EREM 73/4
Journal of Environmental Research, Engineering and Management
Vol. 73 / No. 4 / 2017
pp. 21-30
DOI 10.5755/j01.erem.73.4.19316

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Received 2017/10 | Accepted after revision 2017/11

http://dx.doi.org/10.5755/j01.erem.73.4.19316

Exposure to Total Volatile Organic Compounds from Household Spray Products

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Emission of volatile organic compounds from the use of household spray products has a negative impact on health and environment. Total volatile organic compound (TVOC) concentration levels emitted from 45 registered consumer spray products in Nigeria were measured using the MultiRAE TVOCs gas monitor. Human exposure to emitted TVOCs through inhalation, ingestion or through the dermal route was evaluated using the ConsExpo spray model. The average TVOCs emission from all the investigated samples was in the range of 1,664 and 560,994.7 μg m^{-3} with an average of 63,632.2 μg m^{-3}. Generally for all the samples considered, the average released concentrations, the inhaled doses, the dermal doses, and the average deposition rates values obtained were in the ranges of 1.83E+04 – 1.00E+06 μg m^{-3}; 1.47E+03 – 8.01E+04 μg; 3.41E+04 – 4.84E+05 μg; and 1.79E+01 – 1.01E+03 μg s^{-1}, respectively. The results provide information that could be used to significantly improve human exposure and risk assessment to emitted aerosols from spray products.

Keywords: household products, exposure, total volatile organic compounds, hazard ratio, indoor environment.

Introduction

People use household spray products in indoor environments where they spend a higher percentage of their time. The use of household spray products results in introduction and elevation of volatile organic

compound levels in the indoor environment (Adeniran, Sonibare, & Jimoda, 2015). VOCs have been widely studied and they have been reported to contribute to human health deterioration and depletion of ozone in the atmosphere (Dinh et al., 2015). A number of spray products contain VOCs in the form of fragrances, propellants, and other active ingredients. Some of the undisclosed constituents of the total volatile organic compounds present in household products have been classified as toxic and hazardous under federal laws of most developed countries (Steinemann, 2009).

Human exposure to emitted VOCs can occur through inhalation, ingestion or through the dermal route. The aim of this study is to quantify the concentration levels of TVOCs emitted from consumer spray products and estimate their exposure level.

Methodology

Mass generation rates

Average mass generation rates of household spray products were determined by averaging mass of aerosol released for 3 scenarios (full filled, half-filled and while almost empty). In each case, an individual spray product was weighed using Mettle Toledo weighing balance to obtain the initial weight, sprayed for 10 seconds and reweighed to obtain the final weight. The mass generated for each case was determined using equation 1

$$M_g = \frac{M_i - M_f}{t} \quad (1)$$

where M_g is the mass generation rate, M_i (g) is the initial weight of the sample, M_f (g) is the final weight of the sample and t is the time (s).

A wide range of consumer products was sampled in order to fully capture the behaviour of aerosols being investigated. Forty-five (45) commercially available household spray products in the Nigerian market were considered for investigation. The selected samples include: 15 different brands of air fresheners, 15 different brands of insecticides, 5 different brands of hairsprays, 5 different brands of perfumes, 2 different brands of shoe impregnation sprays and 3 different brands of surface polish (Table 1). Production dates on the selected samples suggested that they had been manufactured not more than 3 months before they were used for the study. Since specific information on

Table 1

Characteristics of household products used in the study

S/N	Product	Product type	Use category	Filling level (ml)	Major listed ingredients
1	2	3	4	5	6
1	INST A	Insecticide	Air space	300	Butane, propane, active ingredients
2	INST B	Insecticide	Air space	300	Butane, propane, active ingredients
3	INST C	Insecticide	Air space	300	Butane, propane, active ingredients
4	INST D	Insecticide	Air space	300	Butane, propane, active ingredients
5	INST E	Insecticide	Air space	400	Butane, propane, active ingredients
6	INST F	Insecticide	Air space	330	Isobutane, propane, active ingredients
7	INST G	Insecticide	Air space	300	Butane, propane, active ingredients
8	INST H	Insecticide	Air space	300	Isobutane, propane, active ingredients
9	INST I	Insecticide	Air space	300	Butane, propane, active ingredients
10	INST J	Insecticide	Air space	400	Butane, propane, active ingredients
11	INST K	Insecticide	Air space	400	Butane, propane, active ingredients
12	INST L	Insecticide	Air space	300	Butane, propane, active ingredients
13	INST M	Insecticide	Air space	305	Butane, propane, active ingredients

