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POPULATION AT RISK ESTIMATION DUE TO GROUNDWATER FLUORIDE POLLUTION IN THE FLUOROSIS ENDEMIC AREA OF ASEMBAGUS, EAST JAVA, INDONESIA

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ABSTRACT

Groundwater is the world's primary source of drinking water. Unfortunately, we face the reality that access to safe drinking water is a worldwide issue nowadays. The presence of fluoride in groundwater is one of the causes of the fluorosis disease, which affects hundreds of millions of people around the world. Asembagus, an endemic area for fluorosis in Indonesia, requires updating its current risk status due to dynamically changing factors over time. Therefore, this research aims to estimate the probability of human health risk based on a population-at-risk calculation. Using an ion-selective electrode, we carefully tested 45 water samples from shallow groundwater in situ. The interpolation technique was used to determine the current potential risk zone and calculate the population at risk. The result showed that the fluoride concentration in the study area varied from 0.29 to 3.41 mg/L, with a mean of 1.22 mg/L. In Asembagus, the threat of skeletal fluorosis still affects 859 people (6.16%), and dental fluorosis affects 1,122 people (8.04%). Based on this study's findings, we concluded that the potential risk of non-carcinogenic fluorosis has significantly decreased since more than 20 years ago. This information is very useful for the government and public health authorities to further mitigate and protect people in the Asembagus area.

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Introduction

Fluoride (F) is the second-most significant geogenic pollutant that must be managed to ensure clean water access [1]. The consumption of fluoride in drinking water at optimum concentrations between 0.5 and 1.0 mg/L is beneficial for the prevention of dental caries, but excessive consumption leads to the development of fluorosis [2]. As low as 1.5 mg/L, fluoride causes mottled enamel or dental fluorosis, whereas greater than 3.0 mg/L causes skeletal fluorosis [3]. Furthermore, fluoride ions in excessive quantities in drinking water can result in not only fluorosis diseases that are already commonly known but also other findings on neurological disorders, cardiovascular issues, and brain issues, possibly contributing to attention deficit hyperactivity disorder (ADHD) and highly hazardous endocrine disruptors for diabetics [4].

Asembagus is an endemic area for fluorosis in Indonesia [5],[6]. The Asembagus community primarily consumes water from shallow groundwater [7]. Consequently, by using groundwater as the source for drinking water, dental fluorosis is developing and posing a risk at the lowest level concentration of 0.5 mg/L [6],[8], while skeletal fluorosis is 1.1 mg/L [7]. Compared to 1.5 mg/L for the Indonesian standard for fluoride in drinking water recommendations based on the Ministry of Health Regulation in 2023, Asembagus is an

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example of a case in Indonesia where 1.1 mg/L fluoride concentration in groundwater already poses a high risk to human health.

The hazard quotient regarding fluoride pollution in the Asembagus was already calculated around 20 years ago by [7]. However, after 20 years since the first risk assessment, the situation is significantly changing. The dug well as a source of drinking water was abandoned by the majority of people in Asembagus. During the field observations in 2022, the people of Asembagus realized that their drinking water was polluted by the volcanic water. So, they left their groundwater as the source of drinking water and changed their drinking water to a gallon of water or used the municipal waterworks. Thus, a re-calculation of potential risk is needed in order to update the current status and further consider a precise countermeasure to overcome the existing problems. In order to efficiently mitigate and overcome the risk potential based on current status, the population at risk estimation calculation is crucially needed [9]. Therefore, this research aims to estimate the number of populations at risk due to fluoride contamination in the groundwater of the Asembagus fluorosis endemic area in the current situation. The information about potentially impacted people can be helpful, as it is fundamental to choosing the best mitigation techniques to reduce the high risk of fluorosis disease [10].

