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ENVIRONMENTAL HEALTH RISK ASSESSMENT ON GROUND WATER AND AIR OF LIMESTONE IN GRENDEN VILLAGE EAST JAVA, INDONESIA

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ABSTRACT

Background: Grenden is a village at Jember, East Java, Indonesia that have limestone area, which also have a lot of industrial and quarry limestone. However, in spite of remarkable contribution towards economic development, some adverse impacts have been noticed such as the health risk among the inhabitant around limestone areas, because limestone contains of chemical which largely contains of calcite ($CaCO_3$). The health risk of $CaCO_3$ exposure from ground water and air could be studied applying Environmental Health Risk Assessment (EHRA). The purpose of this research was to observ environmental health risk from ground water and air that probably contain limestone in Grenden Village.

Materials and Methods: The method of this research was descriptive observational, that have two kind of sample which is environment and human. Environmental sample in this research is 32 well's water of inhabitant and ambient air sample. Each ground water sample and ambient air sample will be measured their hardness (CaCO3) parameter and Total Suspended Particle (TSP) content. Human sample (respondent) was selested from inhabitant whose well water was taken and near the location of air sample. Respondents were subjected to anthropometric surveys for body weight, ingestion rate, exposure times, exposure frequency and duration time.

Result: The averange of water hardness (CaCO3) value was 207,906 mg/L with maximum 530 mg/L and minimum 111 mg/L. While, TSP content in ambient air at point 1, 2 and 3 are 0.024 mg/m^3, 0.054 mg/m^3 and 0.113 mg/m^3 respectively. Risk characterization on population showed that RQs of CaCO3 exposure through ingestion was greater than one (RQs > 1), and ECR of TSP exposure through inhalation was greater than one (ECR < 1). Risk management could be done by reducing CaCO3 concentration on well water until 1.990125 L/day and, reducing TSP concentration until 0.000621 mg/m^3 and exposure duration until 0.27331 year.

Conclusion: The population at limestone area in Grenden Village which exposed with CaCO3 through ground water and ambient air was at risk. The Risk level of non carcinogenic exposure of ground water was greater than one (RQs>1) and carcinogenic exposure of TSP in ambient air was greater than 10^{-4} (ECR).

Keywords: $CaCO_3$, Total Suspended Solid, limestone area, risk assessment,

1.0 Introduction

Indonesia has a lot of natural resources and diverse geographic condition. Limestone mountain is one of the natural resource that spread every province. For instance, East Java province has some limestone mountain at South Pacitan, South Blitar, Nusa Barung Island, Ujung Blambangan, Bukit Rembang, Dander, South Babad, North and South Madura, and also Puger.[1]

Grenden village is one of the areas in Puger District that has a limestone area, wherein the limestone classified into class C extractive limestone and manganese formation. Limestone exploration has been conducted since 1960s with land area of 30 hectares from 279 hectares of hill area with 80 meters high. Each year, limestone mining increase wherein in 2017 Grenden village produced 475,251 tons of limestone. Grenden Village had many kind of limestone processing industri that are mining, burning and the cement making.[2]

The limestone industry in Grenden Village has a posstive impact wherein 13.6% of inhabitant in the limestone area work in the limestone burning and mining industries. The distance between limestone processing industry and residential area isn't far (100-200 meters). Thus indirectly affecting environmental health condition of the inhabitant which is caused by air and water pollution from the industrial process. Based on the data of Community Health Center in 2016, Grenden Village had more acute bronchitis cases compared to other villages, i.e. Puger Kulon, Puger Wetan, mojosari and mojomulyo. The number of bronchitis cases on Grenden were 206 cases. Furthermore, the number of rheumatoid arthritis which included on 15 major diseases in Puger was 786 cases.

