Variability of naturally occurring fluoride in diverse community drinking-water sources, Tanna Island, Vanuatu

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ABSTRACT

Large variations in fluoride concentrations exist in natural waters, many of which are the source of community drinking-water supplies. Determining fluoride concentrations in community drinking waters can be challenging in developing Pacific countries such as Vanuatu that have limited laboratory capacity. Knowledge of naturally elevated fluoride concentrations that cause irreversible, adverse health outcomes may allow communities the opportunity to treat and manage their drinking-water supplies. Community drinking-water samples (n = 69), sourced from groundwaters, roof catchment rainwaters, surface waters and springs, were sampled on Tanna Island, Vanuatu between 2017 and 2020. In an 18 km² area of Western Tanna, a set of 30 groundwater-based drinking-water samples had a median fluoride concentration of 3.3 mg/L, with 20 samples > 1.5 mg/L and seven samples > 4.0 mg/L. These concentrations increase the risk of dental and skeletal fluorosis, respectively. Repeat resampling at five sites showed little variation over the sampling period. Rainwater-fed drinking-water supplies were lower overall and highly variable in fluoride concentrations (0.05–4.0 mg/L, median of 0.53 mg/L), with variable inputs from volcanic emissions from Yasur volcano. We recommend a comprehensive oral health and bone health study for the whole island to determine adverse health effects of excess fluoride in this vulnerable population.

Key words | drinking waters, fluoride, fluorosis, groundwater, Vanuatu, volcanism

HIGHLIGHTS

- Human endemic fluorosis occurs in Vanuatu, but there is no routine monitoring of drinking waters.
- Fluoride concentrations were determined in 69 community drinking waters on Tanna Island, Vanuatu.
- Twenty-three samples exceeded the WHO guidelines for drinking-water quality (1.5 mg/L F).
- Most high-F drinking waters were from groundwater sources.
- A comprehensive oral and bone health study is needed for Tanna Island.

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INTRODUCTION

The Pacific Small Island Developing States (PSIDS) are considered some of the most vulnerable nations on Earth. Pacific Islands often have challenging physical characteristics, undeveloped infrastructures and are exposed to increasing high-impact weather events (Barnett & Campbell 2010). They are listed amongst the nations with the highest disaster risk (United Nations University 2019) due to their exposure to hazards and lack of adaptive and coping capacity.

Water, sanitation and hygiene (WASH) frameworks initiated by the United Nations RES/18/1 (Human Rights Council 2011) allow PSIDS to move towards achieving 2030 Sustainable Development Goal 6 (SDG6) of safe, affordable, acceptable, available and accessible drinking water and sanitation for all (WHO 2016). Working towards this, it is necessary to consider all determinants of public health significance associated with drinking-water. The highest priority determinants are pathogenic microorganisms. However, chemical contaminants in drinking water, such as fluoride and arsenic, known to cause adverse health effects in global populations at concentrations in excess of drinking-water guideline values, also need to be considered when monitoring drinking-water supplies (Edmunds & Smedley 2013; Foster & Willett 2018).

The primary source of human fluoride exposure, absorbed via the gastrointestinal tract, is considered to be drinking-water consumption (Fejerskov et al. 1996). Some fluoride intake is considered beneficial for dental health; however, this is dependent upon daily dose, age, diet and climate. Fluoride, when made bioavailable, easily bonds with calcium-rich structures of the human body such as teeth and bone (Fejerskov et al. 1996). The continuous ingestion of excess fluoride in drinking water from birth to age 8 years causes irreversible dental fluorosis in children with severe forms of dental fluorosis generating porous tooth enamel, which is more vulnerable to tooth decay (Fejerskov et al. 1996). For children and adults consuming fluoride in drinking water over sustained periods, at values exceeding 4.0 mg/L, brittleness in bone increases, leading to an increased risk of fracture, debilitating and often painful permanent skeletal fluorosis (Fewtrel et al. 2006). Children are at most risk, with growing bones and developing adult teeth vulnerable to the attack of excess fluoride, with these health outcomes made worse if the human body is malnourished, or diet depleted in calcium and vitamin C (Edmunds & Smedley 2013). The World Health Organization (WHO) has a recommended upper limit (Guide for Drinking Water Quality, GDWQ) of 1.5 mg/L F in drinking water, for temperate climates (WHO 2011). Individual countries have been encouraged to consider their respective climates, with temperature increases associated with higher daily drinking-water consumption, and to lower their fluoride guideline values accordingly (Akuno et al. 2019).

