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Measurements and analysis of non-methane VOC (NMVOC) emissions from major domestic aerosol sprays at "source"

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ABSTRACT

Non-Methane Volatile Organic Compounds (NMVOCs) from domestic aerosol sprays are emerging pollutants and have substantial negative effects on human health and the environment. This study, for the first time, carried out quantification of the NMVOC emissions from off-the-shelf domestic aerosol sprays, at "source" in the UK. These aerosol sprays contain harmful organic compounds as propellants and products. The results showed that the cosmetic category (i.e. body sprays) have higher concentrations of NMVOCs with 93.7 wt% per can compared to households (i.e. air fresheners) with 62 wt%. Also, water-based products showed less NMVOCs in all analyses compared to solvent-based formulations. Direct replacement of Liquefied Petroleum Gas (LPG) propellants from conventional products with 'clean air' (i.e. nitrogen) showed the potential emission reduction of 50%. Hair spray products, however, have the highest ozone forming potential with about 105.1 g of Ozone per litre of the product compared to other domestic aerosol sprays. The level of global warming contribution of the selected aerosol sprays in the UK was measured to be 129.8 ktCO_2e in 2018 and globally, this can be projected to be 3154.6 ktCO_2e in 2020. Furthermore, NMVOC emissions contribution from the domestic aerosol sprays in the UK was measured as 61.2 kt in 2018 based on annual consumption of 520 million cans. Globally this can equate to 1437.6 kt based on the projected usage of 17.5 billion cans. Therefore, it is vital to expedite replacing LPG propellant with nitrogen in a drive for a 'near-zero' emission in aerosol industry. The results presented in this study can also be used to steer policy makers to the potentially brewing danger from an otherwise passive emission source.

1. Introduction

Air pollution can affect every organ of the human body from head to toe (Schraufnagel et al., 2019a, 2019b). This statement was also re-enforced by the Doctors from the Royal College of Physicians (RCP) and the Royal College of Paediatrics and Child Health (RCPCH) (Royal College of Paediatrics and Child Health, 2020, Smyth and Blakely, 2019) and that "domestic" aerosol sprays (i.e. body spray or air freshener) are the major sources of indoor air pollution. This is mainly due to the high levels of organic compounds such as blends of Liquefied Petroleum Gas (LPG, e.g. butane or propane), often used as the primary propellant and alcohol compounds which are the components of the product contained in each pressurised aerosol can. According to the report published by the Aerosol Dispensers Directive (Risk and Policy Analysts, 2014), about 80% of all aerosol dispensers employ LPG as the propellant. The distribution of the constituents of typical aerosol spray cans is shown in Fig. 1.

The use of LPG was first introduced in 1987, upon the Montreal protocol (Nourian et al., 2015, United Nations Environment Programme, 1987) which banned the use of harmful Chlorofluorocarbon (CFC) gases within the pressurised aerosol products. These aerosol sprays undoubtedly contain Volatile Organic Compounds (VOCs) which are organic chemicals that easily vaporise at room temperature and contribute significantly to indoor air pollution. They can, therefore, have a direct effect on the human health and thereby, reduce life expectancy resulting from respiratory and cardiovascular diseases and even impair lung functions (Dales et al., 2008, Pharmacy Magazine, 2019). Air pollution is known to exacerbate the underlying conditions and therefore, the reduction and management of the exposure to air pollution during the current COVID-19 induced lockdown, by those responsible for the homes (citizens, building managers, and city leaders) is urged (Climate Leadership Group, 2020). New guidelines to tackle indoor air pollution are recommended by C40 Cities; which is a group of 96 cities around the world which take bold climate action in order to preserve and improve the environment for a more sustainable future and are committed to delivering the Paris Agreement at local levels. The guidelines are not