1	2	3	4	5	6
14	INST N	Insecticide	Air space	300	Butane, propane, active ingredients
15	INST O	Insecticide	Air space	600	Butane, propane, water, active ingredients
16	AFN A	Air freshener	Air space	300	Butane, propane, ethane, 1,1-difluoro-, fragrances
17	AFN B	Air freshener	Air space	300	Butane, propane, active ingredients
18	AFN C	Air freshener	Air space	300	Butane, propane, active ingredients
19	AFN D	Air freshener	Air space	300	Butane, propane, active ingredients
20	AFN E	Air freshener	Air space	300	Butane, propane, active ingredients
21	AFN F	Air freshener	Air space	400	Butane, dimethyl ether, fragrances
22	AFN G	Air freshener	Air space	400	Butane, propane, active ingredients
23	AFN H	Air freshener	Air space	500	Butane, isobutane, propane, fragrances
24	AFN I	Air freshener	Air space	300	Butane, propane, disodium tetraborate decahydrate, fragrances
25	AFN J	Air freshener	Air space	300	Butane, propane, active ingredients
26	AFN K	Air freshener	Air space	400	Butane, propane, active ingredients
27	AFN L	Air freshener	Air space	500	Butane, propane, active ingredients
28	AFN M	Air freshener	Air space	320	1,1-difluoroethane, propane, terpenes and terpenoids, sweet orange-oil
29	AFN N	Air freshener	Air space	300	Butane, propane, fragrances
30	AFN O	Air freshener	Air space	330	Butane, propane, fragrances
31	PEF A	Perfume/body spray	Towards person	75	Isobutane, propane, fragrances
32	PEF B	Perfume/body spray	Towards person	75	Isobutane, propane, fragrances
33	PEF C	Perfume/body spray	Towards person	200	Isobutane, propane, fragrances
34	PEF D	Perfume/body spray	Towards person	110	Isobutane, propane, fragrances
35	PEF E	Perfume/body spray	Towards person	75	Isobutane, propane, fragrances
36	HSP A	Hair spray	Towards person	450	Propylene glycol, isopropyl alcohol
37	HSP B	Hair spray	Towards person	450	1,1-difluoroethane, ethanol, methoxyethene, proprietary fragrance
38	HSP C	Hair spray	Towards person	450	Dimethyl ether, methacrylate co-polymer, butylene glycol, sodium benzoate
39	HSP D	Hair spray	Towards person	625	Dimethyl ether, sd alcohol 40-b (ethanol)
40	HSP E	Hair spray	Towards person	450	Ethyl alcohol, propane, butane
41	SPL A	Surface polish	Surface	300	Acetone, n-butane, propane
42	SPL B	Surface polish	Surface	250	White mineral oil, petroleum, distillates (petroleum), hydrotreated light
43	SPL C	Surface polish	Surface	300	Ethanol, 2-butoxy, isopropyl alcohol
44	SSP A	Shoe impregnation spray	Surface	220	1,1,1,2-tetrafluoroethane R134A, dimethyl ether
45	SSP B	Shoe impregnation spray	Surface	200	Hydrotreated light distillate, mineral spirits, aromatic petroleum distillates

the products' comprehensive description and ingredients is rare, the criterion for product selection was the packaging label, which indicates they are aerosolized products. The ingredients list on the labels of most samples selected indicates that they contain the active ingredients and the propellants that account for between 60% and 90% of their total volume. The active ingredients in fragranced products were simply termed 'fragrance'. The most common propellants listed and declared for the selected samples include butane, isobutane and propane.

Sampling procedure

TVOCs measurements were done using the Multi-RAE Gas Monitor (Model PGM50-5P), manufactured by RAE Systems Inc., USA. The monitor is a 9.3 cm × 4.9 cm × 2.2 cm measuring instrument weighing about 0.1 kg with an instantaneous direct readout display through which VOCs concentrations can be continuously monitored in ppm (parts per million). It has a detection range of between 0–200 ppm with 0.1 ppm resolution. It has a facility for short term exposure limit (STEL) from which TVOC concentration for the last 15 minutes can be determined; the time weighted average (TWA) from which the accumulated reading of the TVOC concentration was turned on divided by 8 hours; and the peak reading, which will represent the highest concentration. Each of the aerosol spray products was sprayed for 10 seconds inside the experimental room. Average microclimatic parameters in the experimental room were 55%, 32.0 °C and 9 h^{-1} for relative humidity, temperature and air exchange rate, respectively. Sprays produced by the aerosol products were characterized by the methods proposed by Fédération Européenne Des Aérosols (FEA, 2009). The TVOC sampler was placed at 40 cm above the spraying nozzle or the receptor. The position depends on the protocol that was employed for different spray product categories. Measurements were taken 15 times for each product and averaged (5 each for fully filled, half-filled and almost empty).

