Atmospheric Environment 128 (2016) 1–9

Contents lists available at ScienceDirect

Atmospheric Environment

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

Atmospheric emission inventory of hazardous air pollutants from China's cement plants: Temporal trends, spatial variation characteristics and scenario projections

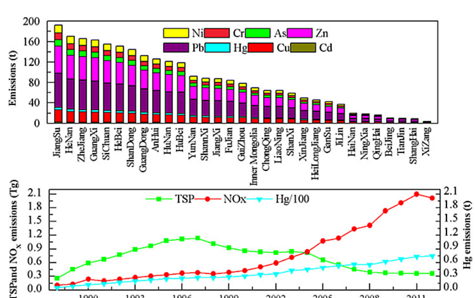
Shenbing Hua [a, b], Hezhong Tian [a, b, *], Kun Wang [a, b], Chuanyong Zhu [a, b], Jiajia Gao [a, c], Yilin Ma [b, d], Yifeng Xue [a, e], Yong Wang [a, b], Shiheng Duan [a, b], Junrui Zhou [a, b]

[a] *State Key Joint Laboratory of Environmental Simulation & Pollution Control, School of Environment, Beijing Normal University, Beijing 100875, China*
[b] *Center of Atmospheric Environmental Studies, Beijing Normal University, Beijing 100875, China*
[c] *Division of Air Pollution Control, Beijing Municipal Institute of Labour Protection, Beijing 100037, China*
[d] *Beijing No.4 School, Xicheng District, Beijing 100034, China*
[e] *National Engineering Research Center of Urban Environmental Pollution Control, Beijing Municipal Research Institute of Environmental Protection, Beijing 100037, China*

HIGHLIGHTS

- We develop a comprehensive emission inventory of HAPs from China's cement industry.
- Dynamic EFs of shaft, new suspension pre-heater and other rotary kilns are proposed.
- We use transformed normal distribution function to reflect historical EFs variation.
- Future trends of HAPs emission till 2050 are predicted with three scenario analysis.
- HAPs emissions from 2961 plants in 2012 are allocated into 15 km × 15 km gridded cells.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 30 September 2015
Received in revised form
21 December 2015
Accepted 23 December 2015
Available online 24 December 2015

Keywords:
Hazardous air pollutants
Emission inventory
Cement production
Temporal trend

ABSTRACTS

A multiple-year comprehensive emission inventory of typical hazardous air pollutants (HAPs) from China's cement industry for the period 1980–2012, has been established by using technology-based dynamic emission factors and detailed annual plant-specific cement production from different types of kilns. Our results show that the total emissions of various HAPs (SO_2, NO_X, CO, PM, Hg, Cd, Cr, Pb, Zn, As, Ni and Cu) have rapidly increased by about 1–21 times at an annual average growth rate of 1–10% over the past three decades. Remarkably uneven spatial allocation features of these pollutants among provinces are observed. HAPs emissions are primarily concentrated in the eastern and coastal provinces due to the concentration of cement plants and their huge volume of coal consumption. We predict the future emission trends of HAPs through 2050 based on industry construction and policy guidance, and our scenario analysis indicates that HAPs emissions will drop substantially because of the combined effects of cement production yields reduction and the increasing application rate of various air pollution

* Corresponding author. Center of Atmospheric Environmental Studies, Beijing Normal University, NO. 19, Xinjiekouwai Street, Haidian District, Beijing 100875, China.
E-mail address: hztian@bnu.edu.cn (H. Tian).

http://dx.doi.org/10.1016/j.atmosenv.2015.12.056
1352-2310/

Spatial distribution
Scenario analysis

control devices. The study suggests that highlighted attention should be paid to this energy-intensive industry to ensure stricter emission standards are implemented for these HAPs emissions in the future.

1. Introduction

China is the largest cement producer and consumer in the world. Total cement production of China is reported to be 22.1 billion tons in 2012, accounting for about 63% of global cement production (CCA, 2013; USGS, 2013). Cement production is highly energy-intensive, and the total energy consumption of this industry has amounted to 155Mtce in 2009 (NBS, 1981–2013). Meanwhile, the enormous quantities of various hazardous air pollutants (HAPs) are emitted into the atmosphere, including the conventional air pollutants such as sulfur dioxide (SO_2), nitrogen oxides (NO_X), carbon monoxide (CO), and particulate matter (PM), as well as various toxic heavy metals (HMs) like mercury (Hg), cadmium (Cd), chromium (Cr), lead (Pb), zinc (Zn), arsenic (As), nickel (Ni) and copper (Cu), which aggravating local and regional environmental problems as well as contributing to human health damage and climate change. Cement production is claimed as the largest source of PM emissions, accounting for 20%–30% of national total emissions and 40% of total industrial emissions in China (CEYEC, 2013). Additionally, various toxic HMs associated with cement production process have received increasing attention. For example, mercury emissions from Chinese cement plants are calculated to be about 23 t in 1999, obviously higher than Hg emissions emitted from cement plants of other countries such as America, Germany and Japan (Streets et al., 2005; Wu et al., 2006).