Health risks in limestone area may arise due to the chemical content which are mostly composed of calcite, a few iron, magnesium, carbonate, silica, alumina and other chemicals. While pure limestone composed of 90-95% CaO or 75-90% $CaCO_3$.[3] The pollution in the limestone's area occur in some media, i.e. air, ground and surfaces water that can enter the body through oral and inhalation. Grenden Village's inhabitant use groundwater as their drinking water which can be seen in Puger Community Health Center data in December 2017 wherein, there were 5,040 household. This shows Grenden's inhabitant have risk of being exposed to CaCO3 through well water.

Based on regulation from The Health Ministry of Indonesia number. 416/menkes/PER/IX/1990, CaCO3 levels in water should not exceed 500 mg/L.[4] otherwise, it can cause several diseases such as hypercalcemia and alkaline syndrome.[5] In addition, cardiovascular disease, cancer, cerebrovascular mortality, Alzhemeir's, atopic dermatitis in children, kidney stones, reproductive health disoreders, indigestion and constipation may result from high levels of CaCO3 consumption.[6] Lime dust limits based on regulation from The Health Ministry of Indonesia number 41 on 1999 are 230 μg / Nm3 for TSP (24 hour measurement).[7] The exceed can cause respiratory tract disease such as respiratory irritation, increased mucus production, airway narrowing, loss of cilium and mucous membranes, and irreversible Chronic Obstructive Pulmonary Disease (COPD).[8]

The study on epidemiological study and the health and safety aspects of mining and limestone processing in Grenden Vilage had been studied but There are no study focus on risk assessments by Environmental Health Risk Analysis (EHRA). The assessment is measuring

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lifetime exposure prediction as well as the time of its fragmentation. EHRA is an approach to scrutinize the potential and magnitude of risks that begin by describing known and recognized environmental problems, and involves setting the risks to human health related to environmental issues. There are several steps that must be implemented that is hazard identificaion, exposure analysis, dose analysis response, risk characterization and risk management.[9] The purpose of the result to give information for goverment to control health risks.[10]

2.0 Materials and Methods

This research used a descriptive study with a case control design and analysis of environmental health risk methods. This study was conducted on may 2018 at residential of limestone area in Grenden Village Puger Sub District. There were two samples that will be taken, which is human and environment sample. The inclusion criteria of those samples were located near the limestone mountain (100 – 400 meters) and the well water was used as drinking water. Human sample (respondent) were inhabitant whose well water already took in this research and also located near to the air sample point. Whereas the environment sample in this research is 32 wells water of inhabitant and 3 sample point of ambient air. The ground water sample and ambient air samples were inspected through their water hardness (CaCO3) parameter and Total Suspended Particle (TSP) of air. All of the process from sample taking until testing were gudied from SNI 6989.58-2008, SNI 06-6989.12-2004, and SNI 19-7199.3-2005.[11,12,13]

$$n = \frac{N}{1+N\,(d)^2} \qquad (1)$$

n : amount of sample
N : amount of inhabitant's well
d : confidence level (0.25)

Table 1. The number of well water sample based on Sub Village

No.	Sub Village	Well number	Sample
1	Dusun Krajan I	1,806	16
2	Dusun Kapuran	887	16
Total			32

The variables used to calculate intake and Risk Quotient (RQ) are CaCO3 and TSP concentration (C), ingestion and inhalation rate (R), time of exposure (tE), frequency of exposure (fE), duration of exposure (Dt), average time periode (duration of exposure x 365 days) and respondent's weight. The equation was used to calcute I and RQ shown in the equation (2) and (3).

$$I = \frac{C\,x\,R\,x\,t_E\,x\,f_E\,x\,D_t}{W_b\,x\,t_{AVG}} \qquad (2)$$

$$RQ = \frac{I}{RfD/RfC} \qquad (3)$$

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3.0 Result

3.1 CaCO3 (Water Hardness) and TSP Concentration

Table 2. The Results of Water hardness (CaCO3 Concentration) at Krajan 1 and Kapuran Sub Village

