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ORIGINAL RESEARCH

# Identification ratio of Si/Ti and Ca/Ti content by X-ray fluorescence in tsunami soil samples

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***Abstract.*** Earthquakes and tsunamis natural disasters have repeatedly occurred on the coast of Aceh province, which lies between the confluence of two plates. The tsunami deposits in this area can provide important information regarding the reconstruction of marine attacks by past earthquakes and tsunamis. In general, tsunami deposits can be identified based on their geological, sedimentological, paleontological, and geochemical characteristics. In our research work, spectrometry X-ray fluorescence (XRF) has been utilized to investigate the geochemical signatures of tsunami-affected soil samples in Aceh province at three tsunami-impacted areas, namely Aceh Besar regency, Banda Aceh City, and Aceh Barat regency. The sampling point is located about a kilometer from the coastal line. Our findings indicate that tsunami-affected soils in Aceh Province after 10 years struck by tsunami contain terrestrial markers such as Fe and Ti, carbonate markers (Mg, Ca), and heavy metals elements (Cr, Ni, Cu, Zn, and Sr). On the other hand, in our study, the concentration ratios of several elements such as Si/Ti and Ca/Ti seem most suitable as a chemical signature for differentiating environmental conditions such as the 2004 Indian Ocean tsunami event. It could be noticed that geochemical analysis by XRF can be applied to characterize the tsunami-affected soils in several coastal areas of Aceh province.

**Keywords**: Aceh-tsunami, soils, chemical signatures, XRF technique

## INTRODUCTION

The 2004 Indian Ocean tsunami triggered by an earthquake was the most destructive natural disaster in recorded history. In addition to claiming millions of lives, the Indian Ocean tsunami disaster also had a serious impact on people living in coastal communities and lowlands areas, such as groundwater pollution and chemical components contamination. Based on the environmental aspect, the damages from these natural disasters have implied that the protection against earthquake and tsunami potential was inadequate [1,2]. Therefore, to improve the preparedness and disaster mitigation of coastal communities, in-depth knowledge is needed for tsunami hazard assessments such as potential tsunami inundation areas and tsunami recurrence. There are various indicators have been used for the identification of marine incursion namely geomorphology, sedimentology, stratigraphy, and palaeontology. However, these parameters could not be applied to identify the tsunami-impacted areas that have low concentrations of microfossils. On the other hand, geochemical analysis has been widely recognized in tsunami research for three decades, specifically the ability of geochemistry to investigate modern and prehistoric tsunami deposits. Research on geochemical analysis and assessment of soil and sediments has been done for various purposes to study [3,4,5]. A study reported that the geochemical indicator of tsunami events was able to predict the ocean background that occurred over 1000 years ago [6,7,8]. Based on the literature previously, geochemical indicators have been accepted as a prospective and susceptive method for evaluating tsunami deposits [9,10]. Previous research reported that the soil sample inundated by the 2004 Indian Ocean tsunami in Thailand has been polluted by salt elements (Na, K, Ca, Mg, Cl, SO42-) and heavy metal elements (Cd, Cr, Ni, Cu, Zn, Pb, and As) [11,12]. Furthermore, a similar investigation was also conducted in Banda Aceh city, the impacted soil sample was contaminated by heavy metals (Cr, Fe, Co, Ni, Cu, Zn, Mn, Cd, Pb) and salt components (K, Ca, Mg, Li, Na), even after 3.5 years of bioremediation. Moreover, it could be noticed that the tsunami-impacted soil collected from Banda Aceh city 10 years after the tsunami event was still contaminated by Ca, Mg, Na, and K elements [13,14]. On the other hand, a study revealed that tsunami deposits in soil samples collected from Lambada regency, Aceh Besar have occurred SiO2 (55%), Fe2O3 (19%), Al2O3 (12%), CaO (9%), K2O (3%), and TiO2 (2%), and the composition of these minerals is the same as from ash that ejected from the Mountain of Merapi, Yogyakarta [15,16]. This phenomenon implies that periodical monitoring of chemical element contamination in tsunami-impacted soil samples needs to be carried out for not to endanger human health and well-being [17,18,19,20]. On the other hand, the chemical pollution inspection of soil samples struck by tsunami events could be utilized for studying and extending historical records, investigating the process of bioremediation, comparison of past tsunamis, and as geochemical signatures for evaluating tsunami events in Aceh province [5]. The principal of our research work is to reveal the effect of the tsunami seawater inundation which occurred in 2004 on the chemical properties of soil in several stricken areas in Aceh province after 10 years caused by the massive Indian Ocean tsunami event. It is expected this research could provide knowledge to enhance the bioremediation process, as well as contribute to the revitalization of affected agricultural land for plants and crop production in the affected area [21,22,23,24,25,26]. According to the previous study, various analytical instruments could be applied for determining and quantifying the chemical elements in soil samples involving Inductively Coupled Plasma-Mass Spectrometry (ICP-MS), Flame Atomic Absorption Spectrometry (FASS), Graphite Furnace Atomic Absorption Spectrometry (GF-AAS), and Atomic Fluorescence Spectrometry (AFS). Nevertheless, these techniques mostly require strong acid as a solvent, complex preparation sample, washing time and energy, and are non-environmentally friendly. To overcome these problems, X-ray fluorescence (XRF) could be utilized for the identification of chemical elements in tsunami-stricken areas because it showed many advantages, such as being cheap, versatile, simple, safe, non-destructive analysis, and no need for routine re-calibration. In addition, XRF technique could be used for quantifying the minor elements with concentrations of 10-20 ppm and having high accurate analyses of a range of elements.

