Particle Size Effects on the Deposition Ratios of Airborne Particles in the Respiratory Tract

Kazutoshi Sugita,*[,a] Sumio Goto,[b] Osamu Endo,[c] Daisuke Nakajima,[b] Hirofumi Yajima,[d] and Tadahiro Ishii[d]

[a]Dia Analysis Service Inc., 8–5–1, Chuo, Ami-machi, Inashiki-gun, Ibaraki 300–0332, Japan, [b]National Institute for Environmental Studies, 16–2, Onogawa, Tsukuba-shi, Ibaraki 305–8506, Japan, [c]National Institute of Public Health, 4–6–1, Shirokanedai, Minato-ku, Tokyo 108–8638, Japan, and [d]Department of Applied Chemistry, Faculty of Science, Tokyo University of Science, 1–3, Kagurasaka, Shinjuku-ku, Tokyo 162–8601, Japan

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Particle size adjusting equipment was manufactured to determine the deposition ratio of airborne particles in the respiratory tract. This equipment is composed of an Andersen low-pressure impactor, one 100 l stainless steel tank, two filter holders, one Tedlar bag, one flow meter, and two pumps. We measured deposition ratios of benzo[*a*]pyrene (BaP) in size-fractionated particles in the respiratory tract using this equipment together with a Hans-Rudolph mask. BaP in particles was extracted using the sonication method and measured with the column concentration HPLC/spectrofluorometric detection method. BaP concentrations in PM0.52 (particles smaller than 0.52 μm) ranged from 0.025 to 0.193 ng/m^3 (CV: 72%), and the CV of deposition ratios of BaP was 23%, (average 44.5%). Furthermore, we measured the deposition ratios of BaP in 6 different sized particles. While high deposition ratios of BaP were observed in PM3.9 air, PM2.5 air, and PM0.76 air, low deposition ratios were found in PM0.52 air, PM0.33 air, and PM0.13 air. We obtained similar results to the theoretical deposition ratio calculated from lung models.

Key words —— airborne particles, particle size, deposition ratio

*To whom correspondence should be addressed: Dia Analysis Service Inc., 8–5–1, Chuo, Ami-machi, Inashiki-gun, Ibaraki 300–0332, Japan. Tel.: +81-29-887-1017; Fax: +81-29-887-5381; E-mail: 6306378@cc.m-kagaku.co.jp

INTRODUCTION

Ambient air contains various trace carcinogenesis-related compounds in airborne particles. Humans inhale these compounds into body continuously, raising concerns over chronic effects such as lung cancer caused by long-term exposure. It is known that particle sizes are distributed over a wide range from a few nanometers to a few 10 μm or more.[1] In particular, the danger of the particles to the human body was reported to be related to the existence of particles smaller than 2.5 μm (PM2.5) in size, compared with those smaller than 10 μm (PM10).[2–4] In addition, atmospheric pollution has been suspected as one of the causes of lung cancer.[5] It is therefore important to clarify the degree of exposure to carcinogenesis-related compounds in airborne particles surrounding humans.

In this study, we manufactured the equipment for preparing air and removing large particles of more than a specified particle diameter ($\times$ μm) from the ambient air. The resulting air is called PMx air. Using this air, we collected particles in inhaled and exhaled air with a Hans-Rudolph mask[6] and measured the deposition ratios of benzo[*a*]pyrene in airborne particles in the respiratory tract.

MATERIALS AND METHODS

Reagents —— Polycyclic aromatic hydrocarbon (PAH) standard reagent: Pyrene, benzo[*a*]anthracene (BaA), and benzo[*a*]pyrene (BaP) were purchased from Wako Pure Chemical (Japan), and benzo[*k*]fluoranthene (BkF), dibenzo[*a*,*h*]anthracene (dBahA), and benzo[*b*]chrysene (BbC) were manufactured by Koch-Light Laboratories, and benzo[*ghi*]perylene (BghiP) was obtained from Aldrich Chemical. As a solvent for extraction, benzene for residual agricultural chemical tests (Wako Pure Chemical) and ethanol for residual agricultural chemical tests (Kishida Chemical, Osaka, Japan) were used. In addition, as a solvent for sample preparation for high-performance liquid chromatography (HPLC), acetonitrile for residual agricultural chemical tests (Wako Pure Chemical) was used. As the mobile phase for HPLC, acetonitrile for HPLC (Kokusan Chemical, Tokyo, Japan) and ion-exchange distilled water were used.