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Nomenclature

AFN	Air Freshener with Nitrogen Propellant
APF	Antiperspirant (Female) With LPG Propellant
APM	Antiperspirant (Male) With LPG Propellant
BSF	Body Spray (Female) With LPG Propellant
BSM	Body Spray (Male) With LPG Propellant
CDE or CO_2e	Carbon Dioxide Equivalency
CFC	Chlorofluorocarbons
C_j	VOC Concentration of each Species
FTSE	The Financial Times Stock Exchange (or Footsie)
GCMS	Gas Chromatograph and Mass Spectrometer
GWP	Global Warming Potential
HC	Hydrocarbon
HSF	Hair Spray (Female) With LPG Propellant
IAQ	Indoor Air Quality
IWB	Insecticide Water-Based with LPG Propellant
LPG	Liquefied Petroleum Gas
MIR	Maximum Incremental Reactivity
NMVOCs	Non-Methane VOCs
NOx	Nitrogen Oxides
OFP	Ozone Forming Potential
RCP	Royal College of Physicians
RCPCH	Royal College of Paediatrics and Child Health
SB	Solvent-Based
SBL	Air Freshener Solvent-Based with LPG Propellant
SOA	Secondary Organic Aerosols
SWB	Insecticide Solvent-Based with LPG Propellant
T_G	Total Annual NMVOC Emissions (g)
VOCs	Volatile Organic Compounds
WB	Water-Based
W_{b1}	Weight of the Collected Bulk Inside the Glass Sampling Chamber (g)
W_{b2}	Weight of The Remnant Bulk After Agitation (g)
WBL	Air Freshener Water-Based with LPG Propellant
W_C	Total Weight of The Aerosol Spray Content (g)
W_{Ce}	Total Weight of Evaporated Content during Test (g)
W_{eth}	Weight of Ethanol Contained in Bulk (g)
W_f	Final Weight of The Domestic Aerosol Can (g)
WHO	World Health Organisation
W_i	Initial Weight of the Domestic Aerosol Can (g)
α	Proportion of Total NMVOCs to Weight of the Total Content

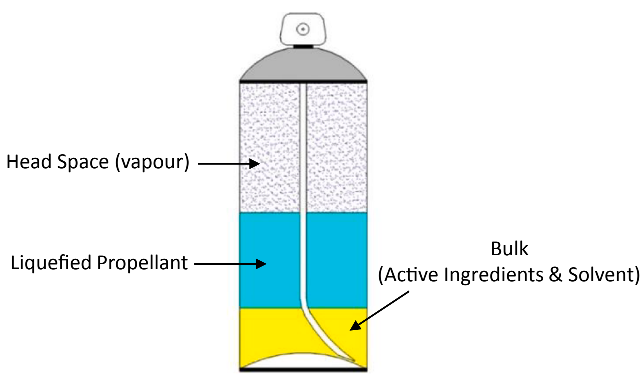

Fig. 1. Schematic illustration of the content's distribution in a typical aerosol spray can (Nasr et al., 2013, Nourian et al., 2015).

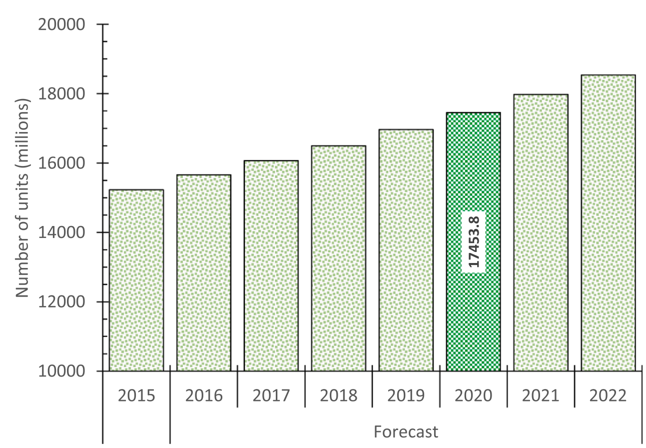

Fig. 2. Forecast of the global aerosol demand from 2015 to 2022 (Grand View Research, 2015).

intended to replace any other guidance by the government in the evolving circumstances of the pandemic (COVID-19). They are simply practical steps taken by citizens in their homes to reduce exposure to indoor air pollution now during the COVID-19 pandemic and even in the future. These include reducing burning of candles, incenses, and woodfires inside homes, and minimise the use of domestic products containing VOCs.

