated with the dynamics of suspended matter in the Belgian coastal zone. They quantified the horizontal transport of suspended matter using a 'transport age'. Complementarily, they also defined a 'resuspension age' to assess the time spent by particles in the water column after their erosion from the bottom. Similar transport ages were also computed by Gong and Shen (2010).

In this paper, we propose a further extension of CART in order to quantify the transport rate of contaminants that are adsorbed on suspended particles. While results of the application of the method to Scheldt estuary are presented, the main objective of this manuscript is to demonstrate the feasibility of the approach and its applicability to more realistic models and case studies.

This paper is organized as follows. Section 2 is devoted to the description of the Scheldt estuary and its hydrodynamic modeling. The modules for the dynamics of suspended particulate matter (SPM) and contaminants are described in Section 3. The developments of CART are explained in Section 4. The information obtained by application of CART is discussed in Section 5. Some concluding remarks are then elaborated.

2. Hydrodynamics of the Scheldt estuary and its 1D modeling

The Scheldt estuary is located at the border between Belgium and The Netherlands (Fig. 1). The Scheldt river has a catchment basin of 22 10^3 km^2 and flows through highly industrialized and densely populated areas. As a result, the estuary receives large inputs of contaminants.

The Scheldt estuary is macrotidal with a tidal range of about 3.8 m at its mouth (Vlissingen). The tidal range remains large up to Ghent (2 m), some 160 km upstream. A system of locks in the vicinity of Ghent stops the propagation of the tidal signal upstream. Thanks to the strong tidal currents and its mean depth of about 10 m, the estuary is well mixed with a small vertical stratification appearing only occasionally in the neighborhood of Antwerp. Taking into account the lateral input from tributaries (Dender, Durme and Ruppel, including Nete, Zenne and Dijle), the total mean fresh-water discharge in the estuary is of the order of 100 m^3/s but with marked seasonal variations (Fig. 2); the average fresh water input reaches 200 m^3/s in winter but decreases to 70 m^3/s or less in summer. Together with the strong tidal signal, these variations form the predominant factors determining the hydrodynamics of the Scheldt estuary (Baeyens et al., 1998a).

For the purpose of this study, we use a one-dimensional model of the tidal part of the Scheldt river and of its main tributaries with a spatial resolution of 2 km. The cross-section and wetted perimeter of each river segment are interpolated from tabulated data as a function of the water level (Laforce et al., 1977).

The hydrodynamic model is based on the continuity and momentum equations integrated overt the cross-section of the river. In this approach, baroclinic processes are neglected as well as the curvature of the river channels. These parameters have however only a small influence in the Scheldt, as suggested by the results obtained in previous studies using the same model approach (De Smedt et al., 1998; Regnier et al., 1997).

The hydrodynamic model is forced at Vlissingen with observed water surface elevation extracted from the DONAR database maintained by the Dutch Ministry of Infrastructure and the Environment (Rijkswaterstaat — Ministerie van Infrastructuut en Milieu) and made available through the WATERBASE online application (http://live.waterbase.nl). At the upstream boundaries, where tidal effects are negligible, the model uses daily mean river discharges obtained from Flanders Hydraulics Research.

The hydrodynamic model has been validated against observed water surface elevations at various stations along the estuary. It provides a reliable description of the propagation and dissipation of the tidal signal in the estuary (Gypens et al., 2013-this issue).