Exposure assessment

The hazard ratio (HR) and ConsExpo spray model approaches were used to assess the risks associated with the use of spray products. The concentration levels obtained were compared with some of the existing air quality standards for TVOCs. Hazard ratio was determined as the ratio of the measured to that of the statutory limits such as the limits of the Nigerian Federal Ministry of Environment, the World Health Organization and literature established TVOCs guidelines (FEPA, 1991; Mølhave & Nielsen, 1992; WHO, 2010; Zabiegała, 2006).

The ConsExpo spray model used was developed by the National Institute for Public Health and the Environment (RIVM), Netherlands. It was used to evaluate human exposure to aerosol released from the spraying activities in the experimental room. ConsExpo has been used internationally to assess the exposure of consumers to substances in consumer products (Bremmer, Prud'Homme de Lodder, & Van Engelen, 2006; Rothe et al., 2011). The main input parameters for the model are the released droplet spectrum, the release rate, the concentration of the pollutant, the spatial and temporal pattern of the release process (surface spraying against floor, ceiling, wall, room spraying), the vapour pressure of the liquid, the size of the room and the ventilation rate. The main output parameters are the concentration calculated values, the concentration versus time diagram and the time-integrated inhaled and deposited dose of the pollutants considered. The model is based on assumptions that the sprayed product is composed of a non-volatile substance dissolved in a solvent with known volatility, and the determination of the inhaled dose is based on the International Conventions on Health-Related Particle Sampling defined for example in CEN481.

