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Historical variations of biogenic volatile organic compound emission inventories in China, 1981–2003

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HIGHLIGHTS

- China's BVOC emissions had increased by 28.01% from 37.89 Tg in 1981 to 48.50 Tg in 2003.
- The increased vegetation biomass caused BVOC emissions increasing by 19.25%.
- The regions with high emissions had been expanding over the years.

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ABSTRACT

To evaluate the variations in temporal and spatial distribution of biogenic volatile organic compound (BVOC) emissions in China, historical BVOC emission inventories at a spatial resolution of 36 km × 36 km for the period of 1981–2003 were developed firstly. Based on the time-varying statistical data and Vegetation Atlas of China (1:1,000,000), emissions of isoprene, 37 monoterpenes, 32 sesquiterpenes, and other volatile organic compounds (OVOCs) were estimated using MEGANv2.1 driven by WRF model. Results show China's BVOC emissions had increased by 28.01% at an annual average rate of 1.27% from 37.89 Tg in 1981 to 48.50 Tg in 2003. Emissions of isoprene, monoterpenes, sesquiterpenes, and OVOCs had increased by 41.60%, 34.78%, 41.05%, and 4.89%, respectively. With fixed meteorological variables, the estimated BVOC emissions would increase by 19.25%, resulting from the increasing of vegetation biomass during the last 23 years. On average, isoprene, monoterpenes, sesquiterpenes, and OVOCs were responsible for 52.40%, 12.73%, 2.58%, and 32.29% of the national BVOC emissions, respectively. β-pinene and α-pinene, farnesene and caryophyllene were the largest contributors to the total monoterpene and sesquiterpene emissions, respectively. The highest emissions were found over northeastern, southeastern, southwestern China, Qinling Mountain, and Hainan and Taiwan provinces. The regions with high emissions had been expanding over the years, especially in the Changbai Mountain, southern China, and southwestern forest regions. The lowest emissions in southern China occurred in 1984–1988. Almost all the provinces had experienced increasing emissions, but their contributions to the national emissions differed significantly over the past 23 years. Yunnan, Guangxi, Heilongjiang, Jiangxi, Fujian, Guangdong, and Sichuan provinces always dominated the national BVOC emissions, excluding in 1977–1981, when the three northeastern provinces had relatively lower emissions.

1. Introduction

BVOCs may have substantial impacts on global environmental chemistry and climate, with isoprene, monoterpenes, acetone, etc., being the important precursors of tropospheric O_3, secondary organic aerosol (SOA) and peroxyacyl nitrates (PAN) (Guenther et al., 1999; Arneth et al., 2011; Carslaw et al., 2010; Sartelet et al., 2012; Nozière et al., 2011). It is essential to establish an accurate BVOC emission inventory for evaluating their roles in atmospheric environment. Global BVOC emissions have been estimated using improving biogenic model since the early 1990s (Guenther et al., 2006, and references therein). It was reported that China's BVOC emissions contributed 2.5% to the total global emissions (Guenther et al., 1995). In the past decades, the national BVOC emission inventories in China have been produced specially by many scholars using different methodologies, landcover and weather data

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(Klinger et al., 2002; Chi and Xie, 2011; Tie et al., 2006; Li et al., 2012, 2013; Fu and Liao, 2012). The regional BVOC emissions were also estimated in China, such as Beijing, Hong Kong, Shandong, and the Pearl River Delta region (Wang et al., 2003; Zheng et al., 2010; Tsui et al., 2009; Wang et al., 2011, 2005; Leung et al., 2010). However, those inventories all focused on China's BVOC emissions for a single or few years, and some paid attention to only one or few VOC species, none for monoterpenes and sesquiterpenes in detail.

The interannual variations of BVOC emissions induced by changes in vegetation cover and climate might be significant. Wu et al. (2012) found the year 2100 global biogenic emission of isoprene simulated with future vegetation will be higher by about 25% than that simulated with present-day vegetation, both using the present-day climate and considering the future impact of CO_2 on vegetation. CO_2-driven climate change would promote the emissions of BVOCs, which were predicted to increase by 22% or 55% from 2000 to 2100 on the basis of fixed distributions of vegetation (Liao et al., 2006; Heald et al., 2008). The increasing BVOC emissions would result in the enhancement of ozone and SOA concentration (Liao et al., 2006; Wu et al., 2012; Jiang et al., 2008). Understanding the interannual variations of BVOC emissions would help us make clear the relative role of biogenic and anthropogenic VOCs to the environmental pollution.

In order to control water and soil erosion effectively and protect the ecological environment, Chinese government has implemented several ecological restoration programs since the 1970s, including the Three-North Protective Forest Program (1978), Rivers Protection Forest Program (the end of the 1980s), the Natural Forest Conservation Program (2000), and the China's Sloping Land Conversion Program (2002). As a result of afforestation and reforestation, the forest area and timber volume in China have been increasing during the past decades, which might lead to the enhanced BVOC emissions. Fang et al. (2001) calculated the carbon storage from forests using an improved estimation method based on the relationship of forest biomass and timber volume, and found the carbon storage increased significantly after the late 1970s from 4.38 to 4.75 Pg of carbon by 1998, mainly due to forest expansion and regrowth. The terrestrial vegetation carbon sinks in China for period of 1981–2000 have increased, so was the vegetation biomass (Fang et al., 2007).

