Journal of Hazardous Materials 360 (2018) 329–339

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

An improved gridded polycyclic aromatic hydrocarbon emission inventory for the lower reaches of the Yangtze River Delta region from 2001 to 2015 using satellite data

Baojie Li[a,b], Shenglu Zhou[a,b,*], Teng Wang[a], Xueyan Sui[c], Zhenyi Jia[a,b], Yan Li[a,b], Junxiao Wang[a,b], Shaohua Wu[a,d,**]

[a] School of Geographic and Oceanographic Sciences, Nanjing University, Nanjing, 210023, China
[b] Key Laboratory of Coastal Zone Exploitation and Protection, Ministry of Land and Resources, Nanjing, 210017, China
[c] Jiangsu Land Consolidation and Rehabilitation Center, Nanjing, 210017, China
[d] Institute of Land and Urban-Rural Development, Zhejiang University of Finance & Economics, Hangzhou, 310018, China

ARTICLE INFO

Keywords:
Emission inventory
Polycyclic aromatic hydrocarbons
Satellite data
Spatial allocation
Yangtze River Delta region

ABSTRACT

An improved gridded polycyclic aromatic hydrocarbon (PAH) emission inventory for the lower reaches of the Yangtze River Delta (YRD) region from 2001 to 2015 was developed using satellite data. Despite rapid increases in energy consumption, the annual total emissions of the 16 PAHs showed overall decreasing trends, from a maximum of 5445 t in 2001 to a minimum of 2619 t in 2015, with the largest decline (84.6%) observed in the residential sector. Different spatial allocation methods used in gridded PAH emission inventories have substantial influences on the distributions of PAHs; therefore, we improved the accuracy of the spatial allocation of industrial and open biomass burning PAH emissions using various satellite data. The gridded secondary and tertiary industrial GDP (GDP_{23}) calculated using corrected nighttime light data was the best spatial proxy for the spatial allocation of industrial PAH emissions in the YRD region. We generated a gridded burned area for 2001–2015 by coupling the MCD64A1 and MCD14ML fire products, which was used to allocate PAH emissions from open biomass burning. Finally, we found that changes in the spatial distribution of PAH emissions were mainly driven by energy consumption and degree of technological advancement in different regions during 2001–2015.

1. Introduction

Airborne polycyclic aromatic hydrocarbons (PAHs) are typically generated by incomplete combustion or pyrolysis of fossil and biomass fuels and various industrial processes [1]. PAH exposure is associated with lung cancer and other diseases [2,3]. Efforts have been made to estimate PAH emissions in some regions (e.g., European Union, United States, China, and East Asia) [4–7], but the long-term trends in PAH emissions require additional research. Research of the long-term trends in PAH emissions have mainly focused on developed countries, which have shown declines in PAH emissions since 1990 because of technological improvements related to traffic sources [5,6,8]. By contrast, studies of the long-term trends in developing countries are scarce, although a few studies have found that PAH emissions in developing countries have shown a slowly declining trend since 1995 due to energy transitions in the rural residential sector [5].

In China, many initiatives (e.g., replacement of residential coal cooking stoves with natural gas stoves) in the rural residential sector have been implemented in recent years [9], which may have further decreased PAH emissions. In addition, owing to rapid urbanization under economic transition, the spatial distribution patterns of fuel consumption and energy structure have been changing rapidly in China [10], which may significantly affect the distribution of PAH emissions. Therefore, the spatiotemporal variations in PAH emissions warrant a closer examination in China.