## METHODOLOGY

### Study Site and Soil Sampling

Aceh province is recorded as the worst destructive area of the Indian Ocean tsunami in 2004. This event has caused seawater to inundate the land 5–6 km inland and leave a tsunami deposit with a thickness of 0.5–1 m. This

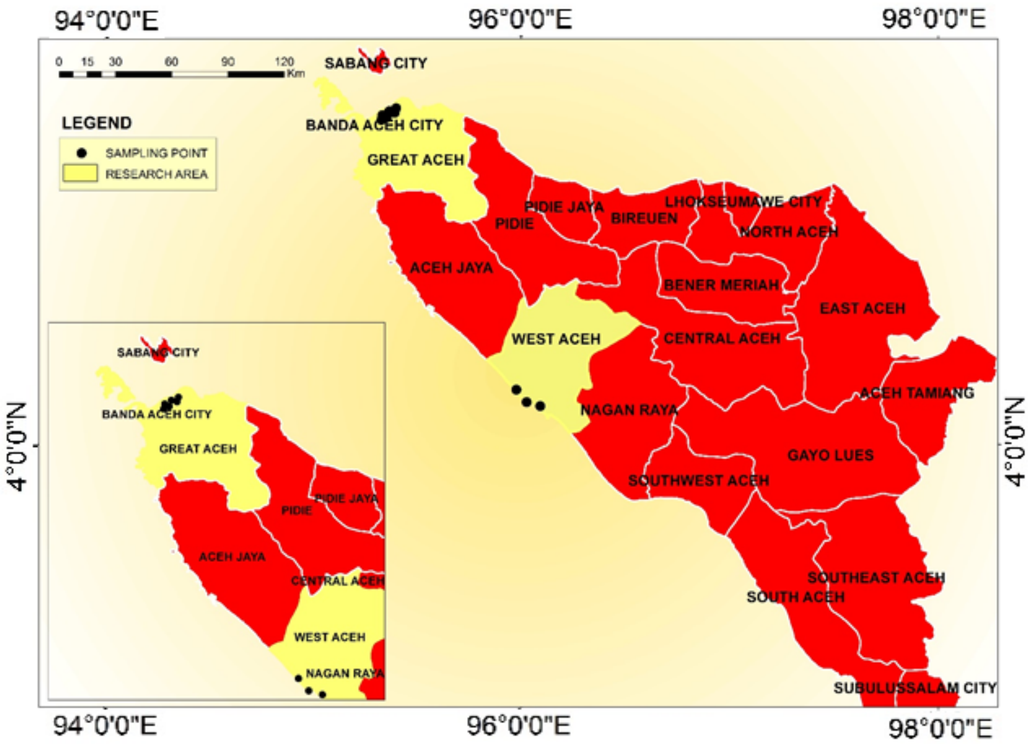


**Figure 1.** The sampling points of soil samples were impacted by the massive Indian Ocean tsunami in 2004 in Aceh province, Indonesia. The sampling regions are highlighted in yellow color, while the sampling points are highlighted in black color.

