



Sulfur-rich geothermal emissions elevate acid aerosol levels in metropolitan Taipei[☆]

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ABSTRACT

Several studies have demonstrated that millions of people globally are potentially exposed to volcanic gases. Hydrogen sulfide is a typical gas in volcanic and geothermal areas. The gas is toxic at high concentrations that predominantly affects the nervous, cardiovascular, and respiratory systems. The WHO air quality guideline for hydrogen sulfide is $150 \mu\text{g m}^{-3}$ (105 ppb). The northwest part of Taipei is surrounded by sulfur-rich geothermal and hot springs. Active fumaroles and bubbling springs around the geothermal area emit acidic gases. In combination with automobile emissions, the pollution of acid aerosols is characteristic of the metropolis. This study considered sulfur-rich geothermal, suburban and downtown locations of this metropolis to evaluate geothermally emitted acid aerosol and H_2S pollution. Acid aerosols were collected using a honeycomb denuder filter pack sampling system (HDS), and then analyzed by ion chromatography (IC). Results indicated that long-term geothermal emissions, automobile emissions and photochemical reactions have led to significant variations in air pollution among regions of metropolitan Taipei. The highest H_2S concentration was 1705 ppb in the geothermal area with low traffic density and the mean concentration was 404.06 ppb, which was higher than WHO guideline and might cause eye irritation. The SO_2 concentrations were relatively low (mean concentration was 3.9 ppb) in this area. It may partially result from the chemical reduction reaction in the geothermal emission, which converted the SO_2 gas into SO_4^{2-} and H_2S . Consequently, very high sulfate concentrations (mean concentration higher than $25.0 \mu\text{g m}^{-3}$) were also observed in the area. The geothermal areas also emitted relatively high levels of aerosol acidity, Cl^- , F^- , PO_4^{3-} , and N-containing aerosols. As a result, concentrations of HNO_3 , NO_2^- , PO_4^{3-} , and SO_4^{2-} in metropolitan Taipei are significantly higher than those in other urban areas considered in other studies. These results provide evidence that geothermal emissions represent a significant source of acid aerosols in metropolitan Taipei. Therefore, in this metropolitan area, the combined health effects of air pollutants from automobiles and geothermal emissions should be particular concern.

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1. Introduction

Air pollution and its impact on human health have been considered a serious problem in active volcanic areas. Millions of people globally are potentially exposed to volcanic gases, the effects of exposure to which may differ from those of exposure to anthropogenic air pollution (Small and Naumann, 2001; Hansell et al., 2006). Toxic emissions materials from volcanic and geothermal areas include carbon dioxide (CO_2), sulfur dioxide (SO_2), hydrogen chloride (HCl), hydrogen fluoride (HF), hydrogen sulfide (H_2S), carbon monoxide (CO), radon (Rn) and various trace elements (Bernstein et al., 1986; Durand and Scott, 2005; Allen et al., 2000; Hansell, 2004; Carapezza et al., 2003). Unlike the

emissions of hazardous air pollutants during eruptions, ground gas emissions from geothermal fields are continuous, and thus, communities in these areas may be chronically exposed to elevated gas concentrations.

Hydrogen sulfide is a typical gas emission from geothermal fields and has significantly environmental impact. Several studies have been performed to measure the concentrations of H_2S from active volcanic and geothermal areas in the world (Vasilakos et al., 2005; Durand and Scott, 2005; Lee et al., 2005), and concentrations were above WHO guideline- $150 \mu\text{g m}^{-3}$ (WHO, 2000). The gas is toxic at high concentrations that predominantly affects the nervous, cardiovascular, and respiratory systems (ATSDR, 1999; WHO, 2003). The World Health Organization (WHO, 2000) indicated that H_2S would produce a unpleasant odor when the 30-min average ambient concentration is above $7 \mu\text{g m}^{-3}$. With exposure to exceed $150 \mu\text{g m}^{-3}$ H_2S (WHO guideline), eye irritation occurs. Between 210 and 350 mg m^{-3} , odor perception is lost due to what has been described as olfactory nerve paralysis.

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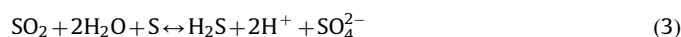
E-mail address: mlchen@ym.edu.tw (M.-L. Chen).

The concentrations exceeding 1400 mg m^{-3} may result in respiration paralysis and sudden death (WHO, 2000).

H_2S increases and become the dominant sulfur species in active volcanic and geothermal areas under the condition of higher pressure, shown as Eq. (1) (Lee et al., 2005; Lino et al., 2004).



On the other hand, SO_2 was reported little in the steam and hydrothermal water which were produced from the interaction between magmatic gas and groundwater (Ohba et al., 2010). During the reaction, SO_2 gas converted into SO_4^{2-} and a reduced form of S such as native S or H_2S via the following Eqs. (2)–(4) (Ho, 2001; Yang et al., 2003; Ohba et al., 2010).



Hydrogen sulfide may also be oxidized to sulfur dioxide but Kristmannsdóttir et al. (2000) thought the conversion of geothermally emitted H_2S to SO_2 in air being slow. Therefore, H_2S concentrations are always much higher than SO_2 in many volcanic and geothermal areas (Vasilako et al., 2005; Durand and Scott, 2005; Kristmannsdóttir et al., 2000; Lee et al., 2005; Ohba et al., 2010). In addition to gas emissions, geothermal sources also emit aerosol pollutants. Obviously, the composition of air pollution in geothermal areas is very complex.

