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Fugitive halocarbon emissions from working face of municipal solid waste landfills in China

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Halocarbons are important anthropogenic greenhouse gases (GHGs) due to their long lifetime and large characteristic factors. The present study for the first time assessed the global warming potential (GWP) of fugitive halocarbon emissions from the working face of landfills in China. The national emissions of five major halocarbons (CFC-11, CFC-113, CH_2Cl_2, $CHCl_3$ and CCl_4) from the working face of municipal solid waste landfills in China were provided through observation-based estimations. The fluxes of halocarbons from working face of landfills were observed much higher than covered cells in landfills hence representing the hot spots of landfill emissions. The annual emissions of the halocarbons from landfills in China were 0.02–15.6 kt·y^{-1}, and their GWPs were 128–60,948 kt-CO_2-eq·y^{-1} based on their characteristic factors on a 100-year horizon. CFC-113 was the dominant species owing to its highest releasing rate (i.e. 15.4 ± 19.1 g·t^{-1}) and largest characteristic factor, resulting in a GWP up to 4036 ± 4855 kt-CO_2-eq·y^{-1}. The annual emissions of CFC-113 from landfills (i.e. 0.61 kt·y^{-1}) made up ~76% of the total national CFC-113 emissions. The GWPs of halocarbons were estimated ~14.4% of landfill methane emissions. Therefore, fugitive halocarbons emissions from working face are significant sources of GHGs in landfill sites in China, although they comprise a small fraction of total landfill gases.

1. Introduction

In 2005, China surpassed the United States and became the largest emitter of anthropogenic Greenhouse Gases (GHGs) globally (Leggett, 2011). According to the 21st Conference of the Parties (COP21) of United Nations Framework Convention on Climate Change (UNFCCC), China proposed to reach carbon emissions peak by 2030 (National Development and Reform Commission, 2014). To tackle the issue, China has pledged to reduce its carbon intensity, namely carbon dioxide (CO_2) emission per unit of GDP (Gross Domestic Product) by 40–45% by 2020 compared to 2005 (National Development and Reform Commission, 2014). Nowadays, waste sector (including disposal and treatment process) is considered as one of the important anthropogenic sources of GHGs, which has attracted worldwide attention (Marchi et al., 2017; Tian et al., 2013). According to the 5th Assessment Report of the Intergovernmental Panel on Climate Change (AR5-IPCC), GHG direct emissions from waste management sector have accounted for 2.9% of the total anthropogenic emissions. Urban waste management has been recognized as a significant part for cutting GHGs emissions by National Climate Change Program of China (National Development and Reform Commission, 2007). Despite methane (CH_4) emission from landfill gas (LFG) being regarded as the major GHG contributor (Myhre et al., 2013; Scheutz et al., 2009), halocarbons are a significant source of GHGs, such as chlorofluorocarbons (CFCs) and chlorinated solvents (Allen et al., 1997; Duan et al., 2014; Fang et al., 2012; Haderlein and Pecher, 1988; Scheutz et al., 2010), when their long lifetime and high Global Warming Potential (GWP, GWP = emissions × characteristic factor) are taken into account.

Halocarbons, categorized as major pollutants under the Montreal Protocol, can persist for several decades in wastes and present as trace components in LFG (Schuetz et al., 2003). Halocarbons are almost all man-made chemical substances, which have been widely used as refrigerants, blowing agents for foaming process, fire extinguishers, and solvent cleaners, they could be released from these applications and disposal of their end-of-life

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Nomenclature

CCl_4	tetrachloromethane
CFCs	chlorofluorocarbons
CFC-113	trichlorotrifluoroethane
CFC-11	trichlorofluoromethane
CFC-12	dichlorodifluoromethane
CH_2Cl_2	dichloromethane
$CHCl_3$	chloroform
CH_4	methane
CO_2	carbon dioxide
ER	emission rate
GDP	gross domestic product
GHGs	Greenhouse Gases
GWP	Global Warming Potential
LFG	landfill gas
IPCC	Intergovernmental Panel on Climate Change
MSW	municipal solid waste
SER	specific emission rate