investigation was conducted at three tsunami-impacted areas in Aceh province, namely Aceh Besar Regency, Banda Aceh City, and Aceh Barat Regency. The sampling point is located about a kilometer from the coastal line. The tsunami-impacted soil samples were collected about 10 years after the massive Indian Ocean tsunami happened in Aceh province. The tsunami-affected soil samples were taken in areas that have a sunken morphology such as rice fields and swamps, are well-maintained areas, and are protected from coastal waves. It can be seen in Figure 1. that these tsunami-stricken areas, such as Aceh Besar, Banda Aceh City, and Aceh Barat are appearing in front of the sea, so it is assumed these areas are the worst impacted by the 2004 Indian Ocean tsunami. For the experimental process, about 5 cm of the surface of soil samples was removed. Figure 1 demonstrates the map of the sampling location of soil samples struck by the 2004 Indian Ocean giant tsunami in Aceh Province, Indonesia. The sampling regions are highlighted in yellow color, while the sampling points are highlighted in black color.

**Soil Chemical Characterization**

The investigation of chemical composition in tsunami-stricken soil samples was carried out using Thermo Scientific Portable X-Ray Fluorescence Analyzer. The soil samples initially were heated in the oven at 65°C for four days to release the water content in the samples below 20%. The content of water in the sample of more than 20% could affect the XRF analysis and change the composition of matrix samples. To realize this research goal, the soil samples were dried by dividing them into aggregates and dispersing them on polyethylene plywood or paper in the open air. During the preparation of this process, it can be predicted that there is no sample contamination from outer sources. Furthermore, to minimize the effects of the soil matrix, those soil samples were mashed and sieved to obtain homogeneous particle sizes of about 75 μm.

## RESULTS AND DISCUSSION

The composition of chemical elements in soil samples taken from tsunami-stricken areas in Aceh province (Banda Aceh, Aceh Besar, and Aceh Barat) was demonstrated in Figure 2. The soil samples were analyzed using XRF. Based on the XRF peaks, the soil samples collected from three stricken areas in Aceh province were contaminated by alkali metal elements identified as salt elements (Ca and K) and transition metal elements involving metals and heavy metal (Al, Si, P, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, and Sr). Table 1-3 shows a data set of the chemical element concentrations in soil samples from the three impacted regions that are previously mentioned. The soil samples collected from Aceh Besar had higher concentrations of Ca and K, as salt elements, compared to the other stricken regions. The maximum concentration of Ca mostly appeared in a sample from Aceh Besar 1 with a value of 15.3 ± 0.6 %, and the minimum content of Ca was in a sample from Aceh Barat 1 (8.31 ± 0.39 %).

Meanwhile, the level of K element was identified as more than 4% in all stricken soils collected from Aceh Besar regency. Fe element, on the other hand, dominantly appeared in soil samples taken from Banda Aceh region, more than 35%. The highest concentration of Fe element was found in Banda Aceh 2 with a value of 48.19 ± 0.29 %, then followed by Banda Aceh 1 (39.7 ± 0.2 %) and Banda Aceh 3 (35.4 ± 0.2 %). The lowest concentration of Fe element was obtained in a soil sample from Aceh Besar 3 (30.2 ± 0.3 %). Each tsunami-stricken area also showed different characteristics associated with other transition elements such as Al, Si, Ti, V, Cr, and Zn. The soil sample in Banda Aceh 1 had a large concentration of Al element with a value of 9.1 ± 0.04 %. and the minimum content of Al was detected in Banda Aceh 2 (6.0 ± 0.08 %). The maximum content of Si elements was obtained in tsunami-stricken soil from Aceh Barat, it is more than 35%. The maximum value of Si element was found in the sample from Aceh Barat 3 (38.6 ± 0.3 %), and the content of Si was low in a sample from Banda Aceh 2 (23.3 ± 0.3 %). Ti had appeared more than 2% in all collected soil samples. The content of Ti in Banda Aceh 2 (4.47 ± 0.04 %) was much higher than others. The minimum value was noted in a sample from Aceh Barat 2 (2.06 ± 0.03 %). The soil samples taken from Banda Aceh 2 had a large content of V (0.23 ± 0.04 %) and Cr (0.56 ± 0.009%). Further, the impacted soil samples in Aceh Besar 1 have a maximum value of Zn element (0.35 ± 0.04 %). The minimum content of Zn is found in Aceh Barat 3 (0.09 ± 0.01).