Several organizations have established gridded PAH emission inventories (e.g., PKU-PAH, EDGARv4, EMEP, and REAS-POP) [4,5,11,12], but have used markedly different spatial allocation methods for industrial and other sources. Such differences in spatial proxies substantially influence PAH distributions in gridded emission

* Corresponding author at: School of Geographic and Oceanographic Sciences, Nanjing University, Nanjing, 210023, China.
** Corresponding author at: Institute of Land and Urban-Rural Development, Zhejiang University of Finance & Economics, Hangzhou, 310018, China.
E-mail addresses: baojieli@smail.nju.edu.cn (B. Li), zhousl@nju.edu.cn (S. Zhou), njuwangteng@163.com (T. Wang), xueyan322@sina.com (X. Sui), zhenyijay@smail.nju.edu.cn (Z. Jia), dg1627013@smail.nju.edu.cn (Y. Li), dz1427034@smail.nju.edu.cn (J. Wang), shaohuawu@126.com (S. Wu).

https://doi.org/10.1016/j.jhazmat.2018.08.011
Received 7 January 2018; Received in revised form 30 July 2018; Accepted 5 August 2018
Available online 10 August 2018
0304-3894/

inventories. For industrial sources, products of gridded population, land cover, and original nighttime light (NL) data have been used as spatial proxies in most PAH emission inventories [4,11,12]. However, these studies are subject to possible biases and limitations. For example, some studies have assumed that the spatial distributions of PAH emission sources do not vary temporally [4,13], or have used spatial proxy data from adjacent years to allocate PAH emissions [14], which may introduce uncertainty. Furthermore, using population data as a spatial allocation proxy involves high uncertainty due to differences in energy consumption per capita.

By contrast, gross domestic product (GDP) or secondary and tertiary industrial GDP (GDP_{23}) may represent good spatial proxies of industrial PAH emissions. Numerous studies have illustrated a strong relationship between GDP and energy consumption [7,15], especially in China, where a long-running equilibrium relationship between economic growth and energy consumption has been found [15,16]. However, few studies have used gridded GDP data as a spatial proxy, because publicly available gridded GDP data are often not continuous [17]. Recent studies have modeled gridded GDP using corrected NL data [18,19], solving the biases of the original NL data (e.g., pixel saturation in urban centers and a lack of continuity/comparability of data from the Defense Meteorological Satellite Program's Operational Linescan System (DMSP-OLS)) [20]. Suomi National Polar-Orbiting Partnership Visible Infrared Imaging Radiometer Suite (NPP-VIIRS) NL data were released in early 2013, greatly improving the accuracy and resolution of NL data [21]; regardless, biases exist in the NPP-VIIRS data, including abnormal light detection and background noise [22].

For open biomass burning, most studies have allocated total open biomass emissions to grid boxes based on population density, land-use type, or biomass area, leading to relatively high uncertainties [23,24]. Other studies have allocated open biomass burning according to satellite data [25,26]; however, using only one satellite fire product to allocate open biomass burning may be problematic. For example, NCAR Fire Inventory (FINN) studies have used the MODIS active fire product MCD14ML, which can capture small fires [27] but may underestimate emissions due to the temporal limitations of the satellite for capturing fire points (10:30 ∼ 13:30) [28]. Meanwhile, another product, MCD64A1, has a 500 × 500-m resolution, and can only identify large fires [29].

The lower reaches of the Yangtze River Delta (YRD), characterized by a high population density and well-developed industries [30], accounted for only 2.2% of China by area, but represented 20.1% of the GDP and 13.7% of the total energy consumption in 2015 [31]. Moreover, rapid industrialization and urbanization has led to a 7.3-fold increase in energy consumption over the past 30 years [31], resulting in severe air and soil PAH contamination [32,33]. The YRD region has among the highest PAH emission densities in China, directly threatening local environmental quality and human health [14,34]. Therefore, it is critical to identify the long-term trends and spatial distribution in PAH emissions in the lower reaches of the YRD.

The main objectives of this study are to (1) improve the accuracy of the spatial allocation of industrial PAH emissions using gridded GDP_{23} data calculated using corrected DMSP-OLS and NPP-VIIRS NL data, (2) improve the accuracy of the spatial allocation of open biomass burning PAH emissions by coupling two types of satellite data, and (3) establish a gridded PAH emission inventory for the lower reaches of the YRD from 2001 to 2015.

2. Data and methodology

2.1. Study area

The lower reaches of the YRD is among the most developed regions in China, with 25 cities and three provinces (Shanghai, Zhejiang, and Jiangsu) (Fig. 1). However, high energy consumption has resulted in severe pollution of various environmental compartments [33,35,36].