Around 17.5 billion aerosol spray cans are produced globally in 2020 (Grand View Research, 2015) and this is expected to grow as the world's population is increasing rapidly. These number of aerosol cans comprise of a variety of products (see Appendix A) such as personal hygiene, cosmetics, household cleaning, polish, air fresheners, surface treatments, lubrication, cooking oils, moistening and protection to people and objects while the list grows each year. The aerosol cans are filled and distributed by the large multinational conglomerates, medium and small national or local companies. Most of the sales are to the public through the large retail organisations and smaller local retailers. Fig. 2 shows the global evolution of aerosol demand within the last five years which shows the aerosol sprays market grew from about 15.3 billion aerosol production in 2015 to about 17.5 billion production in 2020. This is an increase of 2.2 billion cans in demand over a period of 5 years. The statistics also predicted that the aerosol production market will increase by another 1 billion aerosols until 2022 if the same linear trend is assumed.

It has been established that some NMVOCs contained in aerosols products are toxic (Dales et al., 2013) and extensive studies are available in the literature to this effect (Comiskey et al., 2015, Dimitroulopoulou et al., 2015a, Dimitroulopoulou et al., 2015b, McDonald et al., 2018, Rahman and Kim, 2014, Steinemann, 2016, Trantallidi et al., 2015, Wieck et al., 2018, Wolkoff and Nielsen, 2017, Zota et al., 2017). The concentration measurements of the NMVOCs from aerosols at the indoor interface (Bartzis et al., 2015, Batterman et al., 2012, Pelletier et al., 2017, Wieck et al., 2018) and their outdoor impacts in terms of secondary ozone formation (Atkinson and Arey, 2003, Carter, 1994, Dinh et al., 2015, Li et al., 2020, Niu et al., 2017, Rohr, 2013, Yan et al., 2017) have also been highlighted. However, these measurements were based on the ambient concentrations of these harmful air pollutants and their measurements were targeted to evaluate the known VOCs constituents of the domestic aerosol sprays with carcinogenic potential. For example, Dinh et al. (2015) tested different consumer and commercial products like body washes, detergents, air fresheners, insecticides. Bartzis et al.

(2015) also measured the NMVOC emissions from plug-in air fresheners, kitchen cleaning agents, and perfumes. Kwon et al. (2007) analysed household products like glues, paints, pesticides, cleaning products, sealants, and deodorisers and targeted the heavier components and their reoccurrence within the products tested. The analyses of all the aforementioned studies, showed that the majority of the NMVOCs from aerosol spray products are light hydrocarbons (C_2 to C_5), chief amongst which is LPG. This forms the bulk of the NMVOCs sources in domestic aerosol sprays and their harmful emission potential cannot be overemphasised. The NMVOCs undergo physical and chemical processes in the atmosphere leading to their transformation into secondary compounds. Lower atmospheric (tropospheric) ozone (O_3) is one of such secondary compounds which forms by the oxidation of NMVOCs in the presence of nitrogen oxides (NOx) (McDonald et al., 2018). This have long been considered as the major contributor to ozone formation (Bartzis et al., 2015). However, different species of NMVOCs have different ozone forming potential (OFP) according to Carter (1994, 2010) who developed a reactivity scale for the evaluation of relative impacts of different NMVOCs, based on the maximum incremental reactivity (MIR) of each species, on ozone formation. This helps to assess the extent to which a particular volatile compound can go to form ozone at ambient conditions and is widely used for environmental pollution analyses. Aerosol sprays contain several species of organic compounds and thus, an evaluation of their combined OFP at “source” further reiterates the magnitude of the potential danger posed by the use of aerosol sprays which contain NMVOCs on the environment.

Therefore, based on the previous experimental measurements of NMVOCs emissions from aerosol products at ambient conditions, to the authors’ knowledge, there are no available reliable and comprehensive NMVOCs data which separate emissions into the propellants and those from the product formulation at "source”. For this study, the UK will be used as a case study and a global projection will be made from the results obtained.