Method

Research area and data collection

Asembagus is a small town in the Situbondo Regency, East Java, Indonesia. Asembagus is considered for the research area based on a new theoretical approach for the global fluoride belt (GFB) that has been described by [11]. In this theory, Indonesia is grouped as the volcanic fluoride belt, where the source of fluoride in groundwater is associated with volcanic activity. Primary and secondary data were observed and sampled during surveys in the dry season of August 2022. Secondary data, including public health facility locations, clean water municipality networks, and river networks, was collected from the institutional data. All primary samples were collected within the Asembagus area, as shown in **Figure 1**.

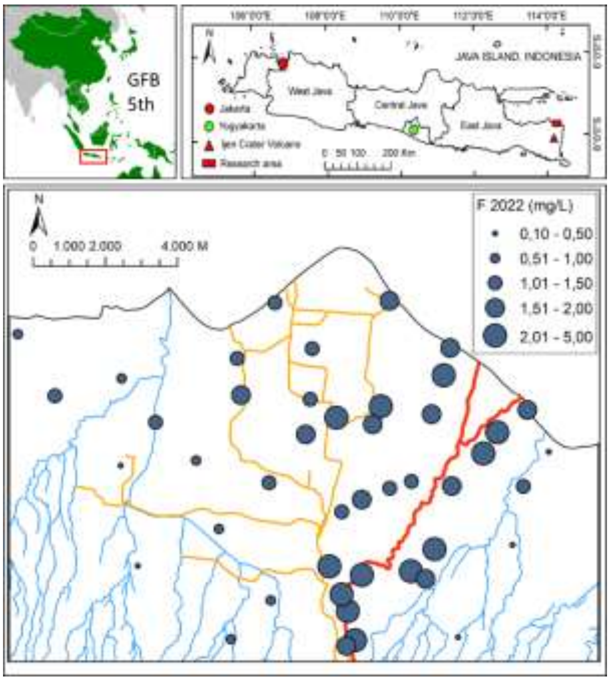

Figure 1. Research area map based on Global Fluoride Belt 5th by [11]

A total of 45 groundwater samples were taken from dug wells during the survey and observation. During the fieldwork activities, the coordinates of groundwater were plotted using Garmin GPSMAP 64s. Fluoride concentration in groundwater was determined electrochemically using a fluoride ion-selective electrode (ISE) with the Fluoride Ion Meter TiN 5101 from TOKO Chemical Laboratories, Japan. In order to reduce the amount of contamination or not-ideal water samples, each well was pumped for 10 to 15 minutes before obtaining fresh groundwater, bottling it, and then sealing it in the sampling bottles. One bottle of samples was prepared for in-situ chemical analysis at the fieldwork home base in the research area. All of the groundwater samples were kept in cold storage and stored in the refrigerator at a temperature of -4 °C and a room temperature of 25 °C at the fieldwork homebase during the fieldwork campaign in August 2022.

Spatial analysis and population-at-risk calculation

The Empirical Bayesian Kriging interpolation technique was used to define the distribution of fluoride concentration in groundwater [12]. This pattern is not only very useful to visualize the high concentration zone area but also to explain the classification level of pollution based on concentration distribution. Basic calculations, data preparation, supporting, and statistical analysis, including minimum, maximum, average, and median quartile, were done using Microsoft Excel 2021. Population at risk was calculated using a combination of population density and total predicted area for classified concentration in raster-based operation following **Equation 1** [13]:

$$Epr = (Gwp/As) \text{ x } Afp \quad (1)$$

where the *Epr* is population at risk (people), *Gwp* is the number of people that used groundwater, *As* is the settlement area in square units, and *Afp* is the predicted fluoride concentration area in square units. The concentrations in this research were categorized and classified using spatial classification based on the lowest level concentration of 1.1 mg/L for skeletal fluorosis described by [7]. The classification is safe (F < 1.1 mg/L), dental fluorosis (1.1< F< 2.5 mg/L), and skeletal fluorosis (F > 2.5 mg/L). The percentage of people using groundwater for drinking water was calculated using data from the statistical agency Situbondo Regency for each subdistrict in the 2022 report.