The tropical PSIDS of Vanuatu (Figure 1) is considered the most disaster risk-prone country on Earth (United Nations University 2019). This ranking is based on the 83+ islands’ extreme exposure to tropical cyclones, floods, droughts, volcanic eruptions, earthquake and tsunami; high vulnerability due to limited infrastructure; and poor adaptive and coping capacity (United Nations University 2019).

In the 2016 mini-census, Vanuatu had a population of 270,459 inhabitants (VNSO 2016) with approximately 75% of the population defined as rural dwellers. Vanuatu’s main urban centres of Port Vila (Efate Island) and Luganville (Espiritu Santo Island) have treated, reticulated drinking-water supplies. Vanuatu has a resource-stretched Department of Water Resources (DOWR) who are responsible for management and national water compliance for reticulated systems. Rural communities, however, are responsible for managing their own delivery of safe drinking waters (DOWR 2019) with basic water facilities in rural villages often burdened with damaged water pipes and storage contamination (DGMWR 2008). There is an annual requirement to test for fluoride on islands, which may be affected by volcanic ashfall (DOWR 2019).

Very high rates of volcanic degassing have been recorded for the Vanuatu island arc (Carn et al. 2016). Cronin & Sharp (2002) sampled drinking-water supplies on two of Vanuatu’s active volcanic islands in 1999 and suggested that Ambrym is a more fluoride-rich system than Tanna. Yasur volcano, Tanna, has been active for centuries,
continuously degassing with sporadic large eruptions which distribute ash widely (Firth et al. 2014). Fluoride concentrations in rainwater samples from Tanna ranged from <0.05 to 0.16 mg/L, compared with higher concentrations on Ambrym, where three samples had an average and a maximum value of 2.1 and 2.8 mg/L F, respectively (Cronin & Sharp 2002), associated with an increased risk of dental fluorosis.

Resampling in 2001 on Tanna yielded a higher concentration of 1.05 mg/L F in surface water, close to the volcano (Cronin & Sharp 2002). Further sampling on Tanna, in 2015, recorded low concentrations of fluoride in rainwaters, 0.38–0.67 mg/L (Stewart et al. 2016). However, a groundwater sample measuring 3.9 mg/L F was recorded (Stewart et al. 2016), associated with increased risks of dental and skeletal fluorosis (Fewtrell et al. 2006), suggesting that volcanic emissions may not be the only, or even the most important, source of excess fluoride in natural waters on Tanna.

Tanna (Figure 1) is inhabited by >32,000 people, more than 10% of Vanuatu’s total population, who are primarily subsistence farmers (VNSO 2016). Further investigations of water composition are important to determine fluoride concentrations present in community drinking waters. Fluoride concentrations in groundwater tend to be stable over time (Amini et al. 2016), meaning that residents could be exposed to sustained high concentrations. In contrast, fluoride concentrations in rainwater supplies may be highly variable, as they are dependent on the strength of volcanic activity and the direction and dispersion of gas and ash emissions (Stewart et al. 2016).

The primary aim of this study was to identify influences on fluoride concentrations in community drinking-water
sources and to map high-fluoride areas. The secondary aim of this study was to gain insight into the variability of fluoride concentrations over time in groundwater- and rainwater-fed supplies.

**MATERIALS AND METHODS**

Samples \((n = 69)\) were collected from as many differing community drinking-water sites as possible across Tanna, between November 2017 and March 2020 (Supplementary Material, Table S1). Samples were taken from sites most used by community members such as schools, communal village sites and sites outside of villages known to be communal collection points. Most households in a village shared the same collection point with reticulation in most villages non-existent. At each location, advice was sought from local community leaders about the most appropriate supplies to sample. Repeat sampling was possible only at some sites and was opportunistic when weather and road conditions were favourable. It was carried out at sites 6, 7, 9, 11, 12, 19 and 31. Samples were taken from groundwater sources at private and public bores \((n = 30)\), which included primary and secondary schools, the main town centre of Lenakel, the regional hospital and the recently built reticulated town supply bore at Isangel. Springwater samples \((n = 21)\) were taken across the whole of Tanna and included three beach springs, indicated as an alternative drinking-water source during drought. Rainwaters collected off sheet metal roofs, at homes and schools, were sampled \((n = 13)\) when available, along with river water sites \((n = 5)\). Two samples were taken from a chamber inside the masiwer or masivier tree, endemic to South West Tanna and an earlier method of water collection by villagers. Three commercial bottled waters were also tested for their fluoride content, along with a town supply tap water sample collected in Port Vila, all sourced from Efate's aquifer. Bottled water is used widely throughout Vanuatu and is often provided as part of emergency responses.

