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COMPOSITION OF ENVIRONMENTAL TOBACCO SMOKE (ETS) FROM INTERNATIONAL CIGARETTES PART II: NINE COUNTRY FOLLOW-UP

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The environmental tobacco smoke (ETS) generated by cigarettes from nine countries was examined to determine selected component yields and ETS marker ratios. The countries tested included: Argentina, Australia, Brazil, Canada, China, England, Japan, Korea, and the United States. Six leading brand styles from each country were smoked in an environmental test chamber. Concentrations of the gas phase ETS components CO, NO, NO₂, nicotine, 3-ethenylpyridine, myosmine, and total volatile organic compounds (TVOC) were measured. Particle mass concentrations were also measured along with the particulate phase markers ultraviolet particulate matter (UVPM), fluorescent particulate matter (FPM), and solanesol. The variation in the ETS generated by cigarettes from these nine countries was greater than observed in a previous test. Ratios of gravimetrically determined respirable suspended particles (RSP) to the surrogate standard concentrations for UVPM and FPM averaged 7.2 and 42, respectively. The average ratio of RSP to solanesol was determined to be 38. ETS component yields were relatively consistent among the countries tested and averaged 14, 59, 41, 1.3, and 0.4 mg/cig. for RSP, CO, TVOC, NO, and NO₂, respectively. ©1998 Elsevier Science Ltd

INTRODUCTION

A number of investigators have performed studies to characterize the environmental tobacco smoke (ETS) generated by American, Canadian, and English cigarettes (Nelson et al. in press; Hodgson et al. 1996; Heavner et al. 1996; Leaderer and Hammond 1991; Martin et al. 1997; Phillips et al. 1994; Rickert et al. 1990). However, few studies have focused on the generation of ETS by cigarettes from other nations. In a recent study, ETS generated by cigarettes from two Asian and eight European countries was measured and compared to the ETS generated by American cigarettes (Nelson et al. 1997). Although there were a few notable exceptions, the composition of ETS among the countries tested was similar.

The ability to differentiate between the particles which arise from the smoking of tobacco products (ETS-RSP) and respirable suspended particles (RSP) derived from other sources is important when performing studies to assess the impact of ETS on indoor particulate matter levels. The three most widely used ETS-RSP markers for apportionment are: ultraviolet particulate matter (UVPM) (Carson and Erikson 1988; Conner et al. 1990; Eatough et al. 1992; Eatough 1993; Guerin et al. 1992; Heavner et al. 1996; Hedge et al. 1993, 1994, 1996; Ingebretsen et al. 1988; Jenkins et al. 1996; Ogden et al. 1990, 1996; Phillips et al. 1994, 1997; Sterling et al. 1996), fluorescent particulate matter (FPM) (Guerin et al. 1992; Heavner et al. 1996;

Jenkins et al. 1996; Ogden et al. 1990, 1996; Phillips et al. 1994, 1996, 1997; Sterling et al. 1996), and solanesol (Eatough 1993; Guerin et al. 1992; Heavner et al. 1996; Jenkins et al. 1996; Ogden and Maiolo 1989, 1992; Ogden et al. 1990, 1996; Phillips et al. 1994, 1996, 1997; Sterling et al. 1996; Tang et al. 1990). UVPM is determined by extracting the PTFE membrane filters, used to collect ETS particles, with methanol and determining the absorbance at 325 nm (Conner et al. 1990; Ogden et al. 1990, 1996). FPM uses the same extract, but fluorescence is examined instead ($\lambda_{\text{excitation}} = 300 \text{ nm}$, $\lambda_{\text{emission}} = 420 \text{ nm}$) (Ogden et al. 1990, 1996). Solanesol is a trisesquiterpenoid alcohol present in the particulate phase of ETS (Ogden and Maiolo 1989). It is quantified by liquid chromatography (LC) UV detection ($\lambda = 205 \text{ nm}$) of an aliquot from the methanol extract of the ETS particles (Ogden and Maiolo 1992; Ogden et al. 1996).