Results and Discussion

Results

The fluoride concentration from the total 45 sample analyses varied from 0.29 to 3.41 mg/L, with a mean of 1.22 mg/L. We further analyzed this pattern using the empirical Bayesian Kriging (EBK) interpolation model, revealing the spatial distribution pattern in **Figure 2**. The spatial model's accuracy was 92%, based on 10 fluoride samples as a validator control. According to the spatial model, the pattern clearly indicated that the middle zone was the most polluted area in the research area. The red color, graded yellow, indicates a high concentration of fluoride in the groundwater, while the green color indicates a low concentration. The red line represents the river, or Banyuputih, while the orange line represents the irrigation networks. Both of them contain extremely toxic water that originates from the crater water of the Kawah Ijen volcano, located approximately 40 km south.

In **Figure 2**, the concentration of interpolation ranges from 0.1 to 3.4 mg/L. A value of 0.1 mg/L indicates low concentration, while the maximum value of 3.4 mg/L is the highest concentration of fluoride in groundwater. We identified two hot spots with a red color in the middle and northern regions of the research area, where the concentration ranged from 1.5 to 3.4 mg/L for the red zone, and a yellow zone marked the medium concentration between 0.8 and 1.5 mg/L. The outer zone, primarily located in the western and eastern parts of the research area, has the lowest concentration of fluoride in groundwater, typically below 0.8 mg/L.

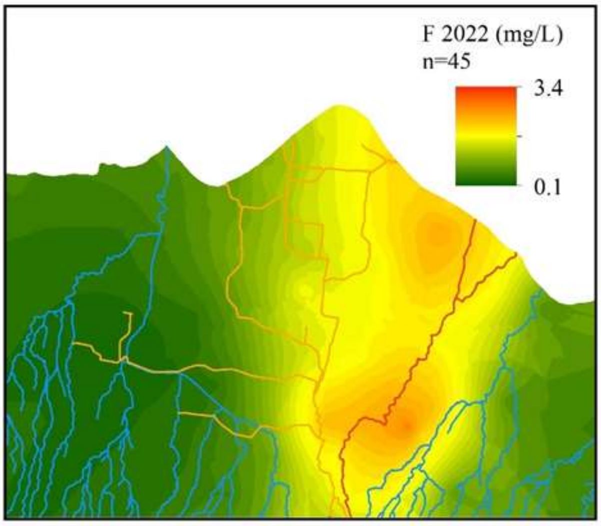

Figure 2. Spatial model of fluoride concentration in groundwater

During the field observations in 2022, we observed that the people of Asembagus realized their drinking water was polluted by the volcanic water. Consequently, they left their groundwater as the source of drinking water and changed their drinking water to a gallon of water or used the municipal waterworks in **Figure 3**. All of these situations will change the risk situation in Asembagus at the current time. Therefore, a re-assessment of non-carcinogenicity due to fluoride exposure in Asembagus should be conducted to reveal the current risk status and efficiently mitigate the problems.