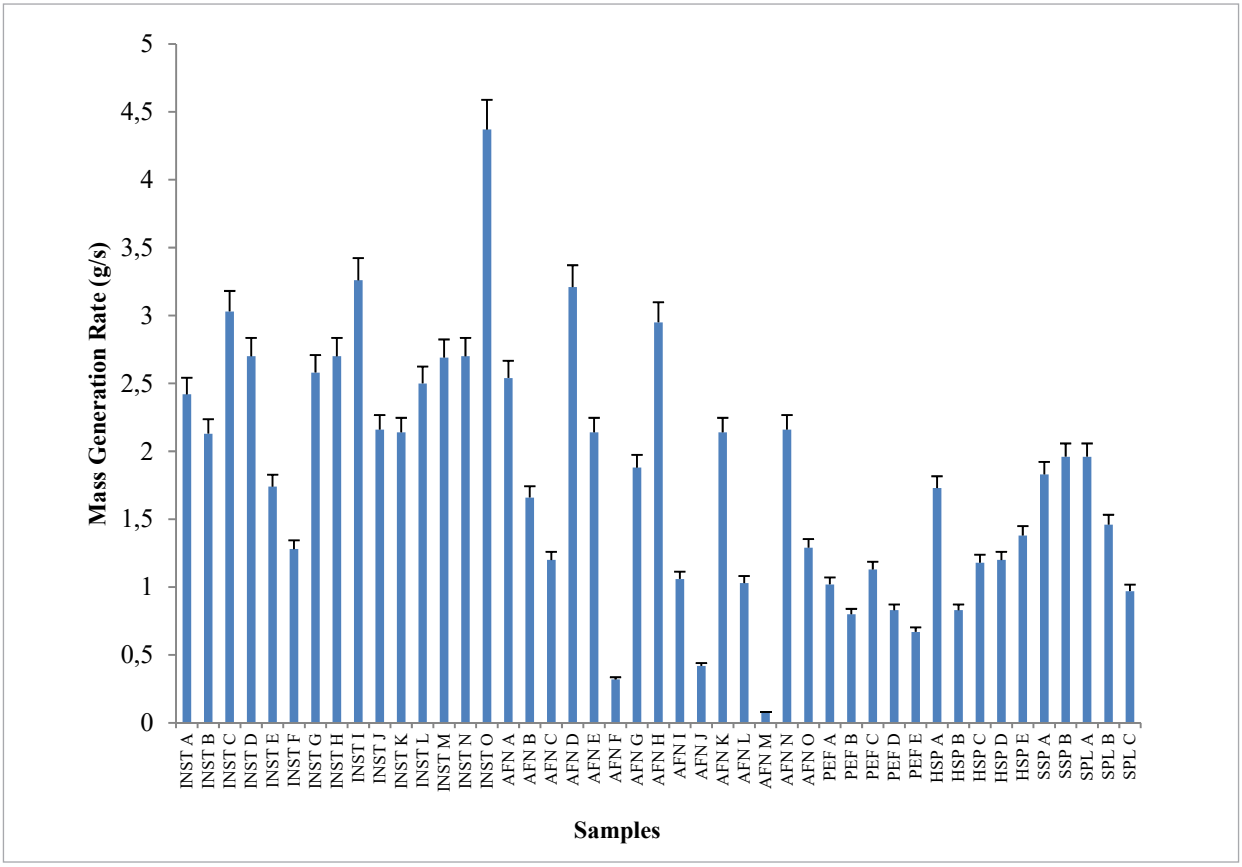
Results and discussion

Mass generation rates

The mass generation rates of the 45 different household spraying product samples selected for investigation were determined and results were presented in Fig. 1. For the insecticide samples considered, INST A has the highest mass generation rate of 4.37 g/s, while INST F has the least mass generation rate of 1.28 g/s. The mean aerosol mass generation rate for the insecticides is 2.56 g/s with the standard deviation

Fig. 1

Mass generation rates of aerosols from spray products



of 0.7 g/s. The mass generation rate for air freshener samples ranged from 0.08 to 3.21 g/s. The mean mass generation rate was 1.61 g/s with a standard deviation of 0.94 g/s. AFN D has the highest mass generation rate, while AFN M has the least mass generation rate.

For the perfume samples considered, the mass generation rate ranged from 0.67 g/s to 1.13 g/s with the mean value of 0.89 g/s and the standard deviation of 0.18. The highest mass generation rate was measured in PEF C and the lowest in PEF E.

Concentration levels of TVOCs in consumer spray products

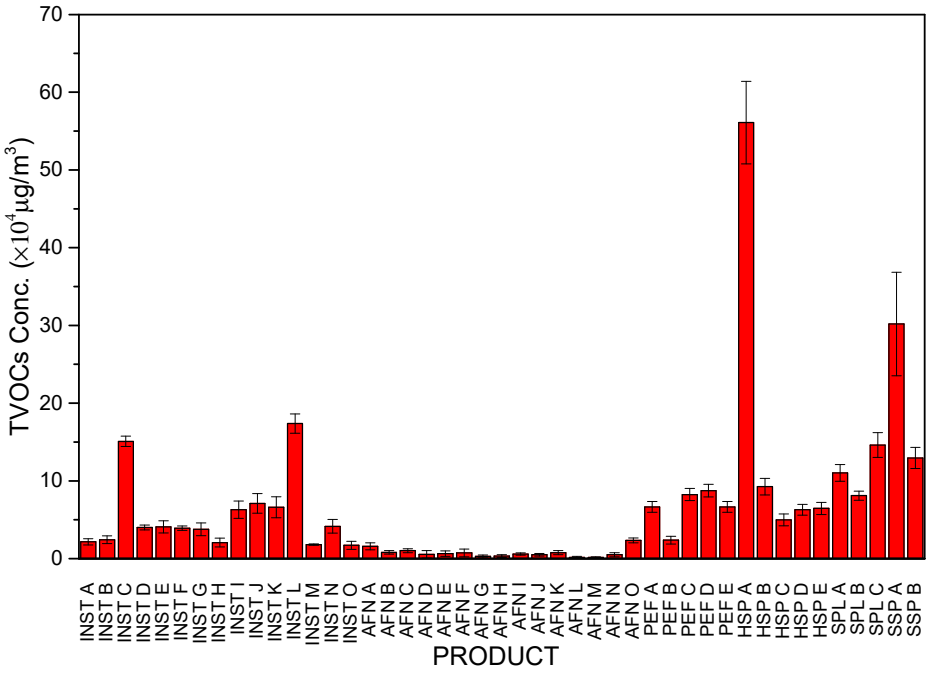
The concentrations of the resulting TVOCs were in the range of 173,765.7 and 17,115.09 μg m^{-3} for insecticides. INST L emitted the highest concentration of TVOCs (Fig. 2), and INST O was the least emitter of TVOCs in all the insecticides investigated. The average concentration of TVOCs emission from insecticides was 55,085.24± 47,007.51 μg m^{-3}.

Air freshener samples had a TVOC concentration range of 1,663.97 and 23,533.25 μg m^{-3}. AFN O emitted the highest TVOC concentration and AFN L emitted the least. The mean emitted concentration is 7,337.30 μg m^{-3} with the standard deviation of 5750.04 μg m^{-3}.