2.2. Gridded emission inventory framework

The 16 PAHs included in this study were naphthalene (NAP), acenaphthylene (ACY), acenaphthene (ACE), fluorine (FLO), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLA), pyrene (PYR), benz(a) anthracene (BaA), chrysene (CHR), benzo(b)fluoranthene (BbF), benzo (k)fluoranthene (BkF), benzo(a)pyrene (BaP), dibenz(a,h)anthracene (DahA), indeno(l,2,3-cd)pyrene (IcdP), and benzo(g,h,i)perylene (BghiP).

In total, 30 emission sources of the 16 PAHs were divided into five sectors (Table S1): power generation, industry, residential, transportation, and agriculture. Among these, the industrial sector included primarily industrial coal and oil combustion, petroleum refinery, primary aluminum production, and the iron–steel industry. The residential sector included primarily domestic coal, indoor straw burning, and firewood combustion. The agricultural sector included outdoor straw burning. PAH emissions (E_{PAH}) were calculated using Eq. (1):

$$E_{PAH} = \sum_{i,k,l} A_{i,k} \times X_{i,k,l} \times EF_{j,k,l} \tag{1}$$

where i, k, and l represent the sector, fuel or product, and technology, respectively; EF is the emission factor of each PAH species j and was derived from previous studies (Table S2) [5,7,37]; and X is the fraction of the activity rate contributed by a given technology, which was calculated using the technology split method, where 17 sources were considered. The time-dependent fractions of these technology divisions were calculated using a series of S-shaped curves (Table S3) [5,38]. Most activity data (A) were obtained directly from statistical yearbooks [31,39–42]. Moreover, the amount of combusted crop residue was estimated using Equation (S2).

To minimize uncertainty in the PAH emission spatial distributions, we first calculated PAH emissions from different sources at a provincial scale during 2001–2015. Then, we applied various methods to spatially allocate the provincial PAH emissions into grid cells (Fig. 2). Finally, we developed a PAH emission inventory with a 6 × 6-km spatial resolution for the lower reaches of the YRD from 2001 to 2015 by combining the gridded PAH emissions from different provinces.

2.3. Spatial allocation of industrial PAH emissions using nighttime light data

Gridded GDP_{23} values for 15 consecutive years were used as spatial surrogates for the industrial PAH emission sources. The gridded GDP_{23} data were modeled using corrected annual DMSP-OLS and NPP-VIIRS NL data. The annual DMSP-OLS data (2001–2013) included data acquired by four DMSP satellites: F14, F15, F16, and F18 (http://www.ngdc.noaa.gov/eog/dmsp/downloadV4composites.html). The monthly NPP-VIIRS data (2013–2015) were derived from https://www.ngdc.noaa.gov/eog/viirs/. Details of the DMSP-OLS and NPP-VIIRS data correction methods are described in Section S3. After correcting the DMSP-OLS (2001–2013) and NPP-VIIRS (2013–2015) data, we allocated the GDP_{23} data with Eq. (2) using an intercept of 0:

$$GDP_{23_{y,pixel}} = (GDP_{23_{y,sum}}/GDP_{y,sum}) \times DN_{y,pixel} \tag{2}$$

where $GDP_{23y,pixel}$ denotes the GDP_{23} in each pixel of each year (2001–2015); $GDP_{23y,sum}$ denotes the total GDP_{23} of each year in the lower reaches of the YRD; $TNL_{y,sum}$ is the sum the total NL of each year in the study area; and $DN_{y,pixel}$ denotes the sum of the digital number values in each pixel of each year.

We assessed the accuracy of five spatial allocation proxies, population data, original gridded DMSP-OLS and NPP-VIIRS NL data (DMSP and NPP-VIIRS), and estimated gridded GDP_{23} data based on the corrected DMSP-OLS/NPP-VIIRS NL data (GDP_{23}-DMSP and GDP_{23}-NPP-VIIRS). Then, we identified the best spatial proxies for industrial PAH emissions. The year 2013 was selected for study because data coverage was available for both DMSP-OLS and NPP-VIIRS NL data. We collected