Furthermore, using Table 4, the ratio of miscellaneous elements obtained included Ca/Ti, Ca/K, Si/Ti, and Si/Al. All those areas affected by the tsunami resulted in the highest ratio of Ca/Ti with a value of 6:1 at the location of Aceh Barat 2. Meanwhile, the lowest Ca/Ti ratio was found in the soil sample taken in Banda Aceh 2 with a value of 2: 1. The Ca/K concentration ratio produced in this study resulted in a comparison with the highest value of 4:1 for soil samples taken from Banda Aceh 2, and the lowest at locations with a value of 2:1 at Banda Aceh 1, Aceh. Besar 2, Aceh Barat 1 and 3. The value of the Si/Al ratio in all tsunami-affected soil samples in Aceh province resulted in the highest comparison with a value of 5:1. Moreover, the Si/Ti ratio produced a large value compared to the ratio of the concentrations of other elements. The highest Si/Ti ratio was found in the soil sample taken in Aceh Barat 2 with a value of 18: 1, while the lowest Si/Ti ratio was found in the sample taken in Banda Aceh 2 with a value of 5: 1.

According to the previous research, it was found that the concentration of chemical elements in tsunami-stricken areas was greater than in unimpacted areas, even after 10 years of tsunami events [14,15,16].

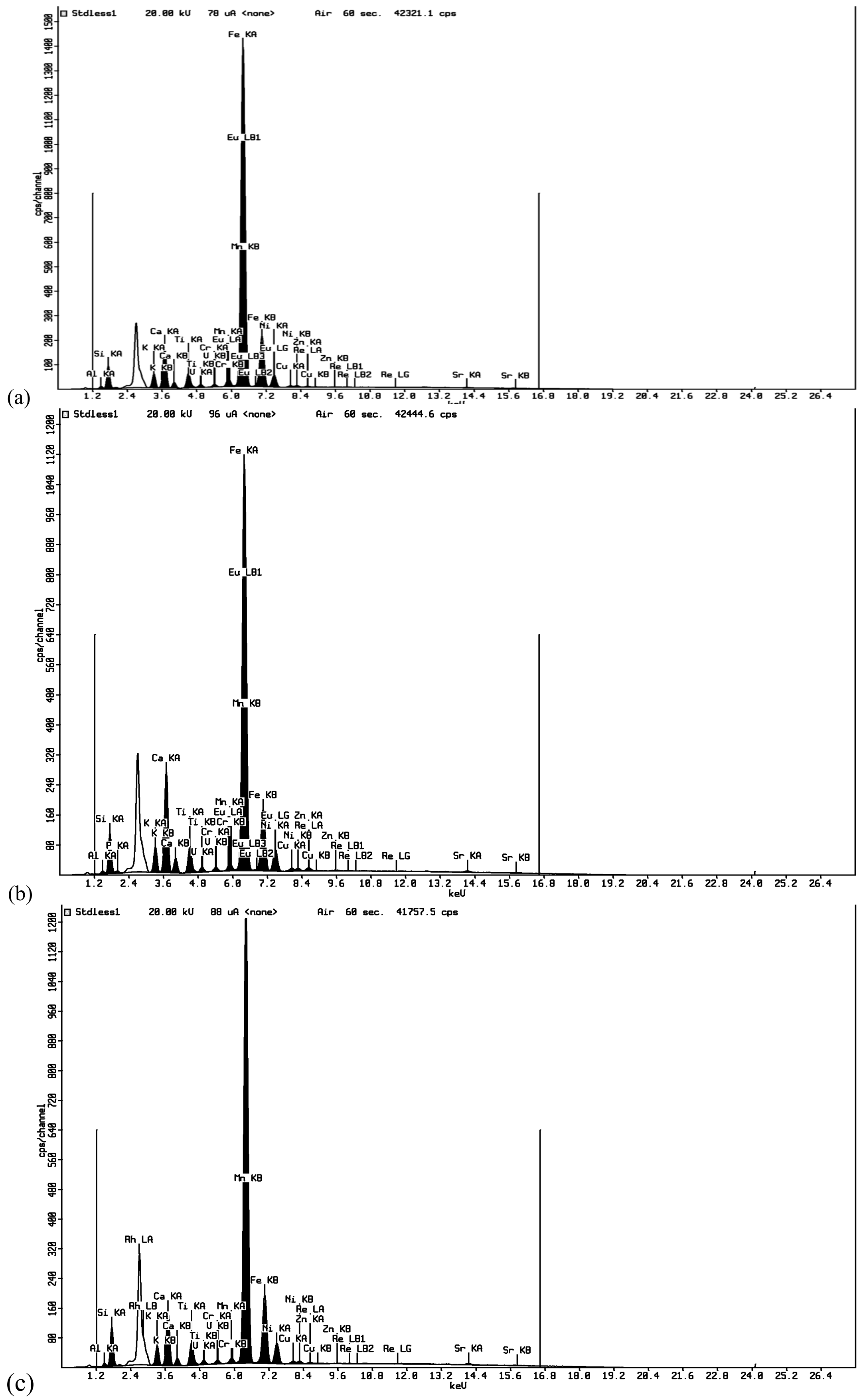


**Figure 2.** The XRF peaks of tsunami affected soil samples collected from (a) Banda Aceh, (b) Aceh Besar, and (c)Aceh Barat.

**Table 1.** The concentration of chemical elements in soil samples collected from Banda Aceh city after 10 years of tsunami 2004 using XRD

| Element | Concentration (%) | | |
|---|---|---|---|
| | **Banda Aceh 1 (Pineung)** | **Banda Aceh 2 (lamgugop)** | **Banda Aceh 3 (Ie Masen)** |