Epidemiological studies have indicated that acid aerosol exposure from anthropogenic sources in urban environments may result in decrements in pulmonary function, increased hospital admissions for asthma and other respiratory ailments, and possibly excess mortality (Spengler et al., 1990; Raizene et al., 1996; Dockery et al., 1992, 1996). Acid aerosols are formed in the air through a series of complex chemical, photochemical, physical, and oxidation reactions (Potukuchi and Wexler, 1995). Most of the strong acid in ambient air is believed to be formed by the oxidation of sulfur dioxide to sulfate, through both homogenous

and heterogeneous processes (Tanner et al., 1981). A series of investigations have been performed to measure the concentrations of acid aerosols in urban, suburban, and rural areas (McCurdy et al., 1999; Lee et al., 1999; Matsumoto and Okita, 1998; Bari et al., 2003; Cheng et al., 2007; Wang et al., 2005; Walker et al., 2006; Plessow et al., 2005; Wu et al., 2009; Biswas et al., 2008). They have found that both the physical and the chemical characteristics of ambient aerosols in the air are determined not only by the rates of emission and chemical transformation, but also strongly by meteorological factors (Zelenka, 1997; Reisinger, 2000).

Taiwan is located in the Circum-Pacific seismic zone and experiences a high level of volcanic activity, resulting in a high density of hot springs (One hundred and forty areas of hot springs have been identified in Taiwan). The northwest part of metropolitan Taipei is surrounded by sulfur-rich geothermal and hot springs. We have previously found that the temperature of spring water in this area to be above 80°C and the pH to be 1.5–2.7 (Wang, 2003). The concentrations of chloride, sulfate, fluoride, nitrous and nitrate compounds in the spring water are in the ranges 26–3459, 75–2480, 0.8–25.6, 0.1–3.9 and 2.4–38.3 mg l^{-1} , respectively (Wang, 2003). Thus, the spring water is highly acidic and rich in chloride and sulfate. Concentrations of airborne H_2S and sulfate close to geothermal areas have been found to exceed 186 ppb and $10 \mu\text{g m}^{-3}$, respectively (Chen et al., 2003). Metropolitan Taipei is located in a subtropical zone. The weather is humid (an annual average humidity of 80%) and hot (an annual average temperature of 23°C). Solar radiation is intense and photochemical reactivity therefore strong. The population and automobile densities are also high (an average of 10000 people per km^2 and 6600 car per km^2). Our earlier study has indicated that the acidic aerosol pollutants in metropolitan Taipei were significantly correlated with traffic densities (Mao et al., 2009). Fortunately, the industrial emissions are low. Obviously, geothermal emissions and automobile emissions contribute predominantly to air pollution in the city.

The three regions investigated in this study were sulfur-rich geothermal, suburban and downtown areas, as seen in Fig. 1. Air was sampled at schools in the selected areas. The acid aerosol

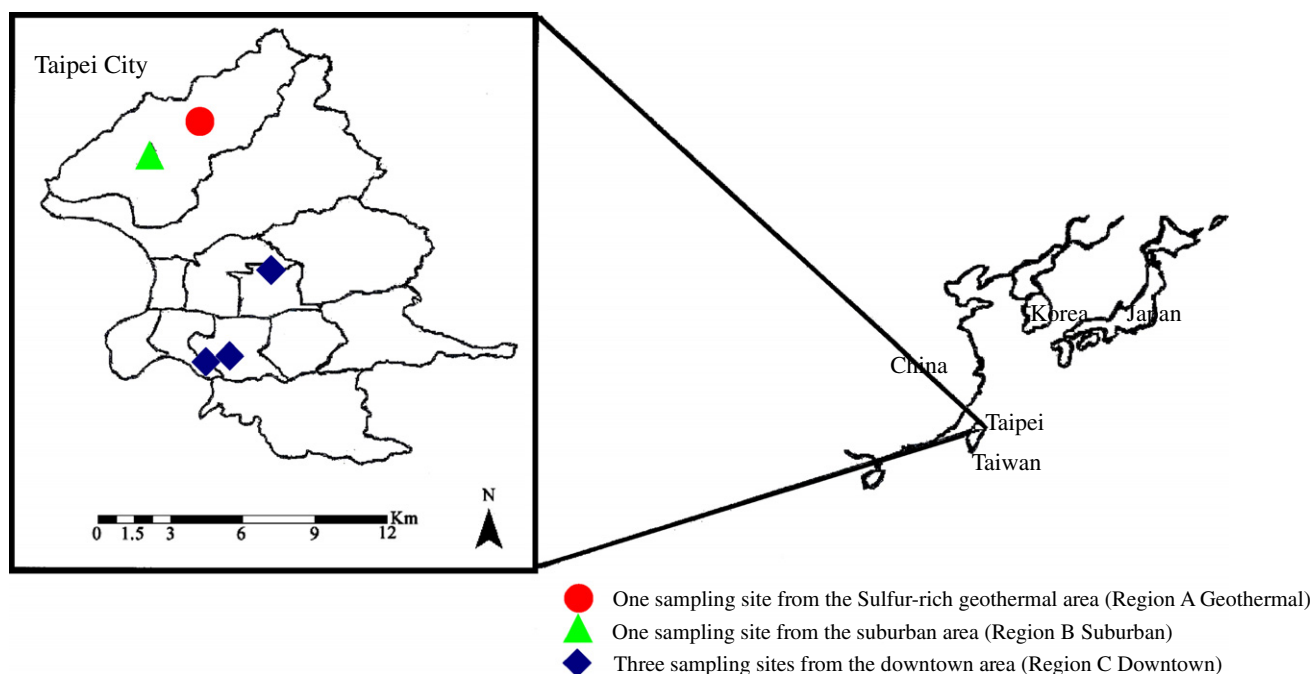


Fig. 1. Location of the sampling sites in different location of metropolitan Taipei.

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