Previously, in the absence of specific apportionment factors for different nations, investigators have had to assume that the values obtained from studies of American cigarettes applied to ETS measurements made in other nations. ETS apportionment factors for UVPM, FPM, and solanesol markers have been determined for 11 countries (Nelson et al. 1997); however, most of these countries were European. Little data on apportionment ratios from other regions of the world have been published. Furthermore, among international cigarettes, there is a lack of per cigarette yield data for commonly measured ETS components.

Concentrations of commonly measured ETS components were measured, and the ratios of UVPM, FPM, and solanesol to ETS-RSP for cigarettes from several countries in Asia, Australia, and North and South America were determined in this study. In addition, yields of RSP, CO, total volatile organic compounds (TVOCs), and oxides of nitrogen were determined for the cigarettes tested.

METHODS

The methods used to perform the current investigation were published previously (Nelson et al. 1997). An abbreviated description of the methods follows here.

Environmental chamber

The studies reported here were performed in a 45 m³ environmental chamber operated in an unventilated mode (0.08 ± 0.01 air changes per hour, as measured by CO decay). Air within the chamber was mixed by a

fan located in one corner of the chamber. The initial chamber temperature (20°C) and relative humidity (57.5%) were controlled prior to the start of each run. Since the heating, ventilating, and air conditioning system was not operated during a run, temperature and relative humidity drifted slightly during the course of a run. Across experiment runs, the average temperature and relative humidity during the sampling period was $23.4 \pm 0.3^\circ\text{C}$ and $56 \pm 6\%$, respectively.

Smoke generation

Three panels of ten smokers were recruited locally to smoke the cigarettes. Six leading cigarette brand styles were identified for each of the nine countries tested. At least one day prior to testing the cigarettes for a given country, the smokers each received one pack of cigarettes from their normal "tar" category; i.e., a full-flavor smoker was given a full-flavor cigarette.

Six smokers entered the chamber prior to each experiment run. Following a 10-min background collection period, the smokers each lit one cigarette of the brand they had received in advance. They then smoked the cigarette *ad libitum* to a line 3 mm from the overwrap. At 20 min, the smokers quickly exited the chamber, which was then resealed. The smokers' exit from the chamber resulted in minimal loss of ETS from the chamber. Pumps for discrete samples started at this point and continued for 60 min. Three replicate experiments were performed for each country.

Analytes

All samples were collected from sampling ports in the wall of the chamber. Concentrations of particles, carbon monoxide, carbon dioxide, oxides of nitrogen, and TVOCs were determined using continuous emissions monitors described elsewhere (Nelson et al. in press). The gas analyzers were calibrated using a Series 2020 Computerized Emissions Monitoring Calibration System (EnviroNics, West Willington, CT) and precision blended calibration gases (Scott Specialty Gases, Raleigh, NC).

Five gravimetric RSP samples were collected from each run in the chamber. An inertial impactor with a 2.5- μm median cut point was located upstream from each filter. After sampling, all the filters were extracted with methanol and analyzed further by LC with UV detection for UVPM and solanesol, and with fluorescence detection for FPM (Ogden et al. 1996).

Duplicate samples of 3-ethenylpyridine, nicotine, and myosmine were collected on XAD-4 sorbent tubes

Table 1. Average concentration of gas phase analytes in environmental chamber. Standard deviation follows each measurement in parentheses.

