



Evaluation of wastewater contaminant transport in surface waters using verified Lagrangian sampling



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HIGHLIGHTS

- Studies evaluating contaminant fate must account for variable effluent discharge.
- Sampling the Lagrangian parcel links hydrologic and chemical processes.
- Small deviations from the Lagrangian parcel result in large concentration changes.
- Concentrations can be corrected to reflect the true Lagrangian parcel.
- Inorganic data are critical for providing information to interpret results.

ARTICLE INFO

Article history:

Received 17 July 2013

Received in revised form 26 September 2013

Accepted 27 September 2013

Available online 26 October 2013

Editor: Eddy Y. Zeng

Keywords:

Lagrangian sampling

Water quality

Conservative elements

Effluent discharge

Normalization

ABSTRACT

Contaminants released from wastewater treatment plants can persist in surface waters for substantial distances. Much research has gone into evaluating the fate and transport of these contaminants, but this work has often assumed constant flow from wastewater treatment plants. However, effluent discharge commonly varies widely over a 24-hour period, and this variation controls contaminant loading and can profoundly influence interpretations of environmental data. We show that methodologies relying on the normalization of downstream data to conservative elements can give spurious results, and should not be used unless it can be verified that the same parcel of water was sampled. Lagrangian sampling, which in theory samples the same water parcel as it moves downstream (the Lagrangian parcel), links hydrologic and chemical transformation processes so that the in-stream fate of wastewater contaminants can be quantitatively evaluated. However, precise Lagrangian sampling is difficult, and small deviations – such as missing the Lagrangian parcel by less than 1 h – can cause large differences in measured concentrations of all dissolved compounds at downstream sites, leading to erroneous conclusions regarding in-stream processes controlling the fate and transport of wastewater contaminants. Therefore, we have developed a method termed “verified Lagrangian” sampling, which can be used to determine if the Lagrangian parcel was actually sampled, and if it was not, a means for correcting the data to reflect the concentrations which would have been obtained had the Lagrangian parcel been sampled. To apply the method, it is necessary to have concentration data for a number of conservative constituents from the upstream, effluent, and downstream sites, along with upstream and effluent concentrations that are constant over the short-term (typically 2–4 h). These corrections can subsequently be applied to all data, including non-conservative constituents. Finally, we show how data from other studies can be corrected.

Published by Elsevier B.V.

1. Introduction

The discharge of wastewater treatment plant (WWTP) effluent into surface waters has long been recognized to have important implications for aquatic ecosystems and downstream water users due to the presence of organic carbon, nutrients, and contaminants (Streeter and Phelps, 1925; Odum, 1956). Although the connections between wastewater

contaminants, drinking water supplies, and aquatic ecosystems have been demonstrated (Focazio et al., 2008; Benotti et al., 2008; Buerge et al., 2009; Purdom et al., 1994; Jobling et al., 1998; Vajda et al., 2008), understanding of the processes that control the fate and transport of these compounds remains incomplete. This is due in part to the fact that the majority of research evaluating the environmental fate of these compounds is based upon laboratory studies, and few studies have evaluated in-stream processes in natural environments (in-situ). While the inherent difficulties involved in conducting in-situ research are substantial, this research is critical because laboratory studies cannot anticipate all of the interacting factors controlling the fates of WWTP contaminants.

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These factors include dynamic changes in wastewater effluent discharge and chemistry (e.g., Coutu et al., 2013) as well as variation in physical characteristics of the receiving waters (such as bed texture, water velocity, and hyporheic exchange); biotic characteristics (such as changing microbial populations and respiration rates); and spatial and temporal variations in chemical characteristics (caused by water sources and sinks, suspended sediment loads, and interactions between all of the above factors). Importantly, in-situ studies can serve to inform and guide the direction which laboratory studies take and verify the models which these studies produce.

In-situ research has generally attempted to account for the myriad concurrent factors which can influence the fate of WWTP contaminants using three main techniques:

- (1) Normalization of concentrations of the targeted compounds to concentrations of intrinsic “conservative tracers” such as boron (Labadie and Budzinski, 2005), bromide (Morrall et al., 2004), and chloride (Barber et al., 2011). The principal idea behind this technique is that changes in normalized concentrations reflect in-stream reactions of the targeted compounds. Although this technique is generally easy to implement, one of its principal disadvantages concerns the effect of changing effluent discharge on normalization, as will be discussed below.
- (2) The use of introduced reactive tracers (Bohlke et al., 2004; Kunkel and Radke, 2011; Writer et al., 2012). The advantage of this technique is that because a known amount of the tracer has been added, its loss or gain within the stream can be directly quantified. However, without substantial knowledge of the hydrologic and chemical characteristics of the stream, the causes of the loss or gain cannot generally be determined. Additionally, political, ethical, and regulatory considerations frequently inhibit this type of study.
- (3) Lagrangian sampling, or sampling the same parcel of water (the Lagrangian parcel) as it moves downstream (Meade and Stevens, 1990; Schultz et al., 2010; Barber et al., 2011, 2013; Writer et al., 2012, 2013). Ideally, this method provides definitive information regarding the in-stream reactions of targeted compounds. However, as will be discussed below, it is critical to verify whether the Lagrangian parcel has been sampled.