Sample number	CaCO3 concentration (mg/L)	Sample number	CaCO3 concentration (mg/L)
1	136	17	149
2	111	18	182
3	120	19	149
4	137	20	115
5	160	21	146
6	153	22	115
7	163	23	140
8	144	24	160
9	418	25	135
10	530	26	221
11	252	27	248
12	364	28	152
13	237	29	245
14	407	30	211
15	272	31	233
16	288	32	160

The results of CaCO3 concentration in inhabitant were described in Table 2. CaCO3 concentration in Krajan 1 and Kapuran Sub Village ranging from 111 mg/L to 530 mg/L and the average was 207.906 mg/L. Based on normality test result, the data wasn't normally distribute wherein the p-value is 0.000 (p-value < 0.05). Therefore, CaCO3 concentration used for calculate population intake was 160 mg/L (median value). While the results of TSP of air concentration were ranging from 0.024 mg/m^3 to 0.113 mg/m^3 and the average was 0.0637 mg/m^3. The normality test result of TSP concentration showed that the data was normally distribute wherein the p-value is 0.645 (p-value > 0.05), so TSP concentration that use for calculate population intake is 0.0637 mg/m^3 (mean value).

3.2 Intake Assessment

The result of anthroprometric survey showed that from 90 respondent consisted of 51 female respondent (56.7%) and 39 male respondent (43.3%). The age was ranging from 7 – 90 years old. The body weight was ranging from 20 – 82 kilograms. Normality test result showed that the data was normally distribute which p-value is 0.2 (p-value > 0.05). For calculating the population intake was used mean value of body weight (52.2 kg). Ingestion rate of all respondent were ranging from 1 litre/day until 6 litre/day where the average was 2.2667 litre/day. The normality test result showed that the p-value was 0.00 (p-value < 0.05) which mean the data was not normally distribute. Therefore, the ingestion rate that use for calculte intake population is 2 litre/day (median value).

Duration exposure of all respondent were ranging from 1 year – 87 years with average 29.7389 year. The normality test result showed data was not normally distribute, which p-value is 0.011 (p-valure < 0.05). Calculating the population intake used median value of

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duration exposure, which is 28 years. While, frequency of exposurew as 365 day/year. The last variable used to calculate population intake was time of exposure which was ranging from 10 – 24 hour/day. The normality test result showed p-value was 0.00, the value that use for calculating population intake was 23 hour/day (median value).

3.2.1 Non Carinogenic Ingestion Intake of Population

CaCO3 concentration (C)	160 mg/L
Ingestion rate (R)	2 litre/day
Frequency of exposure (f_E)	365 day/year
Duration of exposure (D_t)	28 years
Body weight (W_b)	52.2 kg
Average time periode (t_{AVG})	28 years x 365 day/year

$$\text{Ink}= \frac{C \text{ x } R \text{ x } f_E \text{ x } D_t}{W_b \text{ x } t_{avg}} = \frac{160 \frac{mg}{L} \text{ x } 2 \frac{litre}{day} \text{ x } 365 \frac{day}{year} \text{ x } 28 \text{ years}}{52{,}2 \text{ kg x } (28 \text{ years x } 365 \frac{day}{years})}$$

$$= \frac{160 \frac{mg}{L} \text{ x } 2 \frac{litre}{day} \text{ x } 365 \frac{day}{year} \text{ x } 28 \text{ years}}{52{,}2 \text{ kg x } 10220 \text{ day}}$$

$$\text{Ink}= 6{,}130268 \frac{mg}{kg} \text{day}$$

The results of ingestion intake were calculated in the equation above. From the calculation result showed that for one day, the population consumes water containing CaCO3 that enter into the body as much as 6.130268 mg/kg day. The value of intake can also be calculated and expressed for the lifetime exposure of the long-lived population living in the Grenden limestone area up to 30 years. The calculation result of lifetime intake (non-cancer intake) for 5, 10, 15, 20, 25, 30 years ahead showed in Figure 1.