Country	CO ($\mu\text{L/L}$)	TVOC ($\mu\text{L/L}$)	NO (nL/L)	NO ₂ (nL/L)	3-EP ($\mu\text{g/m}^3$)	Nicotine ($\mu\text{g/m}^3$)	Myosmine ($\mu\text{g/m}^3$)
Argentina	6.6 (0.4)	3.0 (0.3)	134 (2)	22 (1)	42 (2)	137 (19)	8.7 (0.7)
Australia	5.4 (0.3)	2.4 (0.1)	ND [†]	ND [†]	34 (2)	121 (17)	6.0 (0.4)
Brazil	6.9 (0.2)	3.0 (0.4)	173 (38)	28 (9)	45 (7)	132 (44)	10 (1)
Canada	6.0 (0.6)	2.4 (0.2)	66 ^{††}	11 ^{††}	33 (3)	125 (51)	6 (1)
China	7.3 (0.2)	3.1 (0.3)	106 (9)	16 (3)	31 (5)	101 (31)	5 (1)
England	7.1 (0.1)	3.3 (0.2)	138 (10)	27 (8)	41 (7)	153 (16)	7.9 (0.9)
Japan	6.4 (0.3)	2.8 (0.5)	130 (23)	38 (5)	40 (7)	154 (30)	8 (2)
Korea	6.5 (0.1)	2.8 (0.1)	ND [†]	ND [†]	45 (4)	145 (25)	9 (1)
United States	6.4 (0.2)	3.0 (0.2)	154 (7)	30 (2)	39 (3)	139 (37)	8 (2)

[†] = not determined - the NO_x analyzer was nonfunctional during tests of Australian and Korean cigarettes.

^{††} The NO_x was nonfunctional for the initial three runs with Canadian cigarettes. One makeup run was performed, and the single value obtained appears in the table.

(SKC, Eighty Four, PA) from each run. At the end of the experiment, the contents of the sorbent tubes were extracted with ethyl acetate modified with triethylamine and analyzed using a gas chromatograph equipped with a nitrogen-phosphorus detector (Ogden et al. 1996).

Yield determination

Yields for each analyte measured in real-time were determined following the method described by Martin et al. (1997). In brief, yields from each run were determined by multiplying the background-corrected, peak analyte concentrations by the chamber volume (43.4 m³ with recirculation system blocked) and dividing the product by the number of cigarettes smoked. The presence of people in the chamber contributes some load of CO, TVOCs, and particles to the chamber. "Yields" calculated for the blank runs (i.e., no ETS) were typically 5% of the yields calculated from smoking runs and ranged from 1% of the average smoking run yield for NO to 9% for TVOCs. Blank "yields" were subtracted from the yields obtained during smoking runs to determine true ETS yield per cigarette.

RESULTS

ETS composition

Average background-corrected gas-phase analyte concentrations are shown in Table 1. In general, analyte concentrations were similar when the cigarettes of each country were smoked. For example, the average CO concentration across the nine countries is 6.6 ppm (6.6 $\mu\text{L/L}$) with a relative standard deviation of 9%.

A notable exception to the agreement among countries is seen for the oxides of nitrogen from Canadian cigarettes. Due to problems with the NO_x analyzer during initial testing of those cigarettes, a makeup session was scheduled. Due to a limited cigarette supply and scheduling problems, the smokers for the make-up session were unable to receive Canadian cigarettes prior to testing. However, the smokers smoked the same brand styles of Canadian cigarettes that they had smoked in a previous session. Good agreement between the other analytes measured and those obtained in the makeup session was observed. The greatly reduced NO and NO₂ concentrations are due to compositional differences between Canadian and other cigarettes. Canadian cigarettes are composed almost exclusively of Canadian flue-cured tobacco, most of which is grown in a limited geographic area (Rickert et al. 1990). Due to differences in agricultural practices between Canada and other countries, Canadian tobacco is low in nitrates - the primary precursor to ETS oxides of nitrogen (Rickert et al. 1987).

Average background-subtracted particulate-phase analyte concentrations appear in Table 2. Values reported for UVPM and FPM are not the "ETS-attributable RSP concentration" that would typically be reported in the literature, but instead are the equivalent concentrations of the surrogate standard (Ogden et al. 1990). As with the gas-phase analytes, there is relatively little difference among the countries in terms of analyte concentration in the chamber. The notable exception is the solanesol concentration measured for Canadian cigarettes. Differences in growing conditions

Table 2. Average concentration of particulate phase analytes in environmental chamber. Standard deviation follows each measurement in parentheses. All units are $\mu\text{g}/\text{m}^3$.