For perfume, hairspray, surface polish and shoe impregnated spray samples, TVOC emission ranged 2,377.10–87,477.14 μg m^{-3}; 49,919.02–560,996.98 μg m^{-3}; 18,9312.27–441,149.70 μg m^{-3} and 118,450.43–161,175.95 μg m^{-3} with respective average concentrations of 61,091.37 μg m^{-3}, 166,254.10 μg m^{-3}, 281,263.19 and 139,813.19 μg m^{-3}. In all the hairspray samples considered in this study, HSP A emitted the highest concentration of TVOCs with a concentration of 560,996.98 μg m^{-3}.

Fig. 2

Concentration levels of TVOCs emitted from selected spray products



Generally, the average TVOC emission from all the investigated samples was in the range of 1,664 µg m^{-3} and 71,039.59 µg m^{-3} with an average of 50,994.7 µg m^{-3}. Mølhave and Nielsen (1992) and Zabiegała (2006) considered TVOC concentration below 200–600 µg m^{-3} to be within the comfort range, 200–3,000 µg m^{-3} as the multifactoral exposure range which is considered to be a health hazard; 3,000–25,000 µg m^{-3} being the discomfort range that could bring strong discomfort to inhabitants of indoor environment and a concentration greater than 25,000 µg m^{-3} as a toxic range.

The measured concentrations of TVOCs from the samples used are in either the discomfort range or the toxic range. This indicates that occupants are often exposed to hazardous and toxic levels of TVOCs whenever the samples are used in the indoor environment. Also, recent investigations have suggested that TVOC concentrations in consumer spray products fall short of the general regulatory requirements as they were found to be far above the recommended limits (Steinemann, 2009).

Exposure assessment

Total volatile organic compounds hazard ratios

TVOC measured and extrapolated readings were compared with the FEPA limit of 1.9 ppm (FEPA, 1991) and the WHO limit of 6.3 ppm (WHO, 2010) for 24 hours measurement to establish the hazard ratio for daily values. Also, TVOC instant/initial values were compared with the limits set by Mølhave and Nielsen (1992) to establish if the instantaneous concentrations would have deleterious challenges for human health.

The HR values for TVOCs for all the samples investigated are summarized in Fig. 3. For insecticide samples, FEPA and WHO 24-hour obtained HR ranges were 0.29–2.77 and 0.12–1.11, respectively, while the Mølhave and Nielsen (1992) HR value for instantaneous release was in the range of 0.68–6.95. The air freshener TVOC HR was in the range of 0.05–0.89, and 0.02–0.36 (0.09±0.09) for the daily FEPA and WHO HR values, respectively.

For perfumes, FEPA and WHO 24-hour HR values were in the range of 0.11–0.46, and 0.01–0.12, while the Molhave and Nielsen obtained HR values were 0.10–3.50. The computed values for hairsprays FEPA and WHO 24- hour HR values were 0.70–6.80 and 0.14–0.32, respectively. Molhave and Nielsen HR value for hairsprays was 2.00–22.44.

Surface sprays and shoe impregnation sprays have the respective Molhave and Nielsen HR levels of the range 4.74–6.45 and 7.57–17.65, respectively. Surface sprays obtained FEPA and WHO 24-hour HR levels ranges were 0.17–0.48 and 0.05–0.13, respectively, while their respective levels for shoe impregnation sprays were in the range of 0.5–1.02 and 0.14–0.28.

The FEPA (1991) limit was breached in 53.33% of the 15 insecticide samples considered. Values close to unity were obtained for 26.7% of the insecticides samples. When the initial concentrations were compared to Molhave and Nielsen's (1992) standards, higher HR values of the range 0.68 and 6.95 were obtained. Values for 10 insecticides exceeded this limit as 3 other HR values was close to unity.