3.2.2 Non Carcinogeni Inhalation Intake of Population

TSP concentration (C)	0.06367 mg/m^3
Ingestion rate (R)	0.83 m^3/hour
Frequency of exposure (f_E)	365 day/year
Time of exposure (t_E)	23 hours/day
Duration of exposure (D_t)	28 years
Body weight (W_b)	52.2 kg
Average time periode (t_{AVG})	28 years x 365 day/year

$$\text{Ink}= \frac{C \text{ x } R \text{ x } t_E \text{ x } f_E \text{ x } D_t}{W_b \text{ x } t_{avg}}$$

$$= \frac{0{,}06367 \frac{mg}{m^3} \text{ x } 0{,}83 \frac{m^3}{hour} \text{ x } 23 \frac{hour}{day} \text{ x } 365 \frac{day}{year} \text{ x } 28 \text{ years}}{52{,}2 \text{ kg x } (28 \text{ years x } 365 \frac{day}{year})}$$

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$$= \frac{0{,}06367\ \frac{mg}{m^3} \times 0{,}83\ \frac{m^3}{hour} \times 23\ \frac{hour}{day} \times 365\ \frac{day}{year} \times 28\ \text{years}}{52{,}2\ kg \times 10220\ day}$$

$$\text{Ink} = 0{,}023285\ \frac{mg}{kg}\ day$$

The results of inhalation intake were calculated in the equation above. The result showed that for one day, the population exposed with TSP in ambient air as much as 0.023285 mg/kg day. The value of intake can also be calculated and expressed for the lifetime exposure of the long-lived population living in the Grenden limestone area up to 30 years. The calculation result of lifetime intake (non-cancer intake) for 5, 10, 15, 20, 25, 30 years ahead can be seen in Figure 2.

3.2.3 Carcinogenic Inhalation Intake of Population

TSP concentration (C)	0.06367 mg/m^3
Ingestion rate (R)	0.83 m^3/hour
Frequency of exposure (f_E)	365 day/year
Time of exposure (t_E)	23 hours/day
Duration of exposure (D_t)	28 years
Body weight (W_b)	52.2 kg
Average time periode (t_{AVG})	70 years x 365 day/year

$$\text{Ink} = \frac{C \times R \times t_E \times f_E \times D_t}{W_b \times t_{avg}}$$

$$= \frac{0{,}06367\ \frac{mg}{m^3} \times 0{,}83\ \frac{m^3}{hour} \times 23\ \frac{hour}{day} \times 365\ \frac{day}{year} \times 28\ \text{year}}{52{,}2\ kg \times (70\ \text{year} \times 365\ \frac{day}{year})}$$

$$= \frac{0{,}06367\ \frac{mg}{m^3} \times 0{,}83\ \frac{m^3}{hour} \times 23\ \frac{hour}{day} \times 365\ \frac{day}{year} \times 28\ \text{years}}{52{,}2\ kg \times 25550\ day}$$

$$\text{Ink} = 0{,}009313\ \frac{mg}{kg}\ day$$

The results of inhalation intake were calculated in the equation above. The result showed that for one day, the population exposed by TSP in ambient air as much as 0.009313 mg/kg day. The value of intake can also be calculated and expressed for the lifetime exposure of the long-lived population living in the Grenden limestone area up to 30 years. The calculation result of lifetime intake (non-cancer intake) for 5, 10, 15, 20, 25, 30 years ahead can be seen in Figure 3.

