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# Collateral variations between the concentrations of mercury and other water soluble ions in volcanic ash samples and volcanic activity during the 2014–2016 eruptive episodes at Aso volcano, Japan

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## ABSTRACT

During 2014–2016, the Aso volcano, located in the center of the Kyushu Islands, Japan, erupted and emitted large amounts of volcanic gases and ash. Two episodes of the eruption were observed; firstly Strombolian magmatic eruptive episodes from 25 November 2014 to the middle of May 2015, and secondly phreatomagmatic and phreatic eruptive episodes from September 2015 to February 2016. Bulk chemical analyses on total mercury (Hg) and major ions in water soluble fraction in volcanic ash fall samples were conducted. During the Strombolian magmatic eruptive episodes, total Hg concentrations averaged  $1.69 \pm 0.87 \text{ ng g}^{-1}$  ( $N = 33$ ), with a range from 0.47 to  $3.8 \text{ ng g}^{-1}$ . In addition, the temporal variation of total Hg concentrations in volcanic ash varied with the amplitude change of seismic signals. In the Aso volcano, the volcanic tremors are always observed during eruptive stages and quiet interludes, and the amplitudes of tremors increase at eruptive stages. So, the temporal variation of total Hg concentrations could provide an indication of the level of volcanic activity. During the phreatomagmatic and phreatic eruptive episodes, on the other hand, total Hg concentrations in the volcanic ash fall samples averaged  $220 \pm 88 \text{ ng g}^{-1}$  ( $N = 5$ ), corresponding to 100 times higher than those during the Strombolian eruptive episode. Therefore, it is possible that total Hg concentrations in volcanic ash samples are largely varied depending on the eruptive type. In addition, the ash fall amounts were also largely different among the two eruptive episodes. This can be also one of the factors controlling Hg concentrations in volcanic ash.

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## 1. Introduction

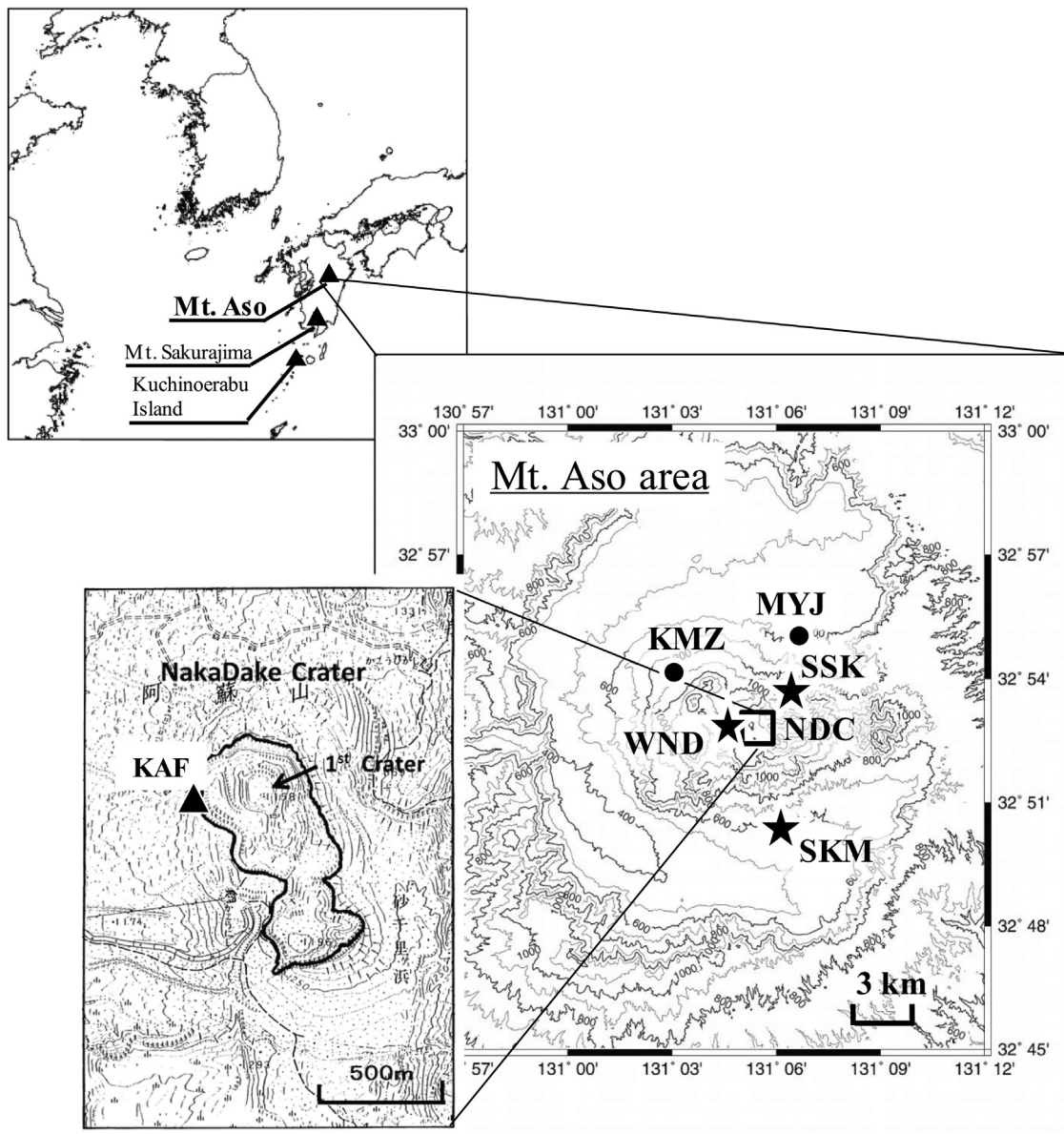
Mercury (Hg) is emitted from anthropogenic emission sources including coal and oil combustion, metal production and gold mining. In addition, Hg is also emitted from volcanic activity, direct emission from sea areas and soil and forest surfaces and by biomass burning (Nriagu, 1989; Schroeder and Munthe, 1998). The emission into the atmosphere was estimated from 6000 to 9000 tons  $\text{yr}^{-1}$  (UNEP, 2013). From the results of the calculation using the recent global models, almost all Hg currently emitted from sea, land and forest is derived from legacy anthropogenic Hg (Amos et al., 2013). In addition, biomass burning is caused by human activity and natural factors such as severe dryness and thunderbolts. Therefore, active volcanoes are the only natural sources of direct mercury emission into the atmosphere (Bagnato et al., 2007, 2010; Nriagu and Becker, 2003; Pyle and Mather, 2003; Siegel and Siegel, 1975; Varekamp and Buseck, 1986). The amount of Hg released into the atmosphere, or deposited on land and sea areas has been reported for many volcanic areas, such as Mt. Etna (Barghigiana et al., 1988;

