Submitted to New Directions-AE, May 6, 2010, revised August 31 and September 7, 2010

New Directions: Need for better understanding of plastic waste burning as inferred from high abundance of terephthalic acid in South Asian aerosols

Terephthalic acid (1,4-benzenedicarboxylic acid; *t*-Ph) is an important industrial material used for making plastics such as polyester fibre and PET (polyethyleneterephthalate) thermoplastics. In term of public health, *t*-Ph promotes a bladder cancer (Heck and Tyl, 1985). Global consumption of plastics reached 150 millions of tons per year in 2000, with annual growth rate of 5.5% (Braun, 2004). Global production of polyester fibre alone was 24.5 million tons (65% of the world chemical fibre market) in 2004, 61% of which come from Asia, followed by North America and Western Europe (Aizenshtein, 2006). Although plastic materials are increasingly recycled in developed countries, they are dumped as the municipal solid wastes (MSW) in developing countries. The MSW are often burned under open-fire conditions to avoid the spread of mosquitoes that transmit malaria near the dumpsite, emitting plastic decomposition products to the atmosphere.

t-Ph has been reported at elevated levels in smoke particles derived from open burning of new plastic bags (907 ng mg^{-1}), roadside litter (5033 ng mg^{-1}) and landfill trash (176 ng mg^{-1}) (Simoneit et al., 2005). In addition, secondary formation by atmospheric oxidation reactions may also be significant. However, *t*-Ph has rarely been reported in ambient aerosols that are not derived from open-burning activities such as those listed above (Table 1).

We have analyzed ambient aerosol samples (PM_{10}) collected from the megacity Chennai, India (13.04 °N 80.17 °E) for molecular distributions of dicarboxylic acids using gas chromatography (GC) and GC/mass spectrometer. The details of the aerosol sampling and analytical procedures are described elsewhere (Pavuluri et al., 2010). Briefly, PM_{10} samples

were collected on the rooftop of the Mechanical Sciences Building (18 m above the ground) at the Indian Institute of Technology Madras campus. The sampling location was not close to a MSW incineration site; rather it is more typical of city background. We detected *t*-Ph with very high concentrations ranging from 7.6-168 ng m^{-3} (average 45 ng m^{-3}) in winter and 24-158 ng m^{-3} (61 ng m^{-3}) in summer. *t*-Ph was found as the second most abundant dicarboxylic acid in most of the samples, in particular in nighttime samples, following oxalic acid (114-696 ng m^{-3}), which is the most abundant water-soluble organic in particles from many locations around the globe (e.g., Kawamura and Ikushima, 1993). Although phthalic acid (Ph) has been reported as the most abundant aromatic diacid in particles globally, Ph and isophthalic (*i*-Ph) acids were rather minor isomers in Chennai aerosols (Pavuluri et al., 2010). *t*-Ph concentrations in Chennai aerosols are 10 times greater than those reported for urban aerosols (TSP) from North America and East Asia as well as marine aerosols (TSP) from the Western Pacific and Southern Ocean (Table 1).

In South Asia, in particular, India, the generation of MSW is very high with a rate of 0.5-0.7 kg $capita^{-1}$ day^{-1} (48 Tg per year; 1997), in which plastics account for 2-8%. >90% of MSW are disposed into open landfills without regulations (Jha et al., 2008), and local residents very often set light to the garbage to avoid the stench from the wastes and the occurrence of mosquito that transmits malaria. Plastics may readily decompose into monomers under open-fire conditions. These monomers can be emitted to the atmosphere and adsorbed onto pre-existing particles. Interestingly, mass concentrations of *t*-Ph in Chennai aerosols were 55-1620 ng mg^{-1} (average 466 ng mg^{-1}) in winter and 305-1890 ng mg^{-1} (764 ng mg^{-1}) in summer. These values are comparable to those reported for smoke particles from plastic bags and wastes as stated above (Simoneit et al., 2005).

These observations indicate that open burning of MSW is the major source of *t*-Ph in the aerosols from Chennai rather than emissions from fossil fuel combustion and secondary

production via the oxidation of its precursor compounds. Thus, the disposal of MSW into landfills followed by firing may deteriorate the local air quality of South India and can lead to climate changes at regional to global scale by emitting significant amount of *t*-Ph and other organics, which can further be oxidized to result in more water-soluble organic acids. For example, *t*-Ph may degrade into maleic/fumaric acid followed by oxalic acid under photo-Fenton oxidation (Thiruvenkatachari et al., 2007). It is of interest to note that the growth rate of municipal waste generation is 61% from 1996 to 2002 (Jha et al., 2008). There is an urgent need to better understand the effect of solid waste burning on the air quality, public health and potential impact on climate changes. Finally, it is also essential for policy makers to enhance the effective recycling of plastic wastes and to regulate the field burning of MSW disposal in South Asia.