Therefore, the objectives of this study are to:

- quantify the level of NMVOCs per sample contained in different categories of aerosol sprays for both solvent and water-based products,
- provide an estimated level of NMVOCs generated by the domestic aerosol sprays,
- compare NMVOCs emitted from the domestic aerosol spray products with the road transport,
- present the potential environmental impact of NMVOCs from the domestic aerosol sprays with LPG propellant to ozone formation and global warming and,
- provide a potential approach to reduce the level of NMVOCs emission from these aerosol sprays.

This work will provide comprehensive data on the quantity of NMVOCs contained in everyday domestic products at “source” in the UK and will present the magnitude of the emissions from these products and also highlight the potential danger posed by their use.

2. Materials and method

2.1. Selection of domestic aerosol samples

Domestic aerosol sprays make up an average of about 70% of the total global aerosol production every year (European Aerosol Federation, 2019, Grand View Research, 2015) and the samples are shown in Table 1, are the major off-the-shelf products from different leading brands (Nourian et al., 2015, Nourian et al., 2014, Nourian et al., 2016). These selected products were further grouped into their targeted applications and according to their bulk compositions. For body sprays and antiperspirants, analysis was made in the male and female targeted subgroups. Air fresheners and insecticides also differ in their bulk compositions i.e. either solvent-based (SB) or water-based (WB). Furthermore, for comparison and evaluation of potential NMVOC reduction, two more air fresheners with nitrogen propellant, which are globally available, were tested. It should be noted that each product was tested three times for repeatability and reliability of quantification/measurement of the total NMVOCs.

2.2. Experimental setup

Fig. 3 shows the experimental setup which was used during this investigation to measure the NMVOC from common domestic aerosol sprays. It is comprised of an electronic mass balance (Metler AJ100) with high accuracy of ±0.001, a mass flowmeter (OMEGA FMA-1600A) with an accuracy of 0.01% to measure the flow rate of the effluent from the sampling chamber, a graduate volumetric glass measuring cylinder with an accuracy of ±0.1 ml, and an Agilent Gas Chromatograph (6890N) and Mass Spectrometer (5873N) setup with Enhanced ChemStation for pre and post processing. The detailed description of these equipment can be found in Agilent Technologies manuals (Agilent Technologies, 2000, 2009). Agilent MassHunter qualitative and quantitative software was used for post analysis after the Gas Chromatograph (GC) separation and Mass Spectrometry (MS) identification of the targeted components of the aerosol sprays.

2.3. Experimental methodology and analysis

The sequence of the experimental methodology that was created and used during this investigation has been divided into THREE different parts:

- Part-I: calibration and method development of GCMS;
- Part-II: analysis of the chemical components of the aerosol propellant;
- Part-III: quantifying the level of NMVOCs within the aerosol propellant.

Table 1
Selection of major domestic aerosol sprays with LPG propellant.

Aerosol Product		Bulk Composition	Propellant	Targeted People	Size (ml)	Code
Households	Air Freshener	WB	LPG	–	300	WBL
		SB	LPG	–	250	SBL
		WB	Nitrogen	–	240	AFN-1
		SB	Nitrogen	–	300	AFN-2
	Insecticide	WB	LPG	–	300	IWB
		SB	LPG	–	250	ISB
Cosmetics	Body Spray	SB	LPG	Male	150	BSM
		SB	LPG	Female	75	BSF
	Antiperspirant	SB	LPG	Male	250	APM
		SB	LPG	Female	250	APF
	Hair Spray	SB	LPG	Female	275	HSF