During the past decades, the Chinese government has issued a series of emission standards and control polices, whereas an official emission inventory of the cement industry is still not available. There are some general emission inventories which simply treating the cement industry as an overall source category based on relevant statistical data in terms of gaseous pollutants and PM but they ignore the effectiveness of technological advances and emission control measures to some extent (Streets et al., 2005; Ma, 2010; Wang et al., 2008; Xue et al., 2014). With regard to the toxic HMs, few studies at national and provincial level have been performed except for the element of Hg (Streets et al., 2005; Mlakar et al., 2010; Wang et al., 2014; Zheng et al., 2012).

The purpose of this article is to summarize the best currently available information on activity level and determine the yearly-varied dynamic emission factors, and then evaluate the historical trend of HAPs emissions from 1980 to 2012, establish a new integrated gridded emission inventory of typical HAPs for the year of 2012 with high spatial resolution, and predict the future trends of emissions from 2015 to 2050 by using scenarios analysis.

2. Methodology and data sources

2.1. The framework of emission inventory

The basic methods of developing an emission inventory have been discussed in previous studies (Tian et al., 2014, 2015). In this study, the comprehensive emission inventory of HAPs from cement production is made of three categories of hazardous air pollutants: gaseous pollutants (SO_2, NO_X, and CO), PM (separated into $PM_{2.5}$, PM_{10} and TSP with respect particle diameter) and typical toxic HMs including Hg, Cd, Cr, Pb, Zn, As, Ni and Cu.

HAPs emissions from the cement industry of each province in China are estimated based on the cement outputs statistics by province and the specific emission factors (EFs) of each kiln type as well as the removal effects of varied control technologies, and then aggregate to the Chinese national level. The calculation approaches used to estimate emissions of various HAPs are summarized in Table 1.

2.2. Activity level and data source

In this work, we follow the previous approaches from many sources to derive activity data (Tian et al., 2012a). Total cement production of each province since 1980 is available from China Statistical Yearbook (CEYRC, 2013). There are mainly two kiln types in China: shaft kilns and rotary kilns, and rotary kilns can be further classified into recently developed suspension pre-heaters (NSP) kilns and other types of rotary kilns (OR). The penetration rate of the three types of cement kilns (see in Figure S1) is calculated based on the historic capacity of precalciner kilns and OR kilns (Xu et al., 2012; Kong, 2005). Here, we estimate coal consumption for cement industry based on typical clinker to cement ratios (CCA, 2001–2013) and energy intensity (consumption of coal per ton of clinker) (Gu et al., 2012) of China's cement industry (see in Table S1 of the separate Supporting Information (SI) file).

2.3. Emission factors

2.3.1. Emission factors of SO_2, NO_X and CO

SO_2 mainly comes from the oxidation of sulfur in fuels such as coal. In NSP kilns, approximately 70% of SO_2 is absorbed by reaction with calcium oxide or calcium carbide, while much more SO_2 is discharged in shaft kilns and OR kilns (Liu, 2006; Su and Gao, 1998) and less SO_2 is emitted into atmosphere from NSP. Liu (1998) summarize the various sulfur in China based on coal reserves, output, coal seam sulfur and other parameters in the 241 coal mines. We assume that 80% of SO_2 is absorbed for NSP kiln process and about 30% for other kilns, respectively. Thus, SO_2 emissions factors from 1980 to 2012 can be calculated.

CO and NO_X are highly influenced by sintering temperature and oxidation conditions. And the burning of fuel in the cement production process is normally treated as the sole source of CO and NO_X emissions. Su and Gao (1998) reported NO_X and CO emission factors for cement kilns of about 0.3 and 20–30 g/kg-cement for

Table 1
Calculation equations of various HAPs from cement production of China.

Pollutants	Equation for emissions estimation
SO_2, NO_X, CO	$E_j = \sum_{i,k} CC_{i,j,k} \times EF_k$
PM	$E_{j,m} = \sum_{i,k} P_{i,j,k} \times EFR_k \times \sum_n r_{k,n,m} \times (1 - \eta_{n,m})$
Heavy Metals (HMs)	$E_j = \sum_{i,k} CC_{i,j,k} \times EF_k + \sum_i CP_{i,j} \times EFH$

E: total emissions of HAPs, *tons/yr.*; *CC*: coal consumption, *tons/yr.*; *CP*: clinker production, *tons/yr.*; *P*: cement production, *tons/yr.*; *EF*: emission factors from coal combustion, *g/kg*; *EFR*: uncontrolled emission factor from coal combustion, *g/kg*; *EFH*: final emission factor of HMs, *g/t*; *i*: each province; *j*: year; *k*: type of kiln, emission source and PM control technology; *m*: different sizes of PM; η: removal efficiency of PM control technology; $\sum rn = 1$ (PM control technology).