References

Aggarwal, S.G., Kawamura, K., 2008. Molecular distributions and stable carbon isotopic compositions of dicarboxylic acids and related compounds in aerosols from Sapporo, Japan: Implications for photochemical aging during long-range atmospheric transport. Journal of Geophysical Research 113, D14301, doi:10.1029/2007JD009365.

Aizenshtein, E.M., 2006. World production and consumption of polyster fibres and thread. Fibre Chemistry 38, 264-271.

Braun, D., 2004. Poly(vinyl chloride) on the way from the 19th century to the 21st century. Journal of Polymer Science: Part A: Polymer Chemistry 42, 578-586.

Heck, H.D'A., Tyl, R.W., 1985. The induction of bladder stones by terephthalic acid, dimethyl terephthalate, and melamine (2,4,6-triamino-s-triazine) and its relevance to risk assessment. Regulatory Toxicology and Pharmacology 5, 294-313.

Jha, A.K., Sharma, C., Singh, N., Ramesh, R., Purvaja, R., Gupta, P.K., 2008. Greenhouse gas emissions from municipal solid waste management in Indian mega-cities: A case study of Chennai landfill sites. Chemosphere 71, 750-758.

Kawamura, K., Ikushima, K., 1993. Seasonal changes in the distribution of dicarboxylic acids in the urban atmosphere. Environmental Science and Technology 27, 2227-2235.

Pavuluri, C.M., Kawamura, K., Swaminathan, T., 2010. Water-soluble organic carbon, dicarboxylic acids, ketoacids and α-dicarbonyls in the tropical Indian aerosols. Journal of Geophysical Research 115, D11302, doi:10.1029/2009JD012661.

Simoneit, B.R.T., Kobayashi, M., Mochida, M., Kawamura, K., Lee, M., Lim, H.-J., Turpin, B.J., Komazaki, Y., 2004. Composition and major sources of organic compounds of aerosol particulate matter sampled during the ACE-Asia campaign. Journal of Geophysical Research 109, D19S10, doi:10.1029/2004JD004598.

Simoneit, B.R.T., Medeiros, P.M., Didyk, B.M., 2005. Combustion products of plastics as indicators for refuse burning in the atmosphere. Environmental Science and Technology 39, 6961-6970.

Thiruvenkatachari, R., Kwon, T.O., Jun, J.C., Balaji, S., Matheswaran, M., Moon, I.S., 2007. Application of several advanced oxidation processes for the destruction of terephthalic acid (TPA). Journal of Hazardous Materials 142, 308-314.

Wang, H., Kawamura, K., Yamazaki, K., 2006. Water-soluble dicarboxylic acids, ketoacids and dicarbonyls in the atmospheric aerosols over the Southern Ocean and Western Pacific Ocean. Journal of Atmospheric Chemistry 53, 43-61.

Acknowledgements

This work is partly supported by funds from JSPS (19204055) and by the Environment Research and Technology Development Fund (B-0903) of the Ministry of the Environment, Japan.

K. Kawamura* and C.M. Pavuluri

Institute of Low Temperature Science, Hokkaido University,

N19 W08, Kita-ku, Sapporo 060 0819, Japan.

*Corresponding author. E-mail: kawamura@lowtem.hokudai.ac.jp

Table 1. Concentrations of terephthalic acid (*t*-Ph) in atmospheric particles collected from different locations over the world.

Sample locale	Concentrations			
	ng m^{-3}		ng mg^{-1}	
	Range	Average	Range	Average
Chennai, India[a]				
Jan.-Feb., 2007	7.6-168	45	55-1620	466
May 2007	24.2-158	61.3	305-1890	764
Gosan site (Jeju Island), South Korea[b,c]				
April 2001		11		47
Sapporo, Japan				
April 2001[b,c]		6.3		43
Sept. 2001[b,c]		0.2		6.5
May-July 2005[d,e]	0.01-5.6	2.6	0.12-79	30
Southern Ocean[f]				
Nov. 1994-Feb. 1995	0.01-0.12	0.04		0.48
Western Pacific Ocean[f]				
Nov. 1994-Feb. 1995	<0.01-0.95	0.11		2.8
Santiago, Chile[b]				
April 1997		3.2		NA[g]
April 1998		0.5		NA
April 1999		21		NA
Nov. 2000		1.1		NA
Los Angeles, CA[b]				
Sept. 1993	0.9-17	5.4		NA
Corvallis, OR[b]				
June 1979		0.1		NA

[a]This study; [b]Simoneit et al., 2005; [c]Simoneit et al., 2004; [d]Aggarwal and Kawamura, 2008; [e]Aggarwal and Kawamura, unpublished data (2005); [f]Wang et al., 2006; [g]NA: Not available.