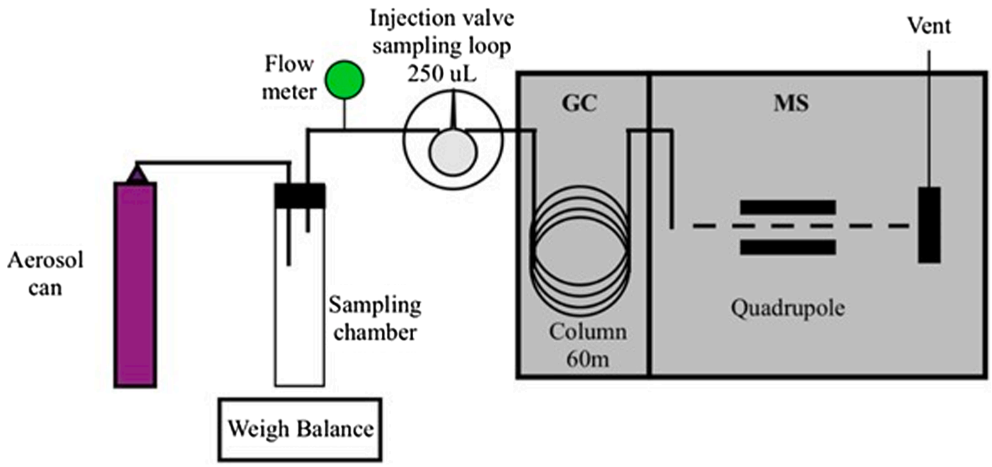

Fig. 3. Schematic diagram of NMVOCs measurement setup.

2.3.1. Part-I: calibration and method development of GCMS

An external calibration gas bottle with high purity and relative analytical uncertainty of ±2% were used from Agilent Technologies UK. The bottle comprised of different component mixture as stated in Table 2. In order to calibrate the quantification of ethanol within the aerosol spray, high purity ethanol was also injected directly with a 5 μL syringe into the system.

The developed and established method from this was then used for the analysis of the aerosol sprays in the subsequent stage. Quantitative and qualitative analyses were carried out using the Agilent GCMS setup with a 250 μL sampling loop. The carrier gas for this set up was pure helium gas. A capillary column (Agilent DB-1) with specification of 60 m × 320 μm × 1 μm with the maximum operating temperature of 260 °C was used to perform the analyses with the temperature program (method) shown in Fig. 4.

The split ratio was set to 950:1 and the scan parameter for the MSD was set to 20 -400 amu with the transfer line temperature held at 280 °C.

2.3.2. Part-II: analysis of the chemical components of the aerosol propellant

This stage showed the quantification of the total propellant in the targeted aerosol sprays. The procedure of measurements is broadly as follows:

- Weight of the sampled aerosol can were first measured and recorded.
- The sample was then agitated and sprayed into the sampling chamber intermittently as predefined intervals (i.e. 5–10 s) to allow the sampling loop to be saturated with the gaseous/volatile aerosol sprays while the heavier components (liquid) were collected in the glass sampling chamber.

Table 2
Composition of calibration gas bottle from Agilent Technologies, UK.

Compounds		Formula	CAS Number	Actual Concentration (mole %)
Hydrocarbons (HCs)	Hexane	C_6H_{14}	110-54-3	0.50%
	Iso-pentane	C_5H_{12}	78-78-4	1.00%
	n-pentane	C_5H_{12}	109-66-0	1.00%
	Iso-butane	C_4H_{10}	75-28-5	1.00%
	n-butane	C_4H_{10}	106-97-8	2.99%
	Propane	C_3H_8	74-98-6	6.00%
	Ethane	C_2H_6	74-84-0	9.00%
	Methane	CH_4	74-82-8	71.11%
Non-HCs	Argon (Ar)	–	7440-37-1	0.50%
	Carbon Dioxide	CO_2	124-38-9	1.00%
	Nitrogen	N_2	7727-37-9	5.90%

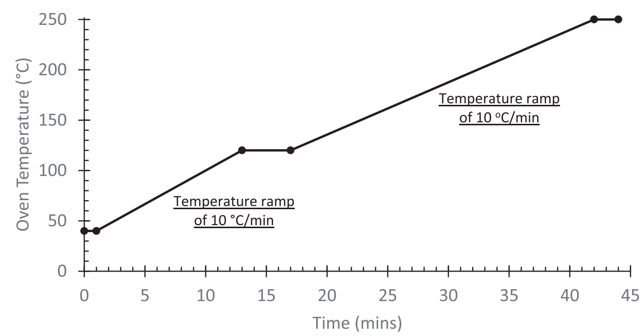

Fig. 4. Temperature program for the GC method used for analysis.