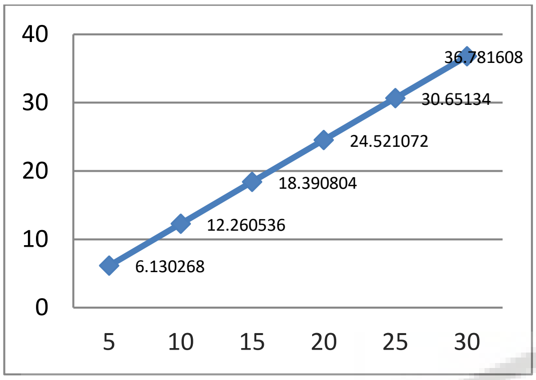

Figure 1. Non Carcinogenic Exposure of Ingestion

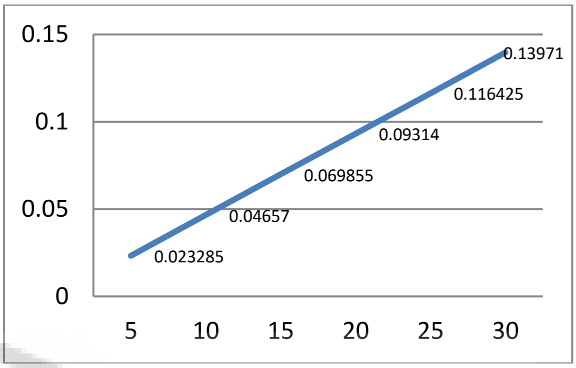

Figure 2. Non Carcinogenic Exposure of Inhalation

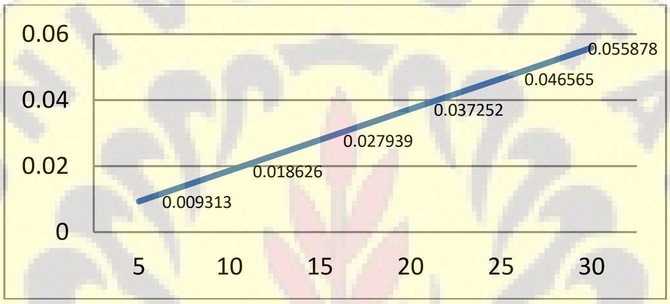

Figure 3. Carcinogenic Exposure of Inhalation

3.3 Risk Quotient (RQ) and Excess Cancer Risk (ECR) of $CaCO_3$ and TSP Exposure On Population

3.3.1 Risk Quotient of CaCO3

$$RQ=\frac{I}{RfD/DNEL}=\frac{6{,}130268\,\frac{mg}{kg}\,day}{6,\ 1\ \frac{mg}{kg}\,day}=1{,}004962$$

From the calculation results can be concluded that the exposure of CaCO3 of 160 mg/L ingestion in the population living in Grenden limestone for 52.2 kg body weight are not safe for frequency exposure 365 days/year .

3.3.2 Risk Quotient of TSP

$$RQ=\frac{I}{RfC}=\frac{0{,}023285\,\frac{mg}{kg}\,day}{2{,}42\ \frac{mg}{kg}\,day}=0{,}009622$$

From the calculation results can be concluded that the exposure of TSP of 0.06367 mg/m^3 ingestion in the population living in Grenden limestone area for 52.2 kg body weight is safe for frequency exposure 365 days/year .

3.3.3 Excess Cancer Risk of TSP

$$\text{ECR=I x SF= } 0{,}009313\,\frac{\text{mg}}{\text{kg}}\text{day x 1,1 kg day/mg=0,010245}$$

From the calculation results can be concluded that the exposure of TSP of 0.06367 mg/m^3 ingestion in the population living in Grenden limestone area for 52.2 kg body weight is not safe for frequency exposure 365 days/year.

4.0 Discussion

4.1 CaCO3 and TSP Concentration

There was one sample (number 10) which had CaCO3 concentration exceed the threshold value. The water samples can be classified as hard (100-200 ppm) and very hard (> 200 ppm)[14] , wherein there were 19 samples hard water (59.375%) and 13 samples very hard water (40.625%). This condition was due to the type of material that composed the stone in Grenden dominated with dolomite (CaMg(CO3)2) and calcite (CaCO3). The type of stone had a significant influence on water resources.[15]

According to montgomerry (2003), one of physical properties of minerals is the hardness of which was calcite has hardness scale of 3 from the range 1 to 10. Beside that, the water hardness level also affected with the depth of the wells. The depth of local wells ranged from 4 meters to 12 meters which is included in shallow wells. Water in shallow well have a lower quality compared with deep well. The calcium consumption exceed from the threshold will caused coorectal cancer, hipertension, stroke, coronary artery, and also kidney stone.