All three of these techniques suffer from a lack of information regarding the influence of unmeasured water sources. We put forth in this paper a method which we call “verified Lagrangian” sampling that allows one to precisely quantify and correct for sampling that may precede or follow the Lagrangian parcel. The importance and utility of a properly designed Lagrangian sampling regime will be demonstrated, and a mechanism for correcting data from samples which were not precisely from the Lagrangian parcel is developed. Finally, the advantages of a sampling scheme which includes a wide variety of inorganic compounds will be established. This information will be useful for future research evaluating the fate and transport of wastewater contaminants.

2. Methods

2.1. Site location

The study was conducted on May 4, 2011 on a 5.6-km reach of Boulder Creek, Colorado, beginning just upstream of the City of Boulder WWTP effluent channel (a site map is available in the Supporting Information, SI). Sampling sites included the WWTP effluent (designated hereafter as “Eff”), Boulder Creek upstream from the effluent channel (“US”), and four sites on Boulder Creek downstream of the WWTP (“BC1”–“BC4”). Two small ditches and a small tributary (Dry Creek), representing the only surface water additions to Boulder Creek observed in the study reach, were also sampled. One small diversion, located immediately upstream of the WWTP effluent outfall, was

removing about 0.057 m³/s from the creek during the study; no other diversions were active.

2.2. Lagrangian tracer study and water quality sampling

In order to determine stream velocity and estimate the time that each site should be sampled to capture the Lagrangian parcel as it moved downstream, a preliminary tracer study was conducted on May 3, 2011 precisely 24 h before the actual study. Rhodamine WT dye was injected into the WWTP effluent channel, and dye concentrations were monitored at the four downstream sites with a self-contained underwater fluorescence apparatus (SCUFA) at intervals of 1 min or less. These were then used to estimate hydraulic transit times to the downstream sites.

The following day (May 4, 2011 at 09:00), under similar hydrologic conditions, both Rhodamine WT dye and sodium bromide were added to the effluent channel. SCUFAs were again employed to measure dye concentrations. Water samples were collected at each downstream site with an autosampler at 8- to 10-minute intervals over a time period bracketing the pulse of the added tracer; these samples were collected for the analysis of bromide and other inorganic constituents.

Two different types of water samples were collected at each downstream site: the autosampler samples, which were collected from a single point within the stream (“grab samples”); and field-duplicate depth- and width-integrated composite water samples at times intended to straddle the bromide tracer pulse and thus capture the Lagrangian parcel. Composite samples are generally regarded as representative of the entire stream at the time and place they are collected, and in unmixed waters may have different chemistries than grab samples (e.g., Moody and Meade, 1993). Composite samples were collected over a period of 1 to 2.5 h. Effluent water samples were collected over a two-day period surrounding the tracer study using an autosampler at two-hour intervals in order to evaluate how effluent water chemistry changed over time.

Stage height was recorded every 15 min at US (the upstream site) at U.S. Geological Survey (USGS) streamgaging station 06730200 (NWIS, 2013). Stage height was also recorded every 5 min (with a pressure transducer) in the effluent outfall (Eff) and the furthest site downstream (BC4). The City of Boulder provided influent discharge values to the WWTP at 5-minute intervals for the period surrounding the study. Discharge was measured at each downstream site using a wading rod and pygmy meter, and each measurement occurred over a period of 30 to 35 min.

2.3. Analytical

Details of the processing and analytical procedures for the composite and autosampler samples can be found in the SI. Bromide from each autosampler sample was analyzed in triplicate. Concentrations of many inorganic and organic species were determined, but for the purposes of this paper, we focus on B, Cl, and Gd, with reference to a few others. B and Gd were analyzed in triplicate and Cl in duplicate.

Bromide tracer recovery was determined by integrating the area underneath the tracer curve (after correcting for background bromide concentrations) and multiplying this value by the idealized discharge directly below the effluent channel (Q_A , defined as the sum of the discharges of the upstream site and the effluent channel). This is described in more detail in the SI.

2.4. Theoretical

To evaluate the impacts of variable effluent discharge on downstream concentrations, a series of equations were developed to describe the behavior of conservative inorganic elements (e.g. B, Cl, Gd); these

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