Environmental Pollution 216 (2016) 738–745

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Health risk assessment for exposure to nitrate in drinking water from village wells in Semarang, Indonesia☆

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A R T I C L E I N F O

Article history:
Received 11 February 2016
Received in revised form
19 June 2016
Accepted 20 June 2016
Available online 8 July 2016

Keywords:
Nitrate
Birth defects
Drinking water
Hazard quotient
Methaemoglobinaemia

A B S T R A C T

The levels of nitrate in 52 drinking water wells in rural Central Java, Indonesia were evaluated in April 2014, and the results were used for a health risk assessment for the local populations by using probabilistic techniques. The concentrations of nitrate in drinking water had a range of 0.01–84 mg/L, a mean of 20 mg/L and a medium of 14 mg/L. Only two of the 52 samples exceeded the WHO guideline values of 50 mg/L for infant methaemoglobinaemia. The hazard quotient values as evaluated against the WHO guideline value at the 50 and 95 percentile points were HQ_{50} at 0.42 and HQ_{95} at 1.2, respectively. These indicated a low risk of infant methaemoglobinaemia for the whole population, but some risk for the sensitive portion of the population. The HQ_{50} and HQ_{95} values based on WHO acceptable daily intake dose for adult male and female were 0.35 and 1.0, respectively, indicating a generally a low level of risk. A risk characterisation linking birth defects to nitrate levels in water consumed during the first three months of pregnancy resulted in a $HQ_{50/50}$ values of 1.5 and a $HQ_{95/5}$ value of 65. These HQ values indicated an elevated risk for birth defects, in particular for the more sensitive population. A sanitation improvement program in the study area had a positive effect in reducing nitrate levels in wells and the corresponding risk for public health. For example, the birth defect HQ50/50 values for a subset of wells surveyed in both 2014 and 2015 was reduced from 1.1 to 0.71.

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1. Introduction

In many developing countries, groundwater wells provide the only source of potable water for much of the population. The groundwater is often used without any treatment and hence there is a significant risk of exposure to contaminants. Nitrate is one of the most significant and widespread chemical contaminants in aquifers and ground water around the world (Spalding and Exner., 1993). There are numerous possible sources of nitrate in groundwater, including improper disposal of sanitary wastes (ARGOSS, 2001), animal feedlots (Burkholder et al., 2007), nitrogen-containing fertilizers (Nolan et al., 1997) and agricultural land on which legumes are grown (Spalding and Exner., 1993).

The total daily intake of nitrate of humans can also be influenced by dietary factors, since certain vegetables (e.g. bok choy, cole and carrots) are known to contain high levels of nitrate, although the bioavailability of the ion has not been clearly defined (Hord et al., 2009). Nevertheless, a significant concern is associated with the presence of elevated levels of nitrate in drinking water. Excessive exposure to nitrate has been implicated as a causative agent in a number of human conditions. Chief amongst these are infant methaemoglobinaemia (World Health Organization (WHO), 2011) and cancers of the digestive tract in adults (Powlson et al., 1995). The mechanism of both effects is incompletely understood, but in

☆ This paper has been recommended for acceptance by Eddy Y. Zeng.

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http://dx.doi.org/10.1016/j.envpol.2016.06.041
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both cases probably involves the initial reduction of nitrate to nitrite (World Health Organization (WHO), 2011; Powlson et al., 1995). More recently, elevated levels of nitrate in drinking water supplied to women during the first trimester of pregnancy have been implicated in birth defects (Brender et al., 2013).

Several studies point to a direct association between methaemoglobinaemia and elevated levels of nitrate in drinking water (Knobeloch et al., 2000; Gupta et al., 2000). But the role of nitrate as a causative agent in infant methaemoglobinaemia is complex (Avery, 1999). It now appears that nitrate may be one of a number of co-factors that play a sometimes complex role in causing the disease (Fewtrell, 2004). There is general agreement that nitrate can play an important role in the incidence of methaemoglobinaemia and that drinking water is one possible source (Richard et al., 2014). As regards nitrate levels in drinking water, the guideline values currently promulgated (as nitrate ion): viz. 50 mg/L (World Health Organization (WHO), 2011) and 46 mg/L (United States Environmental Protection Agency (USEPA), 2015), both pertain to infant methaemoglobinaemia.