- Once the pressure within the system stabilised at ambient condition (to avoid overloading the column), the GCMS method was run to analyse the sample and results recorded.
- The above steps were repeated until the aerosol can was empty and the total mass of the empty can was measured and recorded.

In between each run, pure nitrogen was used to purge the sampling line/loop of any residual samples and to obtain a clean and contaminant-free GC column.

2.3.3. Part-III: quantifying the level of NMVOCs within the aerosol propellant

Post processing of the results obtained from the GCMS showed the amount of each vaporised components (mainly the LPG blends) together with the trace amounts of other volatile organic solvents (mainly ethanol) within those domestic aerosol sprays which are categorised as the solvent-based. The important parameters obtained from the analyses are as follows:

$$\text{Total weight of the aerosol spray content}, W_C(g) = W_i - W_f \quad (1)$$

$$\text{Total weight of evaporated content during the test}, W_{Ce}(g) = W_C - W_{b1} \quad (2)$$

where W_i is the initial weight of the sample domestic aerosol can (g), W_f is the final weight of the can (g), and W_{b1} is the weight of the collected bulk inside the glass sampling chamber (g). The above equations were used to calculate the mass of the propellant which are mainly in gaseous phases. This procedure was adopted in analysing the weight of propellant in other aerosol spray sampled in this study. After the bulk was collected and its mass measured (W_{b1}), a GC liquid sampling was performed using liquid injection method to analyse the solvent compound, mainly ethanol (W_{eth}) contained therein. It should be noted that this method was not done for water-based products because of the risk of water damage to the equipment. Therefore, the total weight of NMVOCs from liquid propellant can be obtained from:

$$NMVOC_{propellant}(g\ \ per\ \ can) = W_{Ce} - W_{eth} \quad (3)$$

Subsequently, a magnetic stirrer was introduced to agitate the bulk to expedite the vaporisation of the volatile components in a controlled environment and also expel some of the dissolved LPG propellant in the bulk. The GCMS method was also used to identify the majority of the volatile compounds from the bulk during the agitation. The agitation continued until there were no more GC spectrum peaks of the targeted (more volatile) components in the collected bulk. The weight of the remnant of the bulk after agitation was measured (W_{b2}). The NMVOC of the bulk contained in the aerosol sample was then measured using the following relation:

$$NMVOC_{bulk}(g\ \ per\ \ can) = W_{b1} - W_{b2} + W_{eth} \quad (4)$$

Therefore, the total NMVOCs per sample can be then expressed as:

$$Total\ \ NMVOC, T_C(g\ \ per\ \ can) = NMVOC_{propellant} + NMVOC_{bulk} \quad (5)$$

It should be noted that, in this study, the measurement was targeted as the bulk of the NMVOCs contained in an aerosol that is LPG and ethanol. Furthermore, for simplicity and scalability, Eq. (6) shows the proportion of total NMVOCs to the weight of the total content (W_C) as a dimensionless quantity which identified in this study as ‘α’.

$$\alpha(wt.\%) = \frac{T_C(g\ per\ can)}{W_C(g)} \times 100 \quad (6)$$

2.3.4. Ozone forming potential (OFP)

Using the approach by Carter (1994), the ozone forming potential (OFB) for each individual NMVOC species was evaluated using Eq. (7):

$$OFP = C_j \times MIR_j \quad (7)$$

where C_j is the VOC concentration of the species j and MIR_j is the maximum incremental reactivity coefficient ($\frac{g\ O_3}{g\ VOC}$).