| Al | 9.1 ± 0.04 | 6.0 ± 0.08 | 8.1 ± 0.1 |
| Si | 31.9 ± 0.2 | 23.3 ± 0.3 | 34.3 ± 0.3 |
| K | 3.77 ± 0.02 | 2.32 ± 0.02 | 3.97 ± 0.03 |
| P | - | 0.48 ± 0.04 | 0.59 0.01 |
| Ca | 8.49 ± 0.05 | 10.8 ± 0.05 | 10.9 ± 0.02 |
| Ti | 2.10 ± 0.01 | 4.47 ± 0.04 | 2.69 ± 0.04 |
| V | 0.12 ± 0.02 | 0.23 ± 0.04 | 0.15 ± 0.01 |
| Cr | 0.26 ± 0.006 | 0.56 ± 0.009 | 0.27 ± 0.001 |
| Mn | 0.82 ± 0.006 | 0.73 ± 0.004 | 0.40 ± 0.01 |
| Fe | 39.7 ± 0.2 | 48.19 ± 0.29 | 35.4 ± 0.2 |
| Ni | 1.96 ± 0.03 | 1.56 ± 0.02 | 1.60 ± 0.02 |
| Cu | 0.26 ± 0.01 | 0.22 ± 0.0005 | 0.26 ± 0.07 |
| Zn | 0.10 ± 0.0008 | 0.23 ± 0.008 | 0.18 ± 0.004 |
| Sr | 0.76 ± 0.06 | 0.64 ± 0.02 | 0.82 ± 0.03 |

**Table 2.** The concentration of chemical elements in soil samples collected from Aceh Besar regency after 10 years of tsunami 2004 using XRD

| Element | Concentration (%) | | |
|---|---|---|---|
| | **Aceh Besar 1 (Miruek Taman)** | **Aceh Besar 2 (Miruek Taman)** | **Aceh Besar 3 (Miruek Taman)** |
| Al | 6.8 ± 0.1 | 7.8 ± 0.2 | 8.3 ± 0.2 |
| Si | 33.4 ± 0.2 | 35.4 ± 0.4 | 37.4 ± 0.5 |
| K | 4.54 ± 0.19 | 4.81 ± 0.01 | 4.76 ± 0.04 |
| P | 0.91 ± 0.04 | 0.62 ± 0.0096 | 0.67 ± 0.05 |
| Ca | 15.3 ± 0.6 | 11.0 ± 0.03 | 12.2 ± 0.06 |
| Ti | 2.83 ± 0.18 | 2.75 ± 0.01 | 2.27 ± 0.02 |
| V | 0.13 ± 0.03 | 0.14 ± 0.007 | 0.12 ± 0.01 |
| Cr | 0.30 ± 0.02 | 0.26 ± 0.003 | 0.29 ± 0.002 |
| Mn | 0.53 ± 0.02 | 0.64 ± 0.02 | 0.40 ± 0.03 |
| Fe | 32.9 ± 2.3 | 33.5 ± 0.2 | 30.2 ± 0.3 |
| Ni | 2.27 ± 0.19 | 1.55 ± 0.01 | 1.91 ± 0.04 |
| Cu | 0.31 ± 0.02 | 0.27 ± 0.005 | 0.25 ± 0.002 |
| Zn | 0.35 ± 0.04 | 0.16 ± 0.01 | 0.10 ± 0.005 |
| Sr | 1.1 ± 0.09 | 0.83 ± 0.02 | 0.89 ± 0.04 |

