

Inventory of PCNs emission to air from secondary nonferrous metallurgy industries in China

Bate and Minghui Zheng

State Key Laboratory of Environmental Chemistry and Ecotoxicology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, P.O. Box 2871, Beijing 100085, China

Introduction

Polychlorinated naphthalenes (PCNs) are a group of compounds that consist of 75 congeners, differing in the number of chlorine atoms and position of substitutions. Before 1980s, technical mixtures of PCNs were used in a wide variety of industries. The use and production of PCNs were banned because of their toxicity, environmental persistence and potential ecological risk in the 1980s¹. However PCNs was lately found in flue gas and fly ash of municipal waste incinerators as unintentional product of incomplete combustion (PICs). The concerns of formation of PCNs in incineration process similar to formation of polychlorinated dibenzo-p-dioxin and dibenzofurans (PCDD/Fs) make it is essential to investigate the potential sources of PCNs^{2,3}. The secondary nonferrous metallurgy industries are listed in main sources of PCDD/Fs catalogues. However the formation of PCNs in secondary nonferrous metallurgy industries has not been investigated. This study focus on the formation of PCNs from 4 kinds of nonferrous recycle production: aluminum, copper, zinc and lead production. The emission factors and total emissions of PCNs emitted to air from these 4 secondary nonferrous metallurgy industries in China were estimated.

Material and methods

In this study, a total of 15 stack gas and 10 fly ash samples were collected from 4 secondary nonferrous recycle industries in China. Stack gas samples were collected using automatic isokinetic sampling system. The samples were spiked with a mixture of ¹³C₁₂-labelled PCNs internal standards before extraction. The concentrated extracts were cleaned up by sulfuric acid, multi-layer silica gel and alumina. The concentrated eluents were spiked with a mixture of ¹³C₁₂-labelled PCNs recovery standards before analysis. PCNs identification and quantification were accomplished with a high-resolution gas chromatograph-high resolution mass spectrometer (HRGC-HRMS) system (DFS, Thermo Electron Corporation) on a resolution of approximate 10000. A DB-5 (J&W, 60 m × 0.25 mm i.d., 0.25 μm film thickness) capillary column was used for the determination of PCN congeners.

Results and discussion

Levels of PCNs from stack gas and ash in secondary nonferrous metallurgy industries

The concentration of PCNs in stack gas and fly ash from secondary nonferrous smelting process are shown in Fig.1 and Fig.2. The average concentration of stack gas released from secondary aluminum, copper, lead, zinc metallurgy processes are 54.9, 112.6, 13.6 and 341.6 pg TEQ/Nm³ respectively. The mean concentration of fly ash samples released from secondary aluminum, copper, lead, zinc production are 852.5, 1607.2, 0.29, and 54.9 pg TEQ/g respectively. The PCNs concentration of secondary zinc production is higher than the other metals production.

PCNs congener profiles in secondary nonferrous metallurgy industries

The congener profiles of PCNs in stack gas samples from secondary nonferrous metallurgy process are shown in Fig.3. The most abundant congener is 1,2,3,4,5,6,7-HpCN (CN 73) in aluminum and copper metallurgy process, 1,2,3,4,6,7-/1,2,3,5,6,7-HxCN(CN 66/67) in zinc production, and 2-MoCN (CN 2) in lead production. The abundance sequences of these 4 metallurgy processes are quiet different. The sum of

abundance of CN-2, 66/67, 73 were from 65.9% to 78.7% in these 4 metallurgy processes. It shows CN-1, 2, 66/67, 69 and 73 are predominant PCNs congeners from secondary nonferrous metallurgy industries.

Estimation of PCNs emissions from secondary nonferrous metallurgy industries in China

The emission factors of PCNs released to air was derived from a specific flue gas flow rate (Nm^3/t metal production) and a concentration of PCNs ($\text{ng WHO-TEQ}/\text{Nm}^3$, 11 % O_2). The total emission was developed by multiplying the emission factors and corresponding activity levels of 2007 of those secondary nonferrous metals metallurgy industries in China⁴. The total emission amount of secondary aluminum, copper, lead and zinc metallurgy industries in China are 2.8, 1.8, 0.031 and 0.028 g I-TEQ/yr respectively.

Acknowledgments

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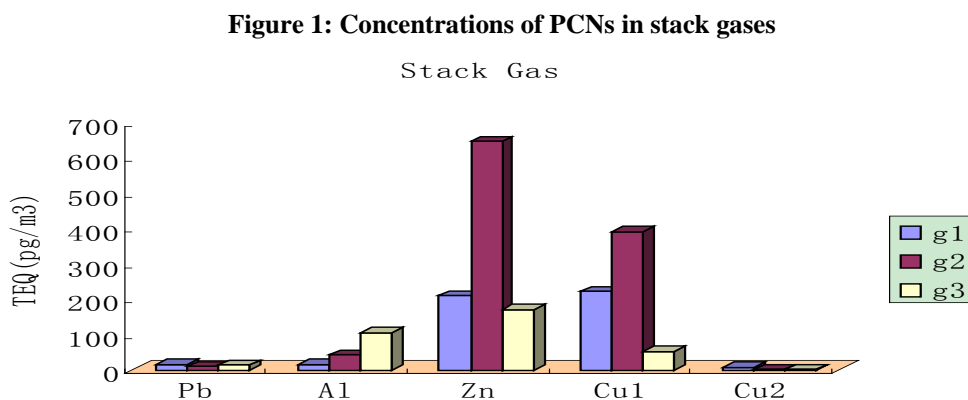


Figure 2: Concentrations of PCNs in fly ashes

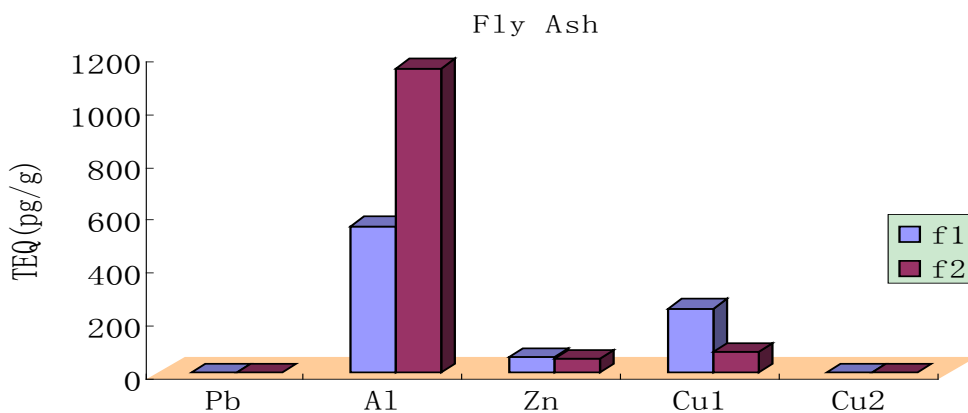


Figure 3: The distribution PCN congeners in stack gases