2.3.5. Global warming contribution

Additionally, the components of the LPG (ethane, butane, and propane) have different abilities to absorb energy as they possess various radiative efficiencies. Greenhouse gases, generally, warm the Earth by the adsorption of energy and trapping it within the lower atmosphere and thus reducing the rate of heat escape. Each of these gases have a specific global warming potential (GWP), which gives a measure of the amount of energy the emissions of a specific mass of gas will absorb over a period of time, often 100 years, compared to the emissions of 1 tonne of CO_2 since carbon dioxide is considered to have a very long residence time in the atmosphere (Vallero, 2019).

GWP as an emission metric can provide a good comparison between the LPG potential in domestic aerosol products and CO_2 which is defined as the measure of the heat absorbed over a given period of time due to emission of a gas. Eq. (8) can be used to calculate the carbon dioxide equivalency (CDE or CO_2e) of the LPG propellants using the GWP of each gas emission.

$$CDE_j = GWP_j \times M_j \quad (8)$$

where GWP_j is the global warming potential of the gas species j for a period of 100 years (which obtained from Forster et al. (2007) and M_j is the mass of each propellant constituent which was measured and evaluated in this study.

3. Results and discussion

3.1. Repeatability and uncertainty

Prior to the actual testing and evaluation, three cans were tested under the same conditions to evaluate the repeatability of the experimental measurement method and the uncertainty of the equipment set up. The combined uncertainty of the set-up is 1.5%. The repeatability results from the three cans tested are shown in Table 3 with a standard deviation of 0.55 g.

3.2. Quantification of NMVOC in domestic aerosol sprays with LPG propellant (at "source")

A quantification of the total NMVOCs in the aerosol sprays were measured and presented in Table 4 and it is also shown in Fig. 5.

As shown in Fig. 5, cosmetic samples presented higher content of the NMVOC compared to the household products. In the household products analyses, the solvent-based air freshener (SBL) contained the highest level of non-methane VOC (98.9 wt%) in this category compared to both water-based (IWB) and solvent-based (ISB) insecticides.

The water-based air freshener (WBL) had the lowest NMVOCs (33.8 wt%) amongst the tested products. As expected, the solvent-based insecticide had higher NMVOCs than the water-based as it stands at 74.9 wt% leaving the water-based at 40.2 wt%. The most abundant volatile organic compounds in all the test products are shown in Table 5. Majority of which are the most volatile components ranging from of C_2 to C_5 as already postulated.

It can be concluded that the water-based aerosol sprays have lower NMVOCs compared to the solvent-based in the household category. The NMVOCs in the water-based samples come largely from the propellant (LPG) with negligible amounts in the bulk of the products as shown in Fig. 6. The NMVOC emitted from the solvent-based household samples are almost twice as much as those from the water-based samples.

All the selected samples contained LPG as the main propellant and major solvent evaluated was ethanol as shown in Table 5. Furthermore, the cosmetic category contains substantially more NMVOCs, which is coming from both propellant and the solvents. The antiperspirants have more propellants, in the cosmetic category, but the body sprays have higher total NMVOCs levels as a result of having higher ethanol content compared to other samples in the same category. These are similar trends observed in previous literature studies (Dinh et al., 2015). Interestingly, ethanol appeared to be the most abundant solvent in the bulk portion of all the products tested in this study. This finding, also, shows the evolution of ethanol in the increased NMVOC emissions from the solvent and industrial emission categories. According to Lewis et al. (2020) ethanol is becoming the most important NMVOC emitted by mass (about 136 kt/annum in 2017 accounting for about 17% of the total anthropogenic NMVOC emissions – 810 kt/annum in the same year) in the UK.

Having quantified the propellant contained in each category, LPG accounts for about 50% of the NMVOCs overall. As shown in Table 5 and Fig. 6, the LPG propellant contributes significantly to the total level of non-methane VOCs contained in the majority of domestic aerosol sprays. Such a high level of emission is extremely disconcerting since these will eventually be emitted indoors in highly insulated buildings and with low infiltration rates. Clearly, this level of pollutants can certainly have a significant impact on indoor air quality as well as the development of

Table 3
Repeatability and uncertainty of experiments.