Preparation of Experimental Air and Sample Collection —— We manufactured the particle size adjusting equipment (PSAE), which is composed of

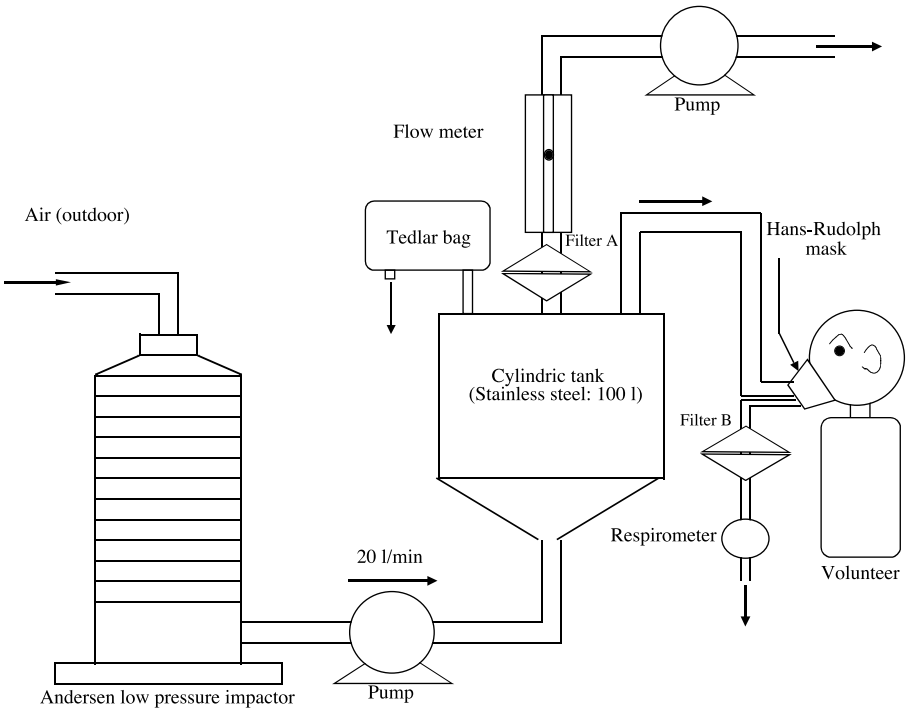

Fig. 1. Particle Size Adjusting Equipment to Determine the Deposition Ratio of Airborne Particles

an Andersen low-pressure impactor (Tokyo Dylec, Tokyo, Japan), one 100 l stainless steel tank, two filter holders, one Tedlar bag, one flow meter, and two pumps (Fig. 1). Ambient air containing airborne particles was drawn into the Andersen low-pressure impactor. Particles bigger than the set particle size were removed by the stage selection of the impactor, and then air containing airborne particles smaller than the set particle size, *i.e.*, PMx air, was prepared.

The particles in PMx air were collected on filter A (Teflon binder filter T60A20, Pallflex Products Co. CT, U.S.A.) in Fig. 1 which is 35 mm diameter using a low-noise and low-volume air sampler. The PMx air containing airborne particles was inhaled through a Hans-Rudolph mask, and simultaneously particles in the exhaled air were collected on filter B in Fig. 1 for 2 hr.[6] After sampling, each filter was stored in a freezer at –80°C until extraction.

Extraction of BaP and Sample Preparation for HPLC Analysis —— The samples were removed from the freezer and allowed to stand until returning to room temperature. The PAHs were extracted from these filters using the sonication method with benzene and ethanol (3 : 1, v/v) for 20 min. After centrifuging for 16 min, aliquots of extracts were dried under a gentle stream of nitrogen. They were stored at –80°C until HPLC analysis, in which the residues were redissolved in acetonitrile.

HPLC Analysis for BaP —— BaP contents in the extracts were measured using the column concentration HPLC/spectrofluorometric detection method described in a previous paper.[7] A stainless steel column (internal diameter: 4.6 mm, length: 30 mm) filled with ODS-60-5 obtained from Tokyo Kasei Kogyo was used as a concentration column. In addition, another stainless steel column (internal diameter: 4.6 mm, length: 250 mm) filled with ODS-60-5 was used as a separation column. An acetonitrile, deionized distilled water, and acetonitrile-water (1 : 1) mixture was used for the mobile phase. The excitation and emission wavelengths were set automatically to detect the target PAH with high sensitivity and selectivity.

RESULTS AND DISCUSSION

PM0.52 Air Deposition Ratios in the Respiratory Tract

To confirm that the flow path of the PSAE was smooth, the deposition ratio in the respiratory tract was measured using the PSAE without impactors. Figure 2 shows chromatograms of inhaled and exhaled air samples collected using the PSAE without impactors. The presence of seven PAHs was confirmed with good resolution. As shown in Fig. 2, BaP

in particular was detected with sufficient sensitivity. BaP is not only a typical carcinogen in airborne particles, but is also suspected of being an environmental endocrine disruptor and being toxic.[8] Therefore measurements of the deposition of particles in the respiratory tract were carried out to determine BaP content. Five separate tests of PM0.52 air were performed. The results are summarized in Table 1. Although the concentration of BaP in fine airborne particles varied considerably over the range from 0.025 ng/m^3 to 0.193 ng/m^3 [average 0.116 ng/m^3 and coefficient of variation (CV) 72%], the mean deposition ratio of BaP was determined to be 44.5% with a CV of 23%. From these results, the deposition ratio with this PSAE could be measured using actual airborne particles.