Disposal of sanitary waste is also a problem in developing countries. For example, data from the World Bank show that in 2012, only 54% of the Indonesian population had access to satisfactory excreta disposal facilities (private or shared). Assessment of such facilities however is frequently made on the basis of hygiene alone. Even in the case of a facility that is deemed satisfactory on hygienic grounds, there may still be a significant potential for contamination of drinking water wells. In such cases, elevated levels of nitrate are commonly encountered in water from adjacent wells (ARGOSS, 2001). It thus follows that schemes aimed at implementing sanitation in a developing country need to consider the possible consequences in terms of nitrate levels in the local groundwater (Templeton et al., 2015).

The present study reports levels of nitrate in drinking water from existing village dug wells and characterisation of the health risks this presents in areas with and without latrines in Semarang, Central Java. The results have been used to produce a comprehensive risk assessment using probabilistic techniques. The techniques of probabilistic risk assessment have been used in evaluating health effects for exposure to a number of chemical contaminants in previous studies, pertaining to a variety of situations or environments (Connell and Yu, 2008; Yu et al., 2012; Edokpolo et al., 2015). Through their ability to model a range of responses across a community, probabilistic techniques provide a more detailed estimate of risks than those obtained by use of stochastic techniques.

2. Materials and methods

2.1. Study area

The present study is focused on 29 villages close to Semarang, Central Java, Indonesia as shown in Fig. 1. All villages are located in the hills surrounding the city. Samples were collected from domestic wells and sampling sites were chosen on the basis of lack of adequate sanitary facilities according to WHO criteria [cf. Stewart and Laksono, 2002]. A number of schemes have been made available to villagers in respect of installation of suitable sanitary facilities. These include the BALatrine (Stewart and Laksono, 2002) and septic tanks. Full details of the initial sanitary survey are given in Park et al. (2015). An active program of improvement of sanitary facilities is ongoing within the study area. All wells sampled in the study were constructed with concrete surrounds at least 0.5 m high and were normally kept covered. None of the wells was in close proximity to a location where farm animals are kept in a confined area. The wells are all used for potable purposes, either by individual households or by a number of households and the water receives no treatment prior to use.

2.2. Collection of water samples and analysis

The main sampling was carried out in April 2014 and involved some 52 wells, with a subsequent re-sampling of 15 wells in 2015. The method of water collection by consumers from wells varied, with water being obtained from some wells by means of a bailer (34 sampling sites) and from others by means of a pump (18 sampling sites). So as to obtain as realistic a picture as possible of the water quality in domestic water, samples of water were collected from the wells by the manner (bailer/pump) which was routinely employed by the householders. In the case of samples collected from pumped wells, the pump was allowed to run for several minutes and the void water wasted, prior to collection of the sample. A subsequent re-sampling of selected wells was carried out in April 2015, so as to provide as direct a comparison as possible of the situation following intervention. Sampling was carried out at the same time of year to avoid any complications due to seasonal effects.

Water samples were collected with a 50 mL syringe from the surface bailer or bucket and passed through a 0.45 μm membrane filter into labelled sample containers. The containers were placed into a cooler with dry ice immediately after, to preserve the water samples, in accordance with AS/NZS 5667.1: 1998 (SAI Global, 1998). The water samples stored and transported frozen, prior to Flow Injection Analysis (FIA). All sample containers were taken from batches which had been shown by prior analysis to be free of significant nitrate and nitrite contamination.

2.3. Quality control

The water samples were sent to Brisbane, Australia for analysis at Queensland Health Forensic and Scientific Services, which holds National Association of Testing Authorities (NATA) accreditation for analysis of nitrate in waters. NATA accreditation requires satisfactory performance in interlaboratory trials and the laboratory concerned participates and performs successfully in the National Low Level Nutrient Collaborative Trial Program. The Limit of Reporting for nitrate in the present study is 0.009 mg/L.

Samples were stored below 5 °C prior to analysis. The laboratory achieves satisfactory performance as regards quality control program through the analysis of relevant certified reference materials, maintenance of control charts and participation in interlaboratory collaborative trials.

2.4. Estimation of water intake values

Human health risk assessments for nitrate in drinking water are presented in terms of three endpoints: infant methaemoglobinaemia; adult toxicity; and birth defects. Because the intake of nitrate from drinking water is a function to the quantity of drinking water consumed, it is necessary to obtain accurate estimates of this parameter, relevant to the study population. There are a number of calculation tools available to estimate daily water requirement. In the present study, H4H Hydration calculator (H4H hydration calculator., 2015) was employed. This tool has received input from Indonesian health professionals and hence offers the facility of calculating requirements specifically for pregnant women under Indonesian conditions.

2.5. Guideline values for nitrate in drinking water

The development of guideline values for nitrate concentration levels in drinking water by various organisations has been based on the endpoint of methaemoglobinaemia in infants during the first

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