Water samples were collected in clean, pre-rinsed plastic bottles and stored in a fridge.

As no commercial internationally accredited laboratory facilities were available in Vanuatu, the samples were transported back to New Zealand. The New Zealand and Vanuatu government do not utilise sample transfer agreements. As this study did not involve human or animal participants, ethical approval was not required. This project was done in partnership with, and the support of, the Vanuatu DOWR.

All samples were imported into New Zealand with the necessary biosecurity permits and securely delivered to Hills Laboratories, Hamilton. This laboratory is an approved biosecurity containment facility and has IANZ (International Accreditation New Zealand) accreditation. Samples were tested within 1–2 weeks with the stability of the fluoride ion stability in solution being in the order of months (Whitford & Riley 2014). Fluoride concentrations were determined using direct measurement by ion-selective electrode (APHA Method 4500-F-C, 22nd edition 2012). The detection limit is 0.05 mg/L. Samples were safely disposed of at the laboratory once tested.

**RESULTS AND DISCUSSION**

Fluoride concentrations for community drinking-water supplies sampled across Tanna are presented in a graphical form in Figure 2 and listed in a summary form in Table 1. The full set of data is available in Supplementary Material, Table S1. The highest fluoride concentrations (up to 4.6 mg/L) were found in groundwater supplies. Twenty of the 30 groundwater samples had fluoride concentrations exceeding the WHO’s recommended upper limit of 1.5 mg/L, and the median concentration was 3.5 mg/L. Fluoride concentrations were lower in the roof-collected rainwater samples \((n = 13, \text{median } = 0.53 \text{ mg/L})\), spring samples \((n = 21, \text{median } = 0.51 \text{ mg/L})\) and surface water \((n = 5, \text{median } = 0.58 \text{ mg/L})\) samples. The set of groundwater drinking-water samples is statistically different \((p < 0.001)\) from the roof-collected rainwater, spring and surface water samples (Table 1). Repeat sampling was carried out at several sites to determine the variability of fluoride concentrations over time (Table 2). Available data from an earlier study have been included for comparison for site 7 (Stewart et al. 2016). Resampled groundwater shows little variability over time in contrast with rainwater fluoride concentrations, site 19, varied from undetectable \((<0.05 \text{ mg/L})\) to very high \((4.0 \text{ mg/L})\).
Twenty-three drinking-water samples, across Tanna, exceeded Vanuatu’s upper limit of 1.5 mg/L F (Table 1). Twenty-six of the 30 groundwater samples were located in West Tanna (Supplementary Material, Table S1), an area that includes the main business area of Lenakel, markets of Blakman Town, the regional hospital, multiple primary schools, kindergartens and a large area high school. Groundwater results from West Tanna show a defined pattern of fluoride concentrations: lower in the north and south with higher yields in the centre of the western coast. Tanna’s western coast is predominantly comprised of limestone (Carney & Macfarlane 1979). The high concentrations of fluoride on west Tanna (>1.5 mg/L) coincide with this area of limestone which spans an approximate 18-km² area, with consistent, very high levels (>4.0 mg/L) clustered around the villages of Loukatai (Figure 2; Table 2).

Lower fluoride concentrations were reported for a newly drilled elevated, inland bore site, 6.5 km east of Loukatai, at D’Imaru Village, Middlebush (0.45 mg/L), as well
Fluoride concentrations in roof-collected rainwater are highly variable (Table 2) with a wide range of fluoride concentrations (<0.05–4.0 mg/L) found during two separate sampling events at Iquarmanu School, immediately adjacent to Yasur volcano. This is likely due to variability in volcanic activity, the direction of dispersion of volcanic emissions and dilution with clean rainfall. A rainwater sample from the village of Lavis, 29 km northwest of Yasur, contained 1.66 mg/L F, indicating that volcanic ash can elevate fluoride concentrations in rainwaters at considerable distances downwind.

