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High-resolution ammonia emissions inventories in Fujian, China, 2009–2015

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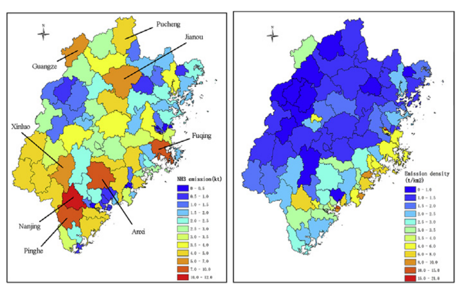
HIGHLIGHTS

- An ammonia emission inventory was developed based on county-level activity data.
- The emission of ammonia in Fujian province was 228.02 kt in 2015.
- Tempo-spatial distributions of ammonia emissions were investigated.
- Uncertainty and correlation analysis were used to evaluate the inventory.

GRAPHICAL ABSTRACT



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ABSTRACT

A high-resolution NH_3 emission inventory was developed based on the corrected emission factors and county-level activity data. To provide model-ready emission input, the NH_3 emission inventory was gridded for the modeling domain at 1 × 1 km resolution using source-based spatial surrogates and a GIS system. The best estimate of total NH_3 emission for the province was 228.02 kt in 2015 with a percentage uncertainty of ±16.3%. Four major contributors were farmland ecosystem, livestock wastes, humans and waste treatment, which contributed 39.4%, 43.1%, 4.9%, and 4.2% of the total emissions, respectively. The averaged NH_3 emission density for the whole region was 1.88 t km^{-2} yr^{-1} and the higher values were found in coastal areas with higher dense populations. The seasonal patterns, with higher emissions in summer, were consistent with the patterns of temperature and planting practices. From 2009 to 2015, annual NH_3 emissions increased from 218.49 kt to 228.02 kt. All of these changes are insignificant compared to the estimated overall uncertainties in the analysis, but indicative of changes in the source categories over this period. Between 2009 and 2015, the largest changes occurred in human emissions and waste treatment plants, which were consistent with the process of rapid urbanization. Meanwhile, the decrease of emissions from pigs was slightly higher than the increased emissions from broilers and the increased emissions from meat goats and beef cattle due to the combine effects of increasingly stringent environmental requirements for pig farms and shift away from pork consumption to beef, chicken and mutton. The validity of the estimates was further evaluated using uncertainty analysis, comparison with previous studies, and correlation analysis between emission density and observed

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ground ammonia. The inventories reflect the changes in economic progress and environmental protection and can provide scientific basis for the establishment of effective $PM_{2.5}$ control strategies.

1. Introduction

Ammonia (NH_3) is an important atmospheric pollutant with a wide variety of impacts. In the atmosphere, the alkaline NH_3 neutralizes a large portion of the acids produced by oxides of sulfur and nitrogen to form secondary sulfate and nitrate aerosols that very effectively penetrate lung tissues and degrade visibilities (Seinfeld and Pandis, 2006; Wang et al., 2011; Ye et al., 2011; Behera and Sharma, 2012; Wu et al., 2015). Our previous study indicated that the sum of sulfate, nitrate and ammonium (SNA) on average contributed 39.8–46.1% of the $PM_{2.5}$ mass and significantly drove the formation of $PM_{2.5}$ during the air pollution episodes at four urban sites in Fujian province, China, demonstrating the importance of ammonium in driving particulate SNA (Wu et al., 2015). Similar trends have also been observed at the urban sites of Beijing, Shanghai, Guangzhou and Suzhou in China (Huang et al., 2014; Tian et al., 2015). In order to meet the $PM_{2.5}$ levels set in the National Ambient Air Quality Standards (GB3095-2012) in China (http://kjs.mep.gov.cn), the combined desulfurization and denitrification units have been applied to flue gas cleanup to reduce the emission of SO_2 and NO_x. Nevertheless, a chemical transport model indicated that the benefit of SO_2 reduction from 2006 to 2015 would be completely offset over all of mainland China due to the significant increase of ammonia nitrate if NH_3 emissions are allowed to increase by +16% (the same percentage increase as NO_x) (Wang et al., 2013). In addition to the adverse effects of atmospheric ammonia on air quality, deposition returns most of the gaseous ammonia and particulate ammonium to the soil or water bodies, where it may contribute to the acidification and eutrophication of aquatic ecosystem (Van Breemen et al., 1982; Paerl et al., 2002; Krupa, 2003; Luo et al., 2014).