**Table 3.** The concentration of chemical elements in soil samples collected from Aceh Barat regency after 10 years of tsunami 2004 using XRD

| Element | Concentration (%) | | |
|---|---|---|---|
| | **Aceh Barat 1 (Lhok Bubon)** | **Aceh Barat 2 (Lhok Bubon)** | **Aceh Barat 3 (Kuala Bubon)** |
| Al | 8.2 ± 0.03 | 8.2 ± 0.01 | 8.7 ± 0.3 |
| Si | 37.8 ± 0.7 | 37.4 ± 0.2 | 38.6 ± 0.3 |
| K | 3.76 ± 0.304 | 4.38 ± 0.03 | 4.98 ± 0.15 |
| P | - | - | - |
| Ca | 8.31 ± 0.39 | 11.8 ± 0.1 | 8.91 ± 0.19 |
| Ti | 2.35 ± 0.14 | 2.06 ± 0.03 | 2.27 ± 0.04 |
| V | 0.11 ± 0.02 | 0.12 ± 0.008 | 0.11 ± 0.03 |
| Cr | 0.27 ± 0.01 | 0.29 ± 0.006 | 0.24 ± 0.002 |
| Mn | 0.40 ± 0.03 | 0.46 ± 0.006 | 0.35 ± 0.04 |
| Fe | 37.6 ± 2.9 | 31.8 ± 0.2 | 31.9 ± 0.4 |
| Ni | 2.27 ± 0.196 | 1.80 ± 0.01 | 1.95 ± 0.04 |
| Cu | 0.31 ± 0.03 | 0.25 ± 0.007 | 0.28 ± 0.01 |
| Zn | 0.1 ± 0.01 | 0.07 ± 0.009 | 0.09 ± 0.01 |
| Sr | 0.97 ± 0.06 | 0.91 ± 0.02 | 0.99 ± 0.04 |

Tsunami seawater inundation has brought marine sediments involving extraordinary chemical elements, resuspended, particulate, then deposited on land in the long-term during tsunami seawater backwash. During this period, the quality of soil has been changing, increasing salt and heavy metal elements, and leaching of marine materials. In this research, we used X-ray fluorescence (XRF) spectrometry as an analysis tool to

**Table 4.** Ratio of elemental concentration of tsunami impacted soil samples to the chemical signatures

| Locations | Ratio of Elements | | | |
|---|---|---|---|---|
| | **Ca/Ti** | **Ca/K** | **Si/Ti** | **Si/Al** |
| Banda Aceh 1 | 4 : 1 | 2 : 1 | 15 : 1 | 4 : 1 |
| Banda Aceh 2 | 2 : 1 | 4 : 1 | 5 : 1 | 4 : 1 |
| Banda Aceh 3 | 3 : 1 | 3 : 1 | 13 : 1 | 4 : 1 |
| Aceh Besar 1 | 5 : 1 | 3 ; 1 | 12 : 1 | 5 : 1 |
| Aceh Besar 2 | 4 : 1 | 2 : 1 | 13 : 1 | 5 : 1 |
| Aceh Besar 3 | 5 : 1 | 3 : 1 | 16 : 1 | 5 : 1 |
| Aceh Barat 1 | 4 : 1 | 2 : 1 | 16 : 1 | 5 : 1 |
| Aceh Barat 2 | 6 : 1 | 3 : 1 | 18 : 1 | 4 : 1 |
| Aceh Barat 3 | 4 : 1 | 2 : 1 | 17 : 1 | 4 : 1 |