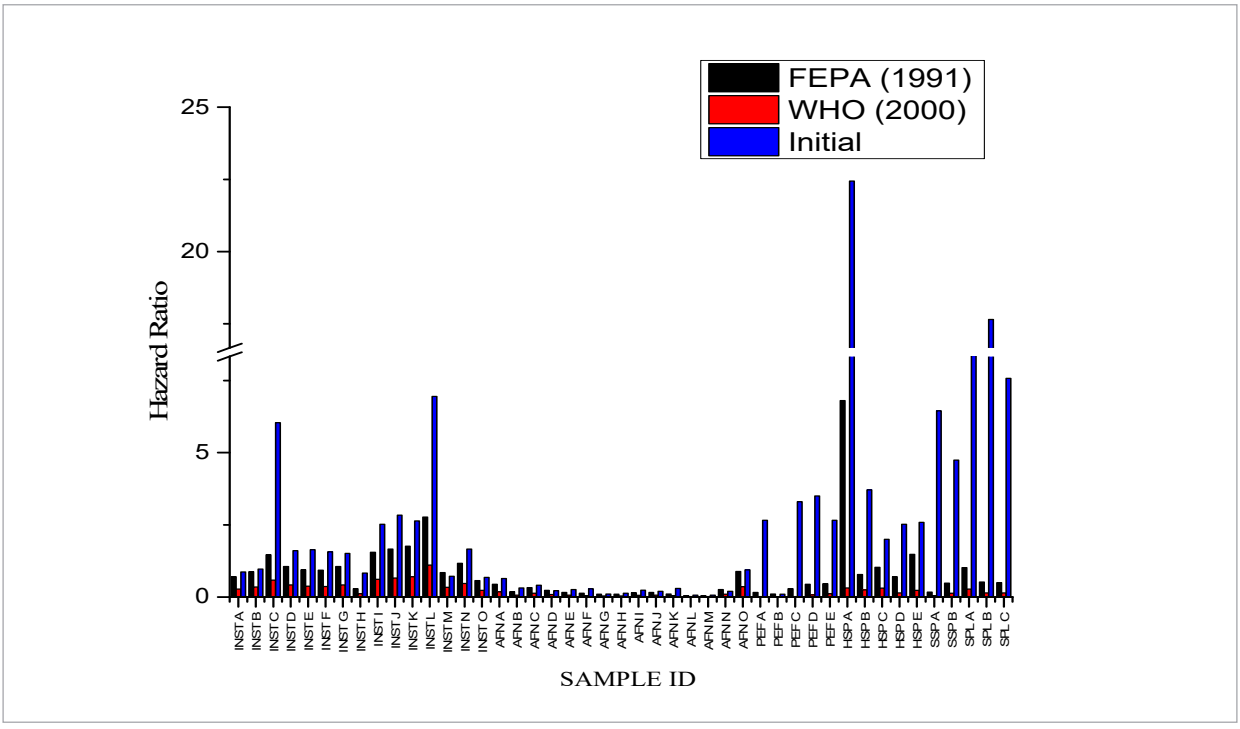
For air fresheners, none of the statutory set limits were breached as HR values for FEPA, WHO and Molhave and Nielsen (1992) TVOC set limits were below unity. This might be because the propellants used in insecticide manufacture/blending contain more TVOCs than the air freshener samples.

For perfumes, hairsprays, surface polish and shoe impregnation sprays, the initial concentration Molhave and Nielson recommended limits were grossly breached by all the samples except one which is suspected to contain some element of water as part of its composition. One hairspray and one surface polish samples gave higher HR levels of 22.44 and 17.65, respectively. This implies that a very high concentration deleterious to humans may be inhaled during use. Besides, 40% of hairspray samples and 33.3% of surface spray samples breached the FEPA 24-hour limit. Also, 20% of hairspray samples, 33.3% of shoe impregnation spray samples, and 100% of surface spray samples breached the WHO limit for 24 hours.

The high instantaneous HR levels obtained in this

Fig. 3

Hazard ratios of TVOCs from spray products



study from the use of consumer spray products could lead to an array of health-related ailments as TVOCs found in household products have been found to be responsible for irritation of the lungs, nausea, headaches, pulmonary diseases, and death (Lakey et al., 2017). Susceptibility to lung cancer and other noncancerous ailments is commonly associated with people frequently exposed to TVOCs in the indoor environment (Norbäck, Hashim, Hashim, & Ali, 2017).

While environmental and health implications of TVOCs cannot be overemphasized, TVOCs from spray consumer products could contribute to the greenhouse effect, acid rains and stratospheric ozone depletion, which are regarded as environmental problems (Erickson III, Sulzberger, Zepp, & Austin, 2015). Those emitted from household consumer products could also serve as precursors for the formation of secondary organic aerosols (SOA), which are presumed to be more harmful than primary aerosols (Erickson III et al., 2015). They could readily react with ambient ozone, a hydroxyl group and nitrates to give complex compounds and particulate matter (Erickson III et al., 2015; Romonosky et al., 2017).