Country	R-t RSP [†]	Grav. RSP	UVPM ^{††}	FPM ^{††}	Solanesol
Argentina	1567 (180)	1509 (188)	211 (24)	36 (3)	61 (9)
Australia	1469 (85)	1372 (82)	187 (14)	32 (3)	48 (4)
Brazil	1529 (116)	1480 (36)	217 (6)	37 (1)	50 (3)
Canada	1315 (86)	1235 (109)	172 (17)	31 (2)	19 (2)
China	2029 (211)	1827 (270)	241 (33)	40 (4)	35 (8)
England	1508 (67)	1421 (54)	200 (8)	34 (2)	42 (2)
Japan	1363 (140)	1303 (148)	185 (20)	31 (4)	36 (4)
Korea	1553 (73)	1397 (152)	198 (16)	37 (4)	39 (5)
United States	1531 (78)	1445 (161)	206 (24)	35 (4)	48 (7)

[†] Real-time RSP by TEOM.

^{††} Results are surrogate standard concentration (Ogden et al. 1990).

and agricultural practices between Canada and other tobacco-growing countries (Rickert et al. 1987; Rickert and Kaiserman 1994; Ogden and Maiolo 1989) probably are responsible for the low solanesol content of Canadian cigarettes.

Despite similarities in aggregate ETS yields among countries, variations in concentrations among individual brand styles may be significantly greater than that seen among the groups of brands smoked to represent each country (Martin et al. 1997).

Cigarettes from the United States were smoked in both the current and a previous study (Nelson et al. 1997). With the exception of solanesol, particulate-phase component concentrations agree well (within ~10%) between the two studies. With the exceptions of nicotine, myosmine, and NO, agreement of gas-phase components between the two studies was equivalent to that observed for the particulate-phase components.

To determine whether statistically significant differences in analyte concentrations existed among the countries, analysis of variance (ANOVA) was performed. Where significant differences were found ($p < 0.05$), a Bonferroni-normalized multiple comparison test was performed to determine which countries differed ($p < 0.05$). Unfortunately, the number of observations ($n=27$) is low relative to the number of comparisons ($n=36$) to be made among the countries. As a result, the sensitivity of the multiple comparison test is limited and may not be sufficient to identify true differences among the countries.

ANOVA indicated that significant differences in analyte concentration existed among countries for all analytes except nicotine. For 3-ethenylpyridine and FPM, differences among groups were detected by

ANOVA but not by the Bonferroni multiple comparison test. For other analytes, relatively few differences among groups were detected. For example, Canadian cigarettes produced significantly less TVOCs than English cigarettes and had a lower UVPM response than Chinese cigarettes.

Australian cigarettes produced statistically significantly lower CO concentrations than the cigarettes from all the other countries tested with the exception of Canada, where the difference was not statistically significant. Canadian cigarettes generated significantly less CO than cigarettes from both China and England. Chamber concentrations of myosmine were significantly higher for Brazilian cigarettes than for Australian, Canadian, and Chinese cigarettes. In addition, Chinese cigarettes produced less myosmine in the chamber than Argentinean cigarettes.

Chinese cigarettes produced significantly less NO than Brazilian cigarettes, and Japanese cigarettes generated higher concentrations of NO₂ than Argentinean and Chinese cigarettes. Canadian cigarettes were not included in the ANOVA for either oxide of nitrogen due to the lack of data collected similarly to the other cigarettes. However, given the marked reduction in NO and NO₂ relative to cigarettes from the other countries and the results obtained previously by Rickert et al. (1987), it is likely that the Canadian cigarettes generate less NO and NO₂ than the cigarettes from the other countries examined.

Among the particulate-phase analytes, Chinese cigarettes generated significantly more RSP than Canadian and Japanese cigarettes and higher UVPM than Canadian cigarettes. Smoking of the Canadian

Table 3. RSP/"surrogate standard" ratios for each country are followed by their standard deviation in parentheses.

