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# The source of naturally occurring arsenic in a coastal sand aquifer of eastern Australia

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#### Abstract

The discovery of dissolved arsenic in a coastal aquifer used extensively for human consumption has led to widespread concern for its potential occurrence in other sandy coastal environments in eastern Australia. The development of an aquifer specific geomorphic model (herein) suggests that arsenic is regionally derived from erosion of arsenic-rich stibnite  $(Sb_2S_3)$  mineralisation present in the hinterland. Fluvial processes have transported the eroded material over time to deposit an aquifer lithology elevated in arsenic. Minor arsenic contribution to groundwater is derived from mineralised bedrock below the unconsolidated aquifer. An association with arsenic and pyrite has been observed in the aquifer in small discrete arsenian pyrite clusters rather than actual acid sulfate soil horizons. This association is likely to influence arsenic distribution in the aquifer, but is not the dominant control on arsenic occurrence. Arsenic association with marine clays is considered a function of their increased adsorptive capacity for arsenic and not solely on the influence of sea level inundation of the aquifer sediments during the Quaternary Period. These findings have implications for, but are not limited to, coastal aquifers. Rather, any aquifer containing sediments derived from mineralised provenances may be at risk of natural arsenic contamination. Groundwater resource surveys should thus incorporate a review of the aquifer source provenance when assessing the likely risk of natural arsenic occurrence in an aquifer.

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## 1. Introduction

In the last decade there has been an increased amount of research surrounding the occurrence of arsenic in groundwater supplies throughout the world. The most notable cases of regionally elevated arsenic in groundwater include aquifers in Argentina (Smedley et al., 2002; Bundschuh et al., 2004; Bhattacharya et al., 2006a), Bangladesh (Nickson et al., 2000; Anawar et al., 2003; Tareq et al., 2003; Ahmed et al., 2004; McArthur et al., 2004; Khalequzzaman et al., 2005; Harvey et al., 2005; Bhattacharya et al., 2006b), Chile (Caceres et al., 1992), Mexico (Romero et al., 2004), Taiwan (Chen et al., 1994), Vietnam (Berg et al., 2003) and many parts of the USA (Schreiber et al., 2000; Welch et al., 2000; Sidle et al., 2001).

Groundwater elevated in arsenic can be derived from both natural and anthropogenic processes. Natural sources include arsenic-rich aquifer matrices. Anthropogenic

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sources include the application of arsenical pesticides, disposal of industrial and mining waste, and leaching from cattle dip sites, cemeteries and tanneries.

Arsenic mobilisation in groundwater can be controlled by several different geochemical processes (Smedley and Kinniburgh, 2002; Bhattacharya et al., 2002, 2004). It is common for aquifers with high arsenic concentrations in groundwater to exhibit low arsenic sediment concentrations, particularly in geologically young, unconsolidated sediments. This phenomenon, and the historical lack of arsenic testing in many groundwater investigations may have led to the delayed discovery of high arsenic groundwater environments across the globe. Localised groundwater arsenic occurrences are now being discovered, with Australian unconsolidated coastal aquifers becoming a potential concern. Coastal areas are often heavily populated making groundwater a valuable resource. Arsenic occurrence in such aquifers is potentially a major issue due to their common use for high yield domestic and irrigation water supplies. Deterioration of coastal groundwater quality can have detrimental effects for human and biological users of these coastal aquifers.

This study was initiated in response to community anxiety over elevated concentrations of arsenic in ground-water of a coastal sandy aquifer in northeastern New South Wales, Australia. Previous studies conducted in this coastal groundwater environment (Smith et al., 2003, 2006) have concluded that arsenic may also be present in other coastal systems, particularly in eastern Australia where coastal aquifers have similar geologic, geomorphic and environmental settings. The source of arsenic deduced from these preliminary studies was thought to be associated with the oxidation of acid sulfate soil (ASS) material and/or desorption of arsenic from marine clays where it was adsorbed during sea level transgressions in the Quaternary Period. Further investigation is recommended to confirm or reject these hypotheses.

The primary goal of this investigation is to deduce the likely source of arsenic within the unconsolidated aquifer matrix. Identification of the correct arsenic source to the aquifer will aid in the successful management of this groundwater resource and potentially identify other aquifers that may be at risk of natural arsenic contamination. In the absence of any strong supporting evidence for anthropogenic contamination sources in the study area, the presence of arsenic in the aquifer is thought to be naturally occurring. Four arsenic source theories are hypothesised:

 i) Arsenic has been contributed to the aquifer matrix via deposition of regionally eroded geological units containing arsenic mineralisation;

- ii) Arsenic is derived from remnant seawater trapped in marine clay units deposited during eustatic changes of sea level in the Quaternary (Smith et al., 2006):
- iii) The oxidation of arsenian pyrite present in ASS material contributes dissolved arsenic to the groundwater (Smith et al., 2006); and/or
- iv) The underlying bedrock contains arsenic, which is being contributed to the aquifer via upwards vertical leakage of groundwater (Smith et al., 2003).

This investigation discusses the validity of each hypothesis and determines which is/are most likely to be the dominant arsenic source(s) in the aquifer.

#### 2. Materials and methods

## 2.1. Study site

Stuarts Point is located approximately 400 km north of Sydney, on the New South Wales (NSW) mid-north coast (Fig. 1). Groundwater for domestic town and farm use is extracted from the Stuarts Point aquifer outlined in Fig. 1.

The aquifer consists of heterogenous unconsolidated sediments overlying bedrock. Numerous geomorphic investigations have been conducted along the NSW coast (Langford-Smith and Thom, 1969; Roy and Thom, 1981; Thom, 2003) and more specifically the mid-north coast (Voisey, 1934; Hails, 1964, 1967, 1968; Walker, 1970). Two dominant depositional environments may have influenced the formation of the Stuarts Point unconsolidated aquifer. The first and most common geomorphic model applied to the northern coast of NSW is the development of inner (Pleistocene) and outer (Holocene) beach barrier systems. Hoyt (1966) proposed the dual barrier system primarily develops due to submergence of dune or beach ridges adjacent to pre-existing shorelines, and are therefore largely controlled by eustatic changes in sea level. The barriers are formed from onshore movement of material during these submergences (Hails, 1968). The second geomorphic model focuses on floodplain development. The Macleay River, which discharges to the sea within the study area, drains hilly tablelands before it broadens into a wide fluvial-deltaic floodplain to the east of Stuarts Point. Walker (1970) examined exposed soil terraces upstream of the floodplain and linked several terraces to buried floodplain deposits downstream, confirming the input of fluvial sedimentation within the Lower Macleay River floodplain.

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