Dedeurwaerder et al., 1982; Ferrara and Masertia, 1990; Ferrara et al., 2000; Martin et al., 2007), the central American volcanic areas (Carr and Rose, 1987; Haynes, 2012; Moya and DiPippo, 2007; Witt et al., 2008), Sakurajima volcano (Ando et al., 2010; Ohki et al., 2016; Tomiyasu et al., 2003), the south America volcanic areas (Guevara et al., 2010), and La Palma-Canary Islands (Martin et al., 2013). In the soil and water near volcanoes and in glacial ice cores, the historical volcanic events are indicated as the principal natural sources of reactive and particulate Hg (Nriagu and Becker, 2003; Barghigiana et al., 1988; Ferrara and Masertia, 1990; Haynes, 2012; Krabbenhoft and Schuste, 2002). Although there are many papers and articles on Hg emitted from volcanic areas, the current and temporal variation of Hg concentrations in gases or ash during one volcano active stages has not been reported.

In Japan, there are a lot of volcanic and geothermal areas and, in recent years, some volcanoes including the Kuchinoerabu Islands, the Sakurajima volcano and the Aso volcano have been particularly active (Fig. 1). The Aso volcano is situated in the centre of the Kyushu Island, Japan and comprises the Aso caldera and post-caldera central cones. The only active cone, Nakadake (1506 m a.s.l.) composed of basaltic andesite to basalt, is one of the most active volcanoes in Japan. The active crater of Nakadake is a composite of seven craterlets aligned N-S. Its

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**Fig. 1.** Location of the sampling sites for volcanic ashes and the seismic station KAF, as shown with solid triangle, in the Aso caldera. The active crater (Nakadake No.1 crater, NDC) is shown with the open square. The weekly or daily sampling sites of volcanic ash are shown with solid stars, WND: West side of NDC, SKM: Shikimi located at SSE 5 km from NDC, and SSK: Sensuikyo located at NE 1 km from NDC. In consideration of wind direction, the volcanic ash sampling were also conducted at the other sampling sites including KMZ (Komezuka) and MYJ (Miyaji), as shown with solid circles.

northernmost craterlet No.1 (the 1st crater) has been active in the past 80 years, although some others were active before the 1933 eruption. During its calm periods, the Nakadake 1st crater is occupied by a hyper acidic hot crater lake (named Yudamari), >200 m in diameter, with temperature of 50–60 °C. During active periods, its volcanic activity is characterized by ash and Strombolian eruptions and phreatic or phreatomagmatic eruptions. The recent eruption activity at the Nakadake 1st crater began on 25 November 2014 with a Strombolian eruption, causing ash falls and volcanic gas emissions. During the following period of intermittent activity, from December 2014 to May 2015, the crater continued to spout out gas, steam, and small ash plumes. At the climax stage of the eruption, from the end of January to the beginning of February 2015, the scoria cone with a vent of 100 m diameter and the depth of 20 m was formed within the crater. From the middle of May to August 2015, the activity was represented by only emissions of gas and steam. Thereafter, the phreatomagmatic and phreatic eruptive episodes occurred from September 2015 to February 2016 and the ash were emitted sporadically (Fig. 2). The Strombolian

eruptions occurred at quasi-regular intervals explosion with the eruption columns of tens or hundreds of meters in height and are supposed to be caused by the generation and subsequent rise of large gas bubble slugs in a liquid magma conduit, often resulting in an explosion when reaching the surface (Zobin and Sudo, 2017). The amplitudes of seismic signals (volcanic earthquakes and tremors) increased around the times of the magmatic, phreatic-magmatic and phreatic eruptions.

In this paper, we focus on Hg in volcanic ash emitted from the 2014–2015 Strombolian eruption at the Aso volcano. The correlation between the amplitude change of seismic signals and the total Hg in volcanic ash are studied. Their correlation is obtained and discussed on the base of the seismic signals associated with the Strombolian explosions at the Aso volcano.

## 2. Nakadake 1st crater in Aso volcano and seismic signals

During a calm period, many tourists are able to easily look inside the active Nakadake 1st crater. At the weakly volcanic active stage, a white-

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