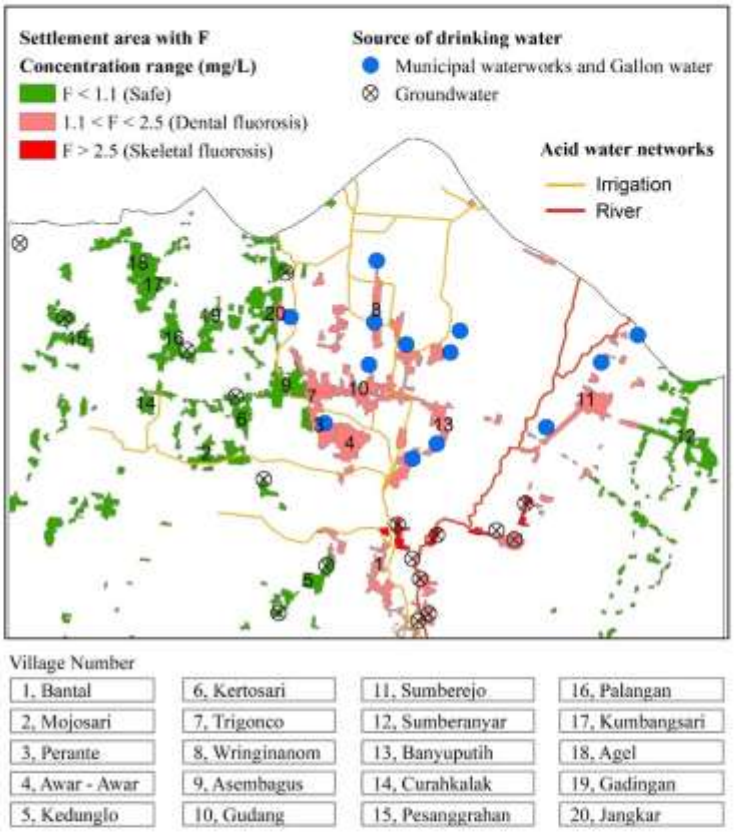

Figure 3. Distribution map of settlement-impacted zone and source of drinking water

To effectively reduce and manage the risk potential, we calculated and presented the population at risk using **Equation 1** in **Table 1**. Asembagus still exposes a total of 859 people to the threat of skeletal fluorosis and 1,122 to dental fluorosis. A total of 10,298 people who could have been exposed to dental fluorosis are no longer relevant because they have not used groundwater as a source of drinking water for the last two decades. Furthermore, we should focus on mitigation and monitoring in villages such as Awar-awar, Sumberrejo, and Banyuputih that still pose a potential risk. This application of population at risk based on the occurrence mechanism status significantly improved management efficiency.

Table 1. Population at risk calculation based on current fluoride hazard zonation

Village	*As*	*Gwp*	*Gwp/As*	*Afp*		*Epr* to fluorosis		Risk
				F = 1.1-2.5	F > 2.5	Skeletal	Dental	status
Bantal	507,740	1,122	.0022	507,740		0	1,122*	Ed
Perante	534,472	1,217	.0023	329,995		0	751	N
Awar – awar	611,279	790	.0013	574,506	31,847	41*	742	Es
Trigonco	614,136	951	.0015	432,167		0	669	N
Wringinanom	1,056,599	1,615	.0015	1,056,599		0	1,615	N
Asembagus	677,321	1,405	.0021	309,405		0	642	N
Gudang	465,365	1,175	.0025	465,365		0	1,175	N
Sumberejo	1,520,302	3,360	.0022	1,245,250	203,262	449*	2,752	Es
Banyuputih	850,143	2,320	.0027	715,080	135,064	369*	1,951	Es
Total	6,837,359	13,955	-	5,636,108	370,172	859	11,420	3Es,1Ed 5N

Notes: *As* = settlement area (m^2); *Gwp* = the number of people using groundwater in the village; *Afp* = predicted fluoride concentration area (m^2); *Epr* = population at risk estimation (people); asterisk symbol (*) = still using groundwater as the source for drinking water; Es = existing potential risk for skeletal fluororis; Ed = existing potential risk for dental fluororis
N = non-existing potential risk.

Discussion

Compared to previous research, the fluoride values in groundwater remained within the range and did not change significantly. This situation may be correlated with the fact that the acid river, which contains high fluoride, has been flowing for more than a hundred years, as reported by [14]. This long exposure time makes it possible to consistently transport fluoride from acid river water to shallow aquifer systems. According to surveyed data from 1979, the range is 0.20 to 2.70 mg/L [6]. As reported by [8], the groundwater fluoride concentration increased in 1999, reaching 0.41 to 3.25 mg/L. In 2001, the range was from 0.10 to 4.20 mg/L, as analyzed by [7]. Since fluorosis disease is the primary concern in the Asembagus area, the risk estimation was focused on the number of people who could potentially be exposed to the risk of skeletal and dental fluorosis after a 20-year hazard quotient (HQ) calculation by [7]. Asembagus continues to expose a total of 859 individuals to the risk of skeletal and 1,122 individuals to dental fluorosis. Because they have not utilized groundwater as a source of drinking water for the past two decades, a total of 10,298 individuals who could have been susceptible to dental fluorosis are no longer pertinent.