Country	UVPM	FPM	Solanesol
Argentina	7.2 (0.3)	43 (2)	25 (1)
Australia	7.3 (0.2)	43 (1)	29 (1)
Brazil	6.8 (0.3)	40 (2)	30 (1)
Canada	7.3 (0.3)	41 (2)	68 (3)
China	7.6 (0.3)	45 (3)	53 (4)
England	7.1 (0.3)	42 (2)	34 (1)
Japan	7.1 (0.2)	42 (1)	36 (3)
Korea	7.0 (0.4)	38 (2)	35 (2)
United States	7.0 (0.2)	41 (2)	30 (1)
Overall	7.2 (0.2)	42 (2)	38 (14)

Table 4. Average ETS yields of components measured in real-time. Standard deviation follows each measurement in parentheses. All units are mg/cigarette.

Country	RSP	CO	TVOC	NO	NO ₂
Argentina	14.5 (1.0)	61 (3)	43 (4)	1.4 (0.0)	0.3 (0.0)
Australia	13.7 (0.4)	49 (2)	35 (3)	ND [†]	ND [†]
Brazil	14.3 (1.2)	62 (2)	44 (6)	1.7 (0.4)	0.4 (0.2)
Canada	12.5 (1.3)	54 (6)	34 (3)	0.65 ^{††}	0.16 ^{††}
China	18.7 (1.7)	66 (2)	46 (3)	1.1 (0.1)	0.3 (0.1)
England	14.0 (1.0)	64 (1)	47 (3)	1.4 (0.1)	0.4 (0.1)
Japan	12.6 (1.3)	59 (3)	40 (7)	1.4 (0.2)	0.6 (0.1)
Korea	13.9 (0.6)	59 (3)	40 (1)	ND [†]	ND [†]
United States	14.5 (0.5)	58 (1)	44 (2)	1.5 (0.1)	0.5 (0.1)

[†] = not determined - the NO_x analyzer was nonfunctional during tests of Australian and Korean cigarettes.

^{††} The NO_x was nonfunctional for the initial three runs with Canadian cigarettes. One makeup run was performed, and the single value obtained appears in the table.

cigarettes resulted in statistically significantly lower solanesol concentrations in the chamber than cigarettes from all the other nations. In addition, Chinese cigarettes generated less solanesol than cigarettes from England, Japan, and Korea.

ETS-RSP apportionment factors

The ratio of the surrogate standard equivalent concentration (Table 3) to the actual ETS-RSP concentration is used to apportion the fraction of RSP attributed to ETS (Heavner et al. 1996). Likewise, if solanesol concentrations are to be used to apportion RSP from ETS, the ratio of the solanesol concentration to the ETS-RSP concentration must be known. Ratios of gravimetric RSP and both the surrogate standard and solanesol concentrations were obtained for each filter collected for a given country. Those ratios were then averaged (Table 4). As was true for the analyte concen-

trations in ETS, the ratios among the various markers and ETS-RSP are also consistent.

The average UVPM factor across all the countries was 7.2 with a standard deviation of 0.2. Results from this test are lower than obtained previously (Nelson et al. 1997) and are somewhat lower than the factors of 8.0 (Ogden et al. 1990) and 7.5 (Heavner et al. 1996) determined by others. ANOVA followed by a Bonferroni multiple comparison test indicated that the UVPM:RSP ratio for Chinese cigarettes was significantly different from all other countries except Australia and Canada. In addition, the UVPM:RSP ratio obtained from Brazilian cigarettes was significantly lower than the ratio obtained from Argentinean, Australian, Canadian, and Chinese cigarettes. Despite the statistical significance of these differences, the apportionment factors are within 6% of the mean value. As a result, the overall apportionment factor of 7.2 may reasonably approximate the factors from those countries.