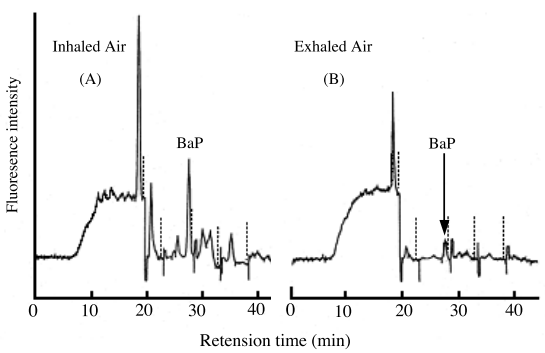

Fig. 2. HPLC Chromatograms of BaP in Suspended Particles from Size-Prepared Air (Inhaled Air) and Exhaled Air

Dependence of Deposition Ratio on Particle Size

To evaluate the effect of particle size on the deposition ratios of particles in the respiratory tract, BaP concentrations in particles were measured for seven different types of PMx air (PM8.5, PM3.9, PM2.5, PM0.76, PM0.52, PM0.33, and PM0.13) and intact air, and simultaneously BaP concentrations in exhaled particles were measured. These results are summarized in Table 2. The levels of BaP in particles varied considerably with particle size, and the deposition ratios for PM3.9 air, PM2.5 air, and PM0.76 air tended to be higher than those for PM0.52 air, PM0.33 air, and PM0.13 air. Initially,

Table 1. Variation of PM0.52 Deposition Ratio in the Respiratory Tract

Test no.	Volume of respiration	BaP concentration (ng/m^3)			Deposition ratio
	(l)	Inhaled (A)	Exhaled (B)	Deposited amount	(%)
1	1.043	0.145	0.078	0.067	46.2
2	1.102	0.188	0.102	0.086	45.7
3	1.044	0.029	0.021	0.008	27.6
4	1.005	0.193	0.087	0.106	54.9
5	1.223	0.025	0.013	0.012	48.0
Mean	1.083	0.116	0.060	0.056	44.5
S.D.	0.085	0.083	0.040	0.044	10.1
CV (%)	7.9	71.9	67.2	78.9	22.8

PM0.52, particles smaller than 0.52 μm in diameter prepared with PSAE.

Table 2. Effect of Particle Diameter on the Deposition Ratio in the Respiratory Tract

Sample	Particle size	No. of	Volume of	BaP concentration (ng/m^3)			Deposition
	(μm)	experiments	respiration (l)	Inhaled (A)	Exhaled (B)	Deposited amount (A − B)	ratio (%)
Intact air[a)]	—	4	0.911	0.511	0.124	0.387	68.1
PM8.5	$\leq$ 8.5	1	1.023	0.662	0.318	0.345	52.0
PM3.9	$\leq$ 3.9	2	0.972	2.637	0.211	2.426	76.7
PM2.5	$\leq$ 2.5	2	0.935	2.324	0.160	2.164	93.9
PM0.76	$\leq$ 0.76	2	1.051	2.700	0.238	2.462	83.4
PM0.52	$\leq$ 0.52	5	1.083	0.116	0.060	0.056	44.5
PM0.33	$\leq$ 0.33	2	1.266	0.130	0.094	0.037	28.7
PM0.13	$\leq$ 0.13	2	1.816	0.137	0.068	0.070	49.7

a) samples prepared without impactor.

we believed that the deposition ratios would increase with decreasing particle diameter, but the deposition ratios for smaller particles tended to be low. Yu and Diu also reported the lowest deposition ratios at particle sizes of 0.1–1 μm using a lung model.[9] Some model studies found that the deposition of larger particles (> 2.5 μm) was dependent on impaction, the deposition of medium-sized particles (2.5–0.5 μm) was dependent on sedimentation, and the deposition of smaller particles (< 0.5 μm) was dependent on diffusion.[10,11] The low deposition ratios for smaller particles (PM0.52, PM0.33, and PM0.13) in this study can be explained by those results from lung model studies. In this study, it is possible that some larger particles were lost when the prepared air passed through the tube and mask to the respiratory tract. Therefore it is necessary to investigate the deposition ratios in detail using adjusted particles for each size range.

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