The result of air samples observation showed that the value below the threshold according to East Java Goverment regulation number 10/2009. The low measurement results probably affected by the air temperature, when the temperature was quite high, the air becomes tenuous so the concentration of the pollution was lower. In addition, the high TSP concentration in other limestone location probably coming from roads or construction areas.[16]. Our measurement point was determined from the distance from the limestone hill which was not near the limestone industry and highway.

The low TSP concentration in ambient air of Grenden limestone area did not mean it had no health impact. TSP concentration less than the threshold value basically can cause environment impat because of the formation of nucleus condensation that can be cluded with water and clouds.[17] In America and Britain, low particulate levels have also been able to cause health effects both in the long and short term[18]. The health effects that caused by particles are have a different levels and types, depending on the type, size and chemical composition. Dust particles air pollution usuallu cause chronic resporatory diseases such as chronic bronchitis, pulmonary emphysema, bronchial asthma and lung cancer.

4.2 Intake Assessment

Ink of ingestion	$6{,}130268 \frac{mg}{kg}$ day
Ink of inhalation	$0{,}023285 \frac{mg}{kg}$ day
Ik of inhalation	$0{,}009313 \frac{mg}{kg}$ day

According to the results, the population exposed to CaCO3 through ingestion 6.130268 mg/kg day.[19] When compared with DNEL values, the value of Ink of ingestion slightly exceeds the DNEL value. This shows that the Ink of population living in Grenden limestone area had exceeded the allowable intake of CaCO3 so that it may pose any risk or health impact.

The Ink of TSP result showed that the population was exposed to TSP daily by inhalation in 0.023285 mg/kg day. When compared with the RfC value, the TSP received every day by the population was still within safe limits. While, the Ik of TSP result means that the population was exposed to TSP daily through inhalation in 0.009313 mg/kg day.

4.3 Risk Quotient (RQ) and Excess Cancer Risk (ECR) of $CaCO_3$ and TSP Exposure On Population

RQ of CaCO3	1,004962
RQ of TSP	0,009622
ECR of TSP	0,010245

The calculation of RQ of CaCO3 result showed that the population were at risk where may cause several diseases such as kidney stone, coorectal cancer, hipertension, stroke and coronary arthery, depend on the exposure that individual received. Beside that, cardiovascular disease may cause by the water hardness in the drinking water, based on the England, Scotland and Wales study. While, the RQ of TSP showed that the population weren't at risk, although some respiration disease may cause by the low TSP concentration (below the threshold). The health impact may accute or chonic depend on the individual condition.[18] In some area of Indonesia study, such as Tulung Agung, Jepara, Tegal, Cirebon and Sukabumi that have limestone area were at risk and can cause the health impact.[10] The ECR of TSP calculation showed that the population were at cancer risk. TSP may cause cancer depend on the particle size.[20] In some study cancer risk happened on the person that have more time exposure and direct contact with $CaCO_3$.[21]

5.0 Conclusion and Recommendation

The exposure of $CaCO_3$ in the Grenden Vilage, East Java, Indonesia has health risk of non carcinogenic through ingestion and carcinogenic through inhalation. This risk can be prevented by decreasing the CaCO3 and TSP concentration until the safe limit, which is

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159,21 mg/L for CaCO3 concentration in well water and 0.000621 mg/m^3 for TSP concentration in ambient air. The process of lime soda can be done to reduce the water hardness by adding soda ash and lime into the wells. In addition to this process, hardness can also be reduced by ion exchange processess which can be done using zeolite. While the TSP concentration can be reduce by using air filter in the industrial area. Beside that, the ingestion rate must be decreased until 1.990125 litre/day and the duration of exposure until 0.27331 year. The local goverment should provide clean water facilities to reduce ingestion rate and duration of exposure.

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