For American cigarettes, a value of 8.5 ± 0.8 was obtained in the previous study of international cigarettes (Nelson et al. 1997). Differences between the previous and current value may have a number of origins. The top six U.S. brand styles changed between the two studies, and this may have had a small impact on the UVPM factor. In addition, the cigarettes would have been blended from different crop-years of tobacco. The variation may also be indicative of the normal variation in UVPM measurement over time. The average across both studies (7.8) is in the middle of the range of other literature values.

Like UVPM, apportionment factors determined for FPM are reasonably consistent across the nine countries (42 ± 2). This apportionment factor is between the recently reported values of 39 (Heavner et al. 1996) and 45 ± 2 (Nelson et al. 1997). Statistical analysis indicated the factors obtained for Chinese and Korean cigarettes were significantly different from all the other countries (except Korea vs. Brazil). In addition, Brazilian FPM apportionment factors were different from those obtained for Argentinean and Australian cigarettes. As with UVPM, the factors do not differ greatly from the mean determined value on a percentage basis.

The average RSP:solanesol ratio was 38 ± 14 . Unlike the other markers, there was considerable variation in the ratios obtained among the different countries. The average ratio determined from this study is comparable to that determined from the previous study (43 ± 6) and close to other previously reported values of 35 ± 5 (Nelson et al. 1994) and 38.5 (Ogden et al. 1990). Statistically significant differences were detected among 30 of the 36 possible combinations of countries. ETS from Canadian cigarettes has been reported to be low in solanesol (Rickert 1991); however, substantial differences were noted among other countries as well. For example, the ratio obtained for Chinese cigarettes was nearly twice that observed for Argentinean cigarettes. As a result, if ETS apportionment is to be performed using solanesol for the countries tested here, it may be prudent to use country-specific apportionment factors.

ETS yields

Yields of ETS components per cigarette are an important factor used when modeling the impact of ETS on indoor air. Although data describing the yields of components in aged and diluted sidestream smoke are available from a number of studies, there are few data

in the literature describing actual yields in true ETS - the aged and diluted combination of both sidestream smoke and exhaled mainstream smoke. Yields of analytes determined in real-time were determined and are presented in Table 4. Average yields across the nine countries tested are 14 ± 2 mg RSP/cig., 59 ± 5 mg CO/cig., 41 ± 4 mg TVOC/cig., 1.3 ± 0.3 mg NO/cig., and 0.4 ± 0.1 mg NO₂/cig.

Emission rates for RSP from the six brand styles of U.S. cigarettes (14.5 mg/cig.) agree well with sales-weighted average values from a 50-brand study of U.S. cigarettes performed by Martin et al. (1997) (13.7 mg/cig.) and are somewhat lower than those determined by Leaderer and Hammond (1991) (17 mg/cig.). The American CO and NO yields of 58 and 1.5 mg/cig., respectively, also agree well with the results of Martin et al. (1997) (55 and 1.6 mg/cig.). Yields of TVOCs (by FID) and NO₂ are somewhat higher (60% and 150%) than previously reported by Martin et al. (1997).

CONCLUSIONS

The ETS generated by simultaneous smoking of six leading brand styles from each of the nine countries was generally similar. Specific ETS-RSP apportionment factors were determined for each country. Overall apportionment factors were determined to be 7.2, 42, and 38 for UVPM, FPM, and solanesol, respectively. These factors were in reasonable agreement with those previously reported in the literature. Apportionment of ETS-RSP can be readily achieved by measurement of marker compounds and application of country-specific apportionment factors. The overall apportionment factors for UVPM or FPM could reasonably be applied to determine the ETS contribution to RSP in any of the tested countries. However, considerable variation in the RSP:solanesol ratio was observed among countries. For the countries tested, application of country-specific solanesol apportionment factors would yield more accurate results than the overall factor. Per cigarette yields of RSP, CO, TVOC (by FID), NO, and NO₂ were determined. Yields were generally similar among the countries tested and averaged 14, 59, 41, 1.3, and 0.4 mg/cig. for each of those analytes, respectively.

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