identify the chemical components in soil samples of tsunami-stricken areas. X-ray fluorescence (XRF) spectrometry is well-known as analysis equipment for identifying the chemical components in various types of samples in the liquid and solid phases. In this system work, an X-ray beam is targeted onto the surface of the soil sample to produce new X-rays. The resulting new X-rays are captured by a detector. Then, the resulting spectrum demonstrated the characteristics of chemical elements composition from the material. We have successfully analyzed the soils of tsunami-stricken land using XRF. Regarding the previous study conducted by Ref [11,13], the composition of chemical elements that resulted in this research work has a similar tendency compared to the soil samples taken from several regions in Aceh province after being struck by the Indian Ocean tsunami 2004. Similar marks could be seen from the elevation of some element concentrations such as Ca and K in tsunami-stricken soils. The area in Banda Aceh had a relatively lower concentration of K element compared to Aceh Besar and Aceh Barat.

The research already stated that the impact of the tsunami in Banda Aceh was terrible, but then they also added that Aceh Barat was the closest region to the Indian Ocean earthquake epicenter that happened in 2004 [7]. Based on the propagation of the tsunami drawn by Ref. [27,28], it can be found that the tsunami first hit Aceh Barat, then continued to Aceh Besar, and then headed to Banda Aceh. So, it can be assumed that the further the location from the earthquake epicenter, the lower the concentration of salt element can be obtained. Nevertheless, the chemical composition of post-deposition could be significantly changed, so that the tsunami deposits from seafloor and land are sometimes difficult to distinguish. Data on single elements in soils sometimes does not provide a clear conclusion, such as the Ca element concentration pattern which messed up the relationship among the data (no Ca concentration gradient is formed from Aceh Barat, Aceh Besar, and Banda Aceh). Previous experiments conducted by Ref. [29,30,31] applied the different elemental concentration ratios in their publications for evaluating the geochemical signature of tsunami events. Several chemical element ratios that had been widely applied for investigating the geochemical signature of tsunami incidents were Na/Ti, Ca/Ti, and Sr/Ba. An increase in the composition of Na element in tsunami-impacted soil samples could be predicted by the deposition of sea salt and feldspar minerals in the soil. A high ratio of Na/Ti was revealed as a good indicator to investigate the influence of tsunami seawater [32]. A geochemical proxy of Na/Ti was found as a good parameter for analyzing the chemical signature of modern and prehistoric tsunami deposits in Tohoku. However, in this work, the ratio of the Na/Ti element could not be obtained because the Na element was not identified. That was presumably due to the high rainfall during sample collection and the high solubility of Na element in the water.

The ratio of Ca/Ti has been widely employed for identifying the chemical signature in marine versus terrestrial environments. The availability of Ca components in coastal systems comes from calcium carbonate (CaCO3) which is produced by shell animals such as ostracods, foraminifera, or gastropods. Regarding previous literature, a high ratio value of Ca/Ti was found to be a good indicator to investigate the chemical signature of a tsunami in Lavericks Bay, New Zealand [33], Kyparissia Bay, Greece, and various prehistoric tsunamis, and to identify the marine versus terrestrial deposits [34]. In this research, Ca/Ti could be used to distinguish the condition of tsunami-stricken areas. It was expressed by the appearance of the Ca/Ti ratio gradient in Banda Aceh, Aceh Besar, and Aceh Barat. Besides that, the high value of the Si/Ti on all tsunami-affected soils could also be used as an indicator to identify the chemical signature of tsunami-affected areas in Aceh province. The Si/Ti ratio had a higher value to distinguish the tsunami-stricken areas rather than Ca/Ti because of a higher range of comparisons. Although Ca/K and Si/Al were already obtained, there was no additional information to retrieve. This still needed to be studied further.

## CONCLUSION

A study on geochemical analysis of soil samples struck by a tsunami in Aceh province using X-ray fluorescence technique has been done. The investigation revealed that the tsunami-affected soils collected in Banda Aceh city 10 years after the big Indian Ocean tsunami in 2004 were still traced by salt content and transition metal elements. All tsunami-affected soil samples taken in Aceh Province were dominated by Fe, Si, and Ca elements, so they could be used as chemical markers to identify tsunami-affected soils in Aceh Province 10 years after the tsunami. Based on the number of chemical elements that can be detected and the high value of the Si/Ti ratio obtained, it could be noticed that the XRF technique is a fast, prospective, and efficient analytical instrument to be applied as a method for investigating chemical markers of tsunami-affected soil samples in the province of Aceh, Indonesia.

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