waste. Amongst halocarbons, trichlorofluoromethane (CFC-11), trichlorotrifluoroethane (CFC-113), dichloromethane (CH_2Cl_2), chloroform ($CHCl_3$) and tetrachloromethane (CCl_4) were chosen as the target substrates. This is not only because of their notable amount in LFG rather than acceptable ambient concentrations, but also due to their impacts on climate change (An et al., 2012; Barletta et al., 2006). CFCs could be enumerated as influential GHGs, which cause a positive radiative forcing of climate directly (Myhre et al., 2013). They have been well known as ozone-depleting substances which indirectly contribute to climate change (Duffy et al., 2013). Long-lived substances, such as CFC-11 (54 years), CFC-113 (109 years) and CCl_4 (35 years) have been clarified as the controlled halocarbon substances by the Montreal Protocol, while CH_2Cl_2 and $CHCl_3$ are identified as the compounds resulting in global warming by AR5-IPCC with defined characteristic factors (Myhre et al., 2013). CH_2Cl_2 was mainly consumed in plastic film production the pharmaceutical industry, cleaning solvent and chemical production (Zhang et al., 2010b); $CHCl_3$ was used as solvent in China (Wang et al., 2014); CCl_4 was largely used as a feed stock for manufacture of CFC-11 (Wang et al., 2014).

Halocarbon emissions from China have been of extensive interest for both multi-scale environmental studies and policy making. This matter is getting more attentions because of high production and consumption of halocarbons in China along with delay in their control in compared to the developed countries. That is why China is anticipated to become a big contributor to the global halocarbon emissions. In the perspective of waste management, halogen-containing substances have already entered the waste stream in China owing to the poor source separation system of municipal solid wastes (MSW). This has been foreseen to be a significant contributor to both entire global and East Asian emissions (Chen et al., 2010). The emissions of CFCs in metric tons from East Asia in 2008 were estimated to represent 15% of the global totals. China has been evaluated to engender at least 80% of the East Asian emissions of halocarbons (Li et al., 2011). There have been a few investigations on specification and measurements of ambient halocarbon concentrations over resent decade throughout China (Eklund et al., 1998; Fang et al., 2012; Jooil et al., 2010; Qin, 2007; Yokouchi et al., 2006), and a systematic research on the historical and future emissions of the halocarbons in China for 1995–2024 has been assessed (Wan et al., 2009). None of them has directly discovered the case study of such compound emissions measurement from MSW landfill, especially on the working face of landfill.

Working face of landfills is a significant source of fugitive volatile compounds emissions (Duan et al., 2014; Liu et al., 2016; Scheutz et al., 2010). In landfill sites studied, fresh wastes were piled up and compacted on working face meanwhile they were exposed to the environment for days even weeks before being well covered. Halogen-containing wastes on working face could be the major sources of fugitive halocarbon emissions. According to IPCC, no inventory method is available for quantifying halogenated gases from other waste sectors so far (Victor et al., 2014), in-situ measurements are still required for the quantification of halocarbon emissions, and such kind of data make great contribution for multi-scale environmental studies later on.

This is the first investigation in China, to our best knowledge, estimated halocarbon emissions from the working face of landfills in China and assessed their impacts on global warming. To determine GWPs by fugitive halocarbon emissions from landfills in China, field measurement of halocarbon emission rates (*ER*s) were conducted from the working face of several typical landfills. The amount of national emissions was estimated on the basis of average *ER* and national MSW productions. Eventually, their GWPs were compared with that of landfill CH_4 emissions. These results have provided new knowledge of fugitive emissions of halocarbons from working face of landfills and their contribution to global warming, which could be compensating the inventory by IPCC and offer methodology for halocarbons measurement from working face of landfills in developing countries.

2. Materials and methods

In the present study, an effective wind tunnel sampling system, reported by the authors (Liu et al., 2016) was adopted to quantify *ER*s of the halocarbons. As the prerequisite, field samplings were undertaken on the working face of three typical anaerobic MSW landfills in China. The *ER*s measured by per unit area and per unit time was converted to the flux by per mass MSW. Annual halocarbon emissions from working face of landfills in China were evaluated on the basis of MSW landfilled, and their global warming contributions were assessed by multiplying annual emissions with characteristic factor values published by the IPCC AR5 in 2013 (Myhre et al., 2013).

2.1. MSW composition and sites description

About 60% of MSW in China is food waste resulting in its high organic and moisture contents (Zhang et al., 2010a). The typical composition of MSW in China reported by Zhang et al. (2010a) is shown in Fig. A1 (Supplementary material). According to Fig. A1 and a previous research, however in China organic garbage accounts for approximately 65% of the total MSW, the large amount of halocarbons released from MSW in China is even more than developed countries (Scheutz et al., 2010). Since no industrial wastes have been disposed in the landfill sites that were studied, the halocarbons emissions could be attributed to the category of plastic and textile and any halogenated-products in waste stream of MSW shown in Fig. A1.

Considering the variation of climate, living habits and economic development between cities, three typical sites located in the north (L1-Beijing), central (L2-Shaanxi, Xi'an) and south (L3-Guangdong,

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