Thus, the aim of this study is to develop China's historical BVOC emission inventories at a spatial resolution of 36 km × 36 km for the period of 1981–2003, to evaluate the variations in temporal and spatial distribution of BVOC emissions in China. In the process of estimating China's BVOC emissions, the most detailed and accurate vegetation investigations were used, including the Vegetation Atlas of China and official statistical data in China. The China's forest inventory data used in this study are for the period of 1977–1981, 1984–1988, 1989–1993, 1994–1998, and 1999–2003. We used data from China Statistical Yearbooks and WRF simulations for the year 1981, 1988, 1993, 1998, and 2003 to estimate the BVOC emission in five periods, respectively. We evaluated the influence of the changes in land cover, mainly forest and crop, on emissions of BVOCs in China over the past 23 years.

2. Methodology

2.1. Emission estimation

The emissions of BVOCs in China were estimated using NEGANv2.1 (http://acd.ucar.edu/~guenther/MEGAN/MEGAN.htm). MEGAN has been proved to be an advanced global and regional biogenic emission model to estimate the net emission rates of BVOCs and other natural gases into the above-canopy atmosphere (Guenther et al., 2006; Sakulyanontvittaya et al., 2008; Guenther et al., 2012). In this study, biogenic emissions of 141 chemical species were simulated, including isoprene, 37 monoterpenes, 32 sesquiterpenes, and 71 OVOCs. A detailed multi-layer canopy environment model was used, which can explicitly determine the effects of leaf temperature and light on emissions in each level of the canopy. The emission rates of BVOCs were estimated by Eq. (1).

$$E = \varepsilon \times \gamma \times \rho \tag{1}$$

where E is the net emission rate (μg compound m^{-2} h^{-1}) of BVOC species; ε is the canopy-scale emission factor at standard conditions, μg m^{-2} h^{-1}; ρ is a factor that accounts for chemical production and loss of compounds in the vegetation canopy. The emission activity factor γ is determined by

$$\gamma = C_{CE} \times \gamma_{PT} \times LAI \times \gamma_{age} \times \gamma_{SM} \times \gamma_{C} \tag{2}$$

where C_{CE} is an adjustment factor so that $\gamma = 1$ under standard conditions, and is parameterized differently for biogenic species with light dependent and independent emissions (Guenther et al., 2006, 2012); γ_{PT} is the weighted average (for all leaves) of the product of the emission activity factors of leaf temperature and PPFD; LAI (m^{-2} m^{-2}) is the leaf area index; γ_{age}, γ_{SM}, and γ_C describes the effects of leaf age, soil moisture, and CO_2 inhibition. In this study, we set ρ to unity, as recommended for isoprene by Guenther et al. (2006) and terpenes by and Sakulyanontvittaya et al. (2008). This will have little impact on the modeling results as mentioned by Sakulyanontvittaya et al. (2008). Due to lacking data, the effect of both soil moisture and CO_2 concentration are not considered, which may introduce uncertainties to the spatio-temporal distribution of BVOC emissions in this study.

2.2. Landcover data

2.2.1. Plant distribution and land area

The landcover data inputted into the MEGAN model includes plant functional type (PFT) distribution and emission factors. In this study, a detailed vegetation classification with 82 plant functional types (including 28 dominant tree genera/species and 6 forest types, 13 primary crop species, 7 shrub subclasses, and 16 grassland types with 28 subclasses) was developed to accurately estimate the BVOC emissions in China (Li et al., 2013). The PFT distribution data derived from the Vegetation Atlas of China (1:1,000,000) (Fig. 1), presenting the most detailed and accurate vegetation distribution of China. The Atlas is based on nationwide vegetation surveys and associated research over the last 3–4 decades, and was published in 2007. Considering that the Atlas updates hardly and represents the average distribution of vegetation in China, the same PFT distribution was used in this study for the estimation of BVOC emissions during the period 1981–2003. MEGANv2.1 uses CLM PFT categories, which is different from the vegetation classification in this study. We computed the emissions of BVOCs from each PFT one by one, by putting each PFT in this study into the CLM PFT category which it belongs to, as listed in Table 1. The 82 PFTs data were allocated into every 36 km × 36 km grid by vegetation area using ArcGIS methodology to provide the MEGAN input.

MEGAN requires the land area index (LAI) of vegetation covered surfaces (LAIv) over a grid cell. MEGANv2.1 supports using the 8-day LAI database to provide a more detailed representation of seasonal LAI variations. However, the monthly LAI data tends to capture the major features (Guenther et al., 2012). In this study, the gridded LAIv distribution originated from the standard monthly LAIv database provided by MEGANv2.04. But this might still have flaws during periods of rapid LAI change. We did not consider the