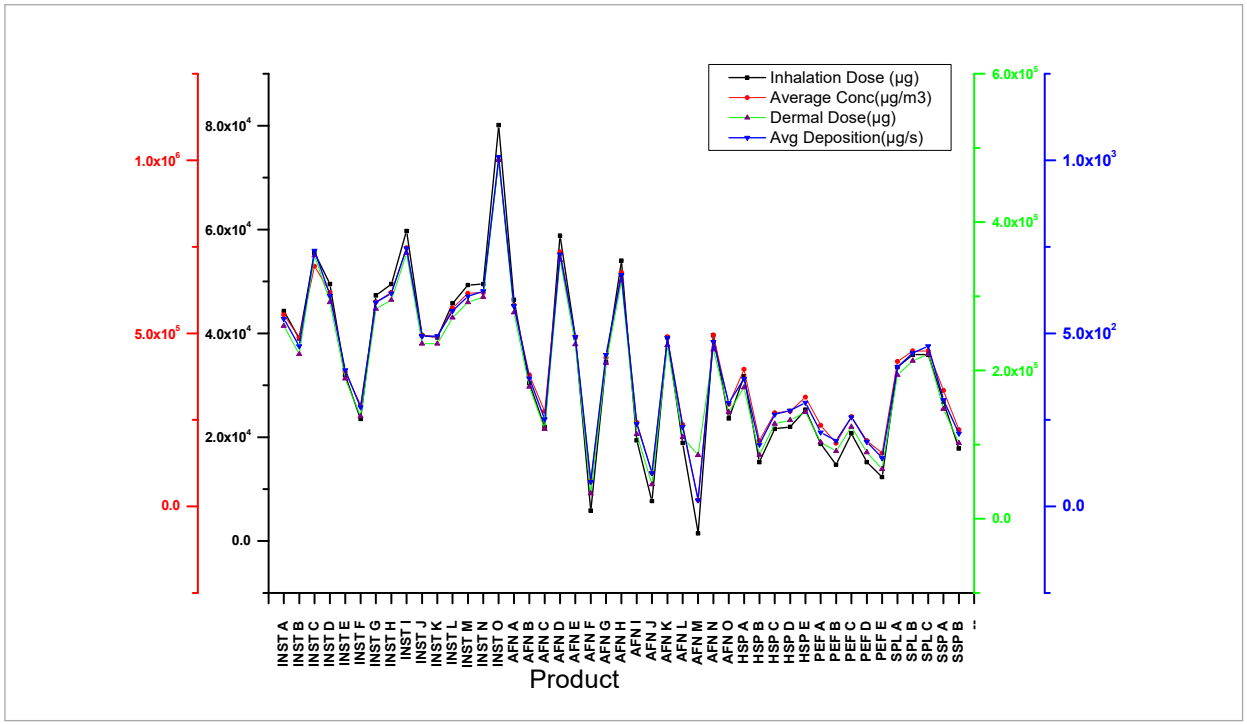
ConsExpo spray model results

For insecticides (Fig. 4), the average concentration was in the range of 2.93E+05 and 1.00E+6 μg m^{-3}. The inhaled dose from insecticides spraying ranged from 2.35E+04 to 8.01E+04 μg, while the dermal dose was between 1.38E+04 and 4.84E+04 μg with the respective average deposition between 2.87E+02 and 1.01E+03 μg s^{-1}.

Average aerosol concentration levels for air freshener spraying (Fig. 4) ranged between 1.83E+04 and 7.35E+05 μg m^{-3}. The simulated amount inhaled was between 1.47E+03 and 5.88E+04 μg, while the dermal dose ranged between 3.41E+04 and 3.49E+05 μg. The average deposition ranged between 1.79E+01 and 7.28E+01 μg s^{-1}. The average released aerosol concentration for hairspray (Fig. 4) ranged between 1.90E+05 and 3.96E+05 μg m^{-3}. The averaged inhaled and dermal dose ranges were between 1.52E+04 and 3.17E+04 μg, and between 8.60E+04 and 1.77E+05 μg, respectively. The average deposition range was between 1.79E+02 and 3.69E+02 μg s^{-1}.

Fig. 4

ConsExpo model exposure assessment results



For perfumes, the average released concentration ranges (Fig. 4) were between 1.54E+05 and 2.59E+05 µg m^{-3}, while the inhaled and dermal dose ranges were between 1.23E+04 and 2.07E+04 µg, and between 6.70E+04 and 1.24E+05 µg, respectively. The average deposition rates varied between 1.40E+02 and 2.58E+02 µg s^{-1}.

For surface polish and shoe impregnation sprays, the average released concentrations (Fig. 4) were in the range of 2.22E+05 and 4.49E+05 µg m^{-3}, while the inhaled and dermal doses were between 1.78E+04 and 3.59E+04 µg, and between 1.02E+05 and 2.22E+05 µg, respectively. The average deposition rates were between 2.11E+02 and 4.63E+02 µg s^{-1}.

