



Use of lithium tracers to quantify drilling fluid contamination for groundwater monitoring in Southeast Asia



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ABSTRACT

Drilling is widely used in groundwater monitoring and many other applications but has the inherent problem of introducing some degree of external contamination into the natural systems being monitored. Contamination from drilling fluid is particularly problematic for (i) wells with relatively low water flow rates which are difficult to flush; and for (ii) hydrogeochemical research studies of groundwaters hosted by incompletely consolidated shallow sediments, which are widely utilized as sources of drinking water and irrigation water across many parts of Asia. Here, we develop and evaluate a method that can be simply used to quantify the extent of drilling fluid contribution to a water sample either to optimize sample collection for reduced contamination, or to allow a correction for contamination to be made. We report the utility of lithium chloride tracers using both field and laboratory analytical techniques to quantitatively evaluate and correct for drilling fluid contamination of casing waters through an investigation of 15 sites in Kandal Province, Cambodia. High analytical errors limit the practicality and resolution of field-based lithium ion selective electrode measurements for purposes other than broad estimates of gross contamination. However, when laboratory analysis is integrated with the method (e.g. via inductively coupled plasma atomic emission spectrometry analysis), lithium tracers can provide a robust and accurate method for evaluating drilling-related contamination if appropriate samples are collected. Casing water is susceptible to contamination from drilling fluid which was shown to be significantly reduced within two to three well volumes of flushing but can still persist above background for greater than seven well volumes of flushing. A waiting period after drilling and prior to water sampling was shown to further decrease contamination due to dilution from the surrounding aquifer, particularly in more permeable wells. Contamination values were generally <3% for 34 monitoring wells across 15 sites after flushing a mean of 4.6 ± 3.8 well volumes, even when lithium-spiked water was directly injected during flushing to remove settled mud/debris. Operational issues can be encountered which can (i) lead to contamination being much higher than the mean if wells are highly unproductive and clay-dominated or (ii) lead to higher flushing volumes than the mean particularly in sandy areas where fine sand may enter the well screening. General correction factors have been provided for typical monitoring wells in poorly consolidated shallow aquifers in Southeast Asia, and examples provided for how to correct other groundwater data for contamination. For most analytes such as sodium or dissolved organic carbon (DOC), specific corrections may not be necessary for the typical magnitude of contamination encountered, particularly when the differences in concentrations between the drilling fluid and groundwater are relatively small. In the particular circumstance where drilling fluid may have much higher DOC than groundwaters, or vice versa with drilling fluid having much lower DOC than groundwaters in organic-rich alluvial sediments, corrections may still be necessary and significant. Similarly, for highly sensitive parameters such as ^{14}C model age or other age-related parameters (such as tritium, chlorofluorocarbons (CFCs) or sulfur hexafluoride (SF_6)), corrections can be significant in typical field scenarios particularly when contamination values are high and/or there is a large difference in age between groundwater and drilling fluid. The lithium method was verified with comparison to changes in concentration of a suite of representative and naturally occurring groundwater constituents as a function

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of well flushing from relatively low and high permeability groundwater monitoring wells to further illustrate the technique.

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1. Introduction

Drilling procedures have the inherent and unavoidable problem of introducing some degree of external contamination into natural groundwater systems. The quantification of contamination resulting from drilling procedures may therefore be essential information required for the scientific interpretation of chemical data from groundwaters obtained from drilled monitoring wells. Chemical tracers can be used to track the distribution of drilling fluid during well drilling (Bath, 2011) and which directly allow for monitoring and quantification of drilling-related contamination in groundwaters.

Drilling tracers have important applications in a number of fields including groundwater investigations (Bottrell et al., 2010; Edmunds and Bath, 1976; Riley et al., 2001; Smart and Laidlaw, 1977), oil and gas exploration (Kleven et al., 1996; Warner et al., 2014), geothermal energy surveys (Adams et al., 1992; Chrysikopoulos and Kruger, 1987) and other geoscientific (McKinley and Colwell, 1996; Smith et al., 2000) and microbiological (Kallmeyer et al., 2006) studies. This is also essential in the context of research into the biogeochemistry of arsenic in shallow reducing aquifers (Charlet and Polya, 2006; Lloyd and Oremland, 2006; Polya and Charlet, 2009), where the quantification of possible introduction of surface water into sediments and groundwaters is particularly important due to the often incompletely consolidated sediments encountered during drilling and the potentially serious implications of drilling contamination on interpretation of inorganic water chemistry, organic water composition and model groundwater dates (Aggarwal et al., 2003; Darling et al., 2012; Goody et al., 2006; Harvey et al., 2003, 2002; Lawson et al., 2013; McArthur et al., 2011; Neumann et al., 2010; Rowland et al., 2007; Sengupta et al., 2008; van Geen et al., 2004; van Geen et al., 2008; van Geen et al., 2003).