In times of drought and disaster response, bottled water may be shipped; however, Tannese are proficient at accessing multiple drinking-water sources (VNSO 2016). For example, staff and students at Yaramanu School in the south-east of Tanna utilise rainwater tanks and a spring water source which is gravity-fed through a pipeline to the school. However, when these sources run dry, drinking-water is then transported daily by students, from an additional spring source approximately 300 m downhill. This practice is consistent with other Pacific communities, where local knowledge is shared amongst community members (McNamara & Prasad 2014). Villages around the volcano also access geothermal springs such as Sulphur Bay Beach (1.49 mg/L F) and Port Resolution Beach (1.12 mg/L F) during times of drought or ash contamination.

To date, little research has been done measuring the fluoride content in other South Pacific Islands. For the Fijian archipelago, which lies east of Vanuatu, fluoride in drinking waters across all islands ranging from 0.01 to 0.35 mg/L was reported during their 2015 National Oral Health Survey (Prasad et al. 2018). Sampling of fluoride in drinking waters in multiple communities at random sites across Vanuatu’s 80+ islands was conducted when possible during the 2017 National Oral Health Survey. Results ranged from <0.05 to 4.37 mg/L F with the highest results reported from West Tanna. However, as some samples from other islands exceeded the GDWQ upper limit of 1.5 mg/L (Ambrym and Efate Islands) (NOHS 2018), endemic fluorosis is likely to be not isolated to Tanna Island with further work required to establish accurate datasets for the whole of Vanuatu.

To date, there have been two studies performed on children to determine dental fluorosis for Tannese residents.

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**Table 1** | Fluoride concentrations in drinking water samples collected across Tanna Island between 2017 and 2020 by source

| N   | Source                  | Median (mg/L) | Range (mg/L) | Number of samples exceeding WHO guideline value of 1.5 mg/L |
|-----|-------------------------|---------------|--------------|-----------------------------------------------------------|
| 30  | Groundwater             | 3.30          | 0.38–4.60    | 20                                                        |
| 21  | Spring                  | 0.51          | 0.08–1.72    | 1                                                         |
| 13  | Roof-collected rainwater| 0.53          | <0.05–4.00   | 2                                                         |
| 5   | Surface water           | 0.58          | 0.11–1.10    | 0                                                         |
| 69  | Total                   | 1.54          | <0.05–4.6    | 23                                                        |

Superscripts a and b denote statistical difference between these sets of data.

**Table 2** | Fluoride concentrations in sites resampled over time, 2015–2020 Tanna Island, Vanuatu

| Site Area | Source               | Year sampled | Range, F (mg/L) |
|-----------|----------------------|--------------|-----------------|
| 6 West Tanna | Groundwater         | 2018, 2019, 2020 | 3.3           |
| 7 West Tanna | Groundwater         | 2015, 2018, 2018 | 3.9–4.0      |
| 9 West Tanna | Groundwater         | 2018, 2018, 2020 | 3.8–5.9      |
| 11 West Tanna | Groundwater       | 2019, 2020    | 4.6            |
| 12 West Tanna | Groundwater       | 2017, 2020    | 4.20–4.37     |
| 19 Whitesands | Rainwater         | 2018, 2020    | 0.05–4.0      |
| 31 South West Tanna | Spring water | 2019, 2020 | 0.17          |

*Bold data from 2015 are from Stewart et al. (2016).*

as the large, six bore water system supplying Lenakel business area, near Isangel (0.96 mg/L) just 3 km south of Loukatai (Figure 2).

Limited repeat sampling of five groundwater-fed supplies suggests a stable level of fluoride over a period of years (Table 2), similar to other studies that have determined temporal trends in groundwater fluoride concentrations (e.g., Amini et al. 2016).

According to the 2016 mini-census, post-Cyclone Pam, villagers in West Tanna use groundwater as one of their alternative drinking-water supplies (VNSO 2016). West Tannese reside on the lee side of the island in a rain shadow and requires constant access to additional drinking-water supplies during extended drought periods (DOWR 2019).
Allibone et al. (2012) reported a 100% prevalence rate of dental fluorosis in a small sample of children \( (n = 26) \), aged 6–18 years, who lived within 4 km of the volcano. However, in 2017, a comprehensive oral health survey was conducted for the whole of Vanuatu (NOHS 2018). Even though the sample sizes were small, due to being part of a large nationwide study, there were children who were examined on Tanna, who had dental fluorosis recorded. Twenty-three children, aged 11–13 years, who were examined at West Tanna had a 71.9% fluorosis prevalence rate, and a further 22 children, described as being ‘from Tanna’, had a 46.7% prevalence rate. Additionally, 26 children, aged 14–16 years examined at West Tanna, had a 57.7% prevalence rate of dental fluorosis and a 64.7% prevalence rate of dental fluorosis for 17 additional children indicated as ‘from Tanna’. Despite these small sample numbers, these results suggest an association between fluoride in drinking water and dental fluorosis for Tanna Island.