It is well established that the major two sources of NH_3 emissions to the atmosphere are the volatilization from the decomposition of livestock and poultry wastes, and from agricultural ecosystems following the application of N fertilizers. High resolution ammonia emission inventories in China indicated that these two major sources contributed 80–90% of total emissions from 1980 to 2012 (Kang et al., 2016). The two sources contributed about 85% of the total emissions in the United States in 1998 (U.S. Environmental Protection Agency, 2002). Although the emissions from livestock and poultry operations and fertilizer applications are large in the magnitude, they are mostly distributed over rural areas and removed from sources of NO_x and SO_x. Conversely, ammonia emitted from urban citizen emissions, mobile sources, waste treatment plants, and fossil fuel processing and combustion in urban area can influence $PM_{2.5}$ formation more significantly because NH_3 can be readily neutralized to form secondary nitrate or sulfate aerosols due to the coexistence of NH_3 and either NO_x or SO_x (Behera and Sharma, 2010; Huang X et al., 2011; Zbieranowski and Aherne, 2012). Ambient levels of NH_3 concentrations in densely populated urban areas are comparable or even higher than those in rural areas (Meng et al., 2011; Zbieranowski and Aherne, 2012; Gong et al., 2011; Ye et al., 2011; Reche et al., 2012). In addition, the seasonal variability of non-agricultural activities in urban areas is not as distinct as that of agricultural activities. A recent research suggested that NH_3 emissions, instead of SO_2 and NO_x emissions, were the key limiting factor for secondary inorganic $PM_{2.5}$ formation in urban regions in China (Wu et al., 2016). Because NH_3 plays such a key role in the formation of secondary $PM_{2.5}$ and atmospheric N deposition, accurate inventories of NH_3 are crucial to the development of realistic air quality modeling and effective regional haze and $PM_{2.5}$ abatement strategies.

Fujian province lies on the southeastern coastal region of China and houses a population of approximately 38 million people and spans an area of about 1.24×10^5 km^2. It faces the island province of the Taiwan across Taiwan Strait to the east and sits between the two economic strongholds, the Pearl River Delta (PRD) to the south and the Yangtze River Delta (YRD) to the north (Fig. 1). Fujian comprises of nine prefectural-level cities and 85 counties or districts at county level (including Kinmen which is currently controlled by the Republic of China on Taiwan). The most popular cities in Fujian include Fuzhou, Xiamen, Quanzhou, Zhangzhou and Putian, which are considered as the main engines of economic growth. There are

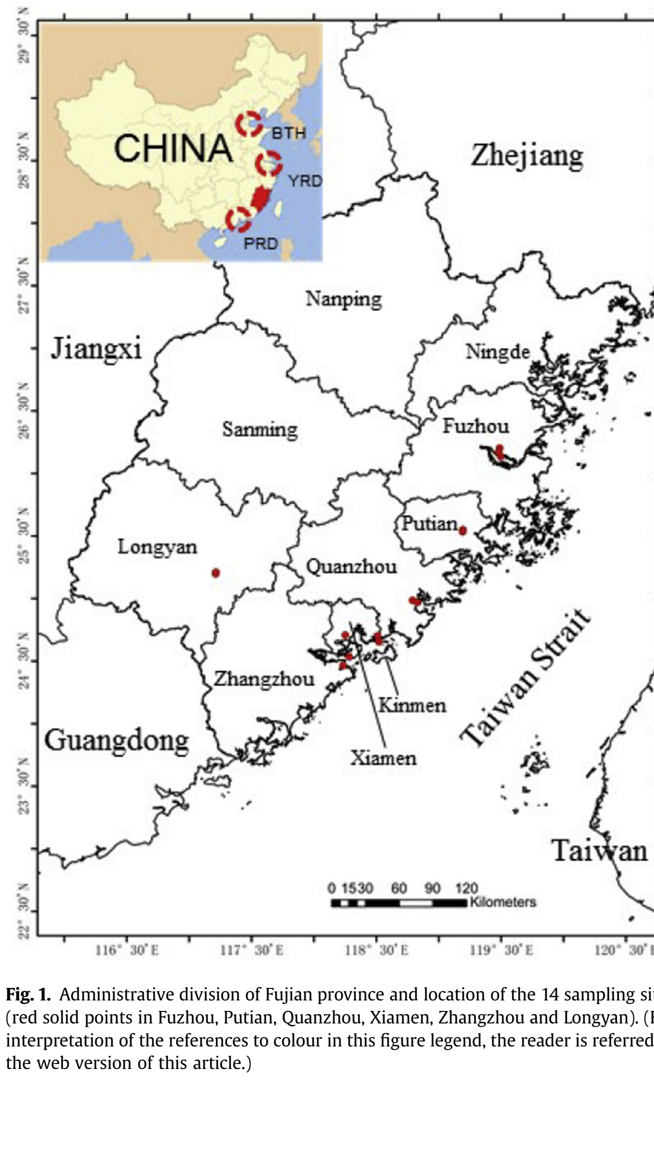

Fig. 1. Administrative division of Fujian province and location of the 14 sampling sites (red solid points in Fuzhou, Putian, Quanzhou, Xiamen, Zhangzhou and Longyan). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)