Various tracers have been used during drilling procedures in the pursuit of different aims (Bath, 2011). The selection criteria for drilling tracers includes background levels; chemical stability in the relevant environment; the degree of conservative and non-sorbing behavior; ease and technical capability of in-field monitoring; analytical detection limits; cost of materials; and minimization of adverse effects on the environment, health (human and/or animal) and/or public perception and acceptability. Tracer selection is thus application-dependent and reflects a balance of considerations. Most tracers fall into two categories: (i) dye compounds and (ii) inorganic compounds (Bath, 2011) and the relative merits of these are discussed briefly below.

Dyes (fluorescent or non-fluorescent) are typically polycyclic aromatic carboxylic organic compounds such as rhodamine WT, rhodamine D, fluorescein, uranine, amino-G acid, fluorobenzoic acid and blue dye (Di Fazio and Vurro, 1994; Penteleit et al., 2006; Sabiani and Austin, 1991; Wandrey et al., 2010). Dyes are very commonly used and have the advantage of easy and rapid on-site analysis. However, organic dyes are much more chemically complex than water, have complex sensitivities to light, pH, salinity, temperature and water composition, and exhibit significant sorption, especially to clays that may be encountered during drilling procedures. These disadvantages severely diminish their value for drilling applications.

Alternatively, inorganic tracers, such as lithium, iodide, bromide, potassium, chloride, nitrate and thiocyanate, exhibit the advantages of simple hydrochemical behavior, simple composition, general insensitivity to environmental conditions such as pH, and improved persistence in drilling fluid as compared to organic dyes. However, inorganic tracers are often avoided in field studies as they generally require more complicated and off-site chemical analysis which may be impractical in the field context. Another potential disadvantage of cationic tracers, in particular, is that ion exchange reactions can occur between a cationic tracer and charged functional groups on the surface of clay materials if unbalanced electrical charges are present. Because these ion exchange reactions are reversible and highly dependent on the nature of the mineral framework within the sediments, they can be variable and difficult to quantify in a natural environment (Bath, 2011; Carroll, 1959; Gast and Klobe, 1971). It is well established that the affinity of various types of clays for cations decreases with increasing hydrated radii, with the order of preference being $\text{Cs}^+ > \text{Rb}^+ > \text{K}^+ > \text{Na}^+ > \text{Li}^+$ (Carroll, 1959; Gast and Klobe, 1971).

Thus, of the possible cationic tracers, lithium exhibits the lowest ion-exchange capacity with clay materials (Carroll, 1959; Gast and Klobe, 1971), indicating its comparative potential as an excellent tracer. Furthermore, background levels of lithium are generally low (for example ranging from <1.0 to 34 $\mu\text{g/L}$ in UK aquifers) (Kinniburgh and Smedley, 2001; Shand et al., 2007) ensuring that the majority of lithium observed is typically derived from tracer spiking. Indeed, lithium has previously been used as a drilling tracer for various applications such as site characterisation (McCartney and de L.G. Solb e, 1999), tracing in porous aquifers (Ptak et al., 2004; Vereecken et al., 2000), hydraulic tracing in wetlands (Dierberg and DeBusk, 2005), the evaluation of fluid-sediment reactions in volcanoes using isotopes (Chan and Kastner, 2000), fracking (Warner et al., 2014), and evaluating interstitial waters of marine sediments in ocean drilling (You et al., 1995; Zhang et al., 1998). Notwithstanding this, lithium is often avoided partly due to difficulties in in-field detection and, in particular, to our knowledge, lithium has not been used as a tracer to specifically evaluate the drilling-induced contamination of groundwater collected from shallow monitoring wells (<45 m in depth) drilled in poorly consolidated sediments.

Such poorly consolidated sediments hosting heavily exploited aquifers are widespread in many parts of the world but particularly in densely populated lowland river basins of circum-Himalayan Asia (Charlet and Polya, 2006). The use of these aquifers as a source of drinking water has led to massive detrimental health outcomes to those exposed to arsenic contained in the waters (Charlet and Polya, 2006; Polya and Charlet, 2009). Although this arsenic is largely held to be of geogenic origin, there is an ongoing debate about the importance of irrigation-enhanced ingress of surface-derived waters (Aggarwal et al., 2003; Harvey et al., 2002; Polya and Charlet, 2009) and detailed hydrogeochemical studies are critical to resolving this. Kandal Province, Cambodia is one of several areas in southern and south-east Asia broadly typical of such arsenic-impacted aquifers (Charlet and Polya, 2006) and is the focus of significant research studies of arsenic biogeochemistry (Buschmann and Berg, 2009; Charlet and Polya, 2006; Lawson et al., 2013; Polizzotto et al., 2008; Polya et al., 2008; Polya and Charlet,

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