The current groundwater conditions in Asembagus have presented significant challenges. For example, the abandonment of numerous dug wells due to groundwater pollution with fluoride has consequences for socioeconomic problems, as reported by [15]. Most individuals in Asembagus have discontinued using the dug well as a drinking water source. Field observations conducted in 2022 revealed that the residents of Asembagus were aware that volcanic water had contaminated their drinking water. As a result, they abandoned their reliance on groundwater as a drinking water source and instead began using either a gallon of water or the municipal waterworks. These scenarios will alter the risk status and increase society's vulnerability to the economic crisis in Asembagus.

On the other hand, even though previous research clearly stated that Asembagus is an endemic fluorosis area, the fluorosis data record is not available. Previous research conducted by [16] was the only recorded public data on fluorosis in Asembagus. In Asembagus, the local public health center, located in every subdistrict, provides information on fluorosis diseases. Unfortunately, the public health center only recorded the sick people who consulted and received treatment in their facility. The Asembagus public health center recorded about 260 cases of dental fluorosis in 2014. However, a large number of people, as indicated, do not want to go to the public health center due to economic factors, a lack of knowledge, or a sense of shame. This situation has persisted until now and has impacted the limitations of fluorosis data in Asembagus.

In addition to fluorosis, local public health data revealed that dental caries is also a common disease in Asembagus. In Asembagus, the average number of dental caries cases reached 2,631 per year [17]. Fluoride hazard zonation, based on the intrinsic acid irrigation model, confirmed this data, revealing a 26% concentration among dental caries sufferers, 12% in the recommended health range, 56% in the potential chronic health effects of fluorosis-mottled teeth, and 6% in the potential chronic health effects of dental fluorosis and skeletal fluorosis [18]. Thus, dental caries also appears in the low fluoride concentration zones in Asembagus, mainly in the eastern and western parts of acid drainage networks.

The current risk situation of fluoride concentrations in drinking water, coupled with its chronic health effects, has resulted in dental caries and fluorosis-mottled teeth due to fluoride-polluted groundwater in Asembagus. This study provides an effective and efficient technique for analyzing the population's at-risk potential based on a spatial distribution model. This technique offers a significant advantage in assessing crucial public health information, serving as a preliminary step towards further mitigating the fluorosis disease problem [19]. An approach such as education to the people, engineering design of clean water networks, and of course, public health treatment in the impacted area will need the number of population at risk to help better technically plan [20].

Based on the current situation and the number of people exposed to the high fluoride concentration in the fluorosis endemic area, we can propose some countermeasures to effectively reduce the potential risk of fluorosis from chronic exposure. It is also a priority to develop a monitoring system and coordination among the local government and stakeholders, such as medical institutions and clean water supply agencies [21]. Despite the benefits of this research, there is still room for improvement. This study did not take into account the actual fluorosis cases in the research area. The difficulty in obtaining actual case information stems from the social stigma associated with sharing information about physical health conditions related to fluorosis. In future research, the actual data about flurorosis disease should be identified as an important factor in validating the distribution model of the high concentration zone. Another crucial step involves educating the affected society to gain a deeper understanding of their health issues, and fostering confidence in them to openly discuss their health problems.

Conclusion

The current calculation shows a significant reduction in the potential population at risk of non-carcinogenic fluorosis in Asembagus, from 20 villages in the previous estimation to only 3 villages in the high-potential-risk area. The critical changing factor is due to intake being almost zero in the highly polluted area since the people abandoned their wells and switched to gallon or municipal clean water networks for their drinking water sources. The current risk status analysis is an efficient process for mitigating and protecting people from fluoride pollution problems.

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