The presence of dental fluorosis in Tannese children surrounding the volcano, where samples yielded lower fluoride median concentrations in drinking waters, also suggests that residents may be exposed to additional sources of fluoride (Thompson et al. 2007).

A recent study on Vanuatu’s Ambrym Island (which hosts the two active cones of Benbow and Marum), considered additional fluoride intake from consuming locally grown foods, inhalation of airborne HF and accidental ingestion of soils (Calkins unpublished data). Staple foods such as taro and yam, grown in acidic, volcanic soils, on Ambrym have a high-fluoride content (Cronin & Sharp 2002).

Children in Vanuatu do not have access to any fluoride treatments prescribed and delivered by oral health professionals. Toothpastes are available in some stores but not often used; the most recent national oral health survey reports that only 21.8% of children and 47.8% of adults use fluoride toothpaste with a further 50% of children and 38.3% of adults responding they had no knowledge of whether their toothpaste contained fluoride (NOHS 2018). In this same survey, only 21.3% of children aged 5–7 years, who are the at-risk age group for developing dental fluorosis, report brushing their teeth twice a day \( (n = 403) \) (NOHS 2018).

Testing of rainwater-fed community drinking waters is an annual practice on Tanna, but only in areas affected by volcanic ash (DOWR 2019). We suggest that high-frequency testing across the entire island would be helpful to construct a comprehensive profile of fluoride concentrations in roofed rainwaters alongside a food web study to determine the total fluoride contamination for this environment (Calkins, unpublished data). It would also be beneficial to carry out a large comprehensive oral health survey of both DMFT/dmft (decayed, missing and filled teeth) and fluorosis to determine the optimal fluoride level in drinking waters for Tannese (Edmunds & Smedley 2013). Additionally, to date, no study of bone health has been implemented on Tanna and is also recommended, as long-term consumption of fluoride levels, greater than 4.0 mg/L, is consistent with the risk of developing skeletal fluorosis and, greater than 4.30 mg/L, consistent with debilitating skeletal fluorosis and hip fractures (Li et al. 2001).

Results suggest that remediation is urgently required in areas exceeding the upper GDWQ limit of 1.5 mg/L F. Popular fluoride removal techniques vary in effectiveness and cost with many methods requiring trained operators, a continuous electricity supply, additional treatment chemicals and regular monitoring and testing to ensure compliance (Edmunds & Smedley 2013). The use of low-cost precipitation techniques such as treatment with clay, bone or ash may not be efficient (Dhillon et al. 2017), it may be difficult to monitor at a village level and may require cultural approval for use. Low-cost remediation techniques used successfully in other endemic fluorosis regions (such as the addition of activated alumina) would require extensive piloting to confirm success on Tanna Island due to differing environmental field conditions (Dhillon et al. 2017). The lack of a reliable, accredited method for fluoride determination in the country is a further challenge.

**CONCLUSION**

This novel data set suggests an endemic fluorosis risk on western Tanna. Twenty samples of drinking waters from groundwater sources, collected in West Tanna between 2017 and 2020, exceed the WHO GDWQ for fluoride (1.5 mg/L), placing infants and children at risk of permanent dental fluorosis. Of these samples, seven contained >4.0 mg/L F. Consumers of these water supplies are at
risk of developing skeletal fluorosis. Limited repeat sampling over time of some of West Tanna groundwaters showed little variation in fluoride concentrations, suggesting that communities are likely to be consuming high levels of fluoride on a long-term basis, with children and infants at the highest risk of permanent adverse health effects. A comprehensive oral health survey of Tanna is recommended to determine the optimum fluoride level for drinking waters as well as bone studies to determine bone health. Further work is urgently required to develop locally appropriate fluoride remediation methods for Tanna Island.

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CONFLICT OF INTEREST

The authors of this study declare no conflicts of interest.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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