Generally for all the samples considered, the average released concentrations, the inhaled doses, the dermal doses, and the average deposition rates values obtained were in the ranges of 1.83E+04 - 1.00E+06 µg m^{-3}; 1.47E+03 - 8.01E+04 µg; 3.41E+04 - 4.84E+05 µg ; and 1.79E+01 - 1.01E+03 µg s^{-1}, respectively.

With reference to the ConsExpo modeling results, the average inhalation exposure from insecticides was about 1.57 folds, 2 folds, 2.83 folds, and 1.54 folds of the levels obtained from air fresheners, hairsprays, perfumes, surface and shoe impregnation sprays, respectively. Similarly, the average dermal dose from air fresheners, hairsprays, perfumes, surface and shoe impregnation sprays respectively were about 63.86%, 47.60%, 33.83% and 62.64% of the levels obtained for insecticide application. Besides, the average deposition for air fresheners, hairsprays, perfumes, surface and shoe impregnation sprays respectively were about 62.10%, 47.56%, 33.82% and 62.50% of the average rate obtained for insecticides.

Individuals might receive higher inhalation and dermal doses from insecticides spraying when compared with other household product samples used in this study. The results provide information that could be used to significantly improve human exposure and risk assessment to emitted aerosols from spray products. These results will be sufficient in calculating exposures for long time exposures but may yield an underestimation of concentration levels for short-term exposure duration (Bremmer et al., 2006).

Conclusion

This study investigated TVOC emission from 45 registered household spray products. Hazard ratio and ConsExpo spray model approaches were used to assess human exposure to the emitted pollutants. The results showed that high concentration levels of TVOCs were emitted from the use of spray products in the indoor environment as the average TVOC concentration from the selected samples ranged between 1,664 µg m^{-3} and 71,039.59 µg m^{-3}. The ConsExpo spray model exposure assessment findings indicated that individuals receive extremely high inhalation and dermal doses from the use of spray products. Hazard ratios for the instantaneous releases of the products were the highest and pollutant concentration levels decrease with time. The HR values obtained for the instantaneous release of aerosols from the samples reached over 17 folds of the reference limit. The study observed that individuals would receive a higher dose from the use of insecticide samples. The results obtained provided relevant information that could be used to significantly improve human exposure and risk assessment to emitted aerosols from spray products.

Acknowledgements

This study was supported by Prof. Ayo Francis Ogunye Trust Fund, University of Lagos (PAFOTFUL), under the Prof. Ayo Ogunye Annual Research Fund awarded to JAA. The authors wish to acknowledge the support of the management of the University of Ilorin, Ilorin, Nigeria for providing a conducive environment for the study. Many thanks to the National Institute for Public Health and the Environment (RIVM), Netherlands, for providing the ConExpo model used in the study.

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Purškiamos buitinės chemijos poveikis aplinkai įvertinant visus lakiuosius organinius junginius

Gauta:
2017 m. spalis
Priimta spaudai:
2017 m. lapkritis

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Lakieji organiniai junginiai iš buitinės chemijos priemonių neigiamai veikia sveikatą ir aplinką. Tiriamajame darbe bendras lakiųjų organinių junginių (TVOC) koncentracijos lygis nustatytas iš 45 registruotų vartotojų purškiamų buitinės chemijos produktų Nigerijoje. Buvo matuojamas naudojant MultiRAE TVOCs emisijų stebėjimo sistemą. Žmogaus ekspozicija TVOCs per įkvėpimą, nurijimą ar per odą buvo įvertinta naudojant ConsExpo purškimo modelis. Vidutinė visų ištirtų mėginių TVOC emisija buvo 1,664 ir 560,994,7 μg m^{-3}, vidutiniškai 63,632,2 μg m^{-3}. Visų tirtų mėginių vidutinė išleista koncentracija, įkvepiamos dozės, odos dozės ir vidutinis nusėdimo greitis buvo 1,83E + 04 - 1,00E + 06 μg m^{-3}; 1.47E + 03 - 8.01E + 04 μg; 3.41E + 04 - 4.84E + 05 μg; ir 1.79E + 01 - 1.01E + 03 μg s^{-1}, atitinkamai. Rezultatuose pateikiama informacija gali būti naudojama siekiant pagerinti poveikio aplinkai ir žmonių sveikatos vertinimą.

Raktiniai žodžiai: buitinė chemija, poveikis aplinkai, lakieji organiniai junginiai, rizikos santykis, patalpų aplinka.