Total weight of the aerosol spray content (W_C), g				
TEST #1	TEST #2	TEST #3	Average	Standard Deviation
147.35	148.22	147.21	147.59	0.55

Table 4
NMVOC of bulk and propellant within each selected domestic aerosol sprays.

Aerosol Sprays		Code	NMVOCs (g per can)			
			Bulk	Propellant	Total	Ave.
Households	Air Freshener	WBL	0.2	87.6	87.8	119.6
		SBL	90.3	65.4	155.7	
	Insecticide	IWB	47.3	57.8	105.1	
		ISB	64.5	65.1	129.6	
Cosmetics	Body Spray	BSM	47.9	46.0	93.9	116.9
		BSF	28.6	20.0	48.6	
	Antiperspirant	APM	42.8	85.4	128.2	
		APF	39.4	93.8	133.2	
	Hair Spray	HSF	147.6	33.1	180.7	

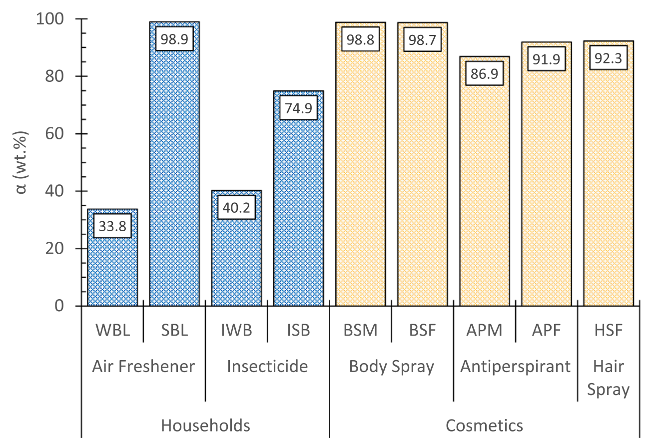

Fig. 5. Total NMVOCs (wt%) within selected aerosol sprays with LPG propellant.

serious health-related problems. This impending air pollution problem was noted and highlighted recently by the health specialists (Schraufnagel et al., 2019b, 2019a) where they discussed the extent of air pollution on health and the quality of life. To exemplify, according to the recent study (Hadei et al., 2018, Ma et al., 2010) hairspray products in their current design contain many toxic pollutants such as benzene, toluene, formaldehyde, xylene, acetaldehyde and ethylbenzene and these are evidently dangerous to human health leading to a possible cancer risk, airway irritants, bronchitis, mucosal irritation, developing asthma including the increased risk of cardiovascular problems.

3.3. Estimation of total NMVOC emissions from domestic aerosol sprays with LPG propellant

The total annual NMVOC emissions (T_G) in kiloton (kt) from domestic aerosol sprays can be evaluated by:

$$T_G(\mathrm{kt}) = \frac{T_C(\mathrm{g\ \ per\ \ can}) \times \mathrm{No.\ of\ aerosol\ sample\ (million\ units/year)}}{10^9} \quad (9)$$

Therefore, using Eq. (9), the average NMVOCs in cosmetics and households from Table 4, and considering the demand of domestic aerosol sprays in the UK in 2018 (377 million units of cosmetics and 143 million units of households), the total NMVOC emissions in the UK can be evaluated as 61.2 kt. The magnitude of T_G based on the global aerosol sprays demand (showed in Appendix B), was estimated and shown in Fig. 7. This estimation was made under the assumption that all domestic aerosol sprays with LPG propellant have similar composition as those selected and tested samples in the UK for this study. The uncertainly in the global estimations was a direct reflection of the NMVOC measurement per spray can which equates to, also, 1.5% as previously stated in Section 3.1. As demonstrated, using Eq. (9), the total NMVOC emissions from 12.2 billion domestic aerosol sprays (cosmetic and household categories) is globally estimated to be 1437.6 kt per annum. Europe contributes massively to NMVOC emissions with about 615.2 kt (43% of the total global emissions) from domestic aerosol sprays because of the demand for and consumption of these aerosol spray products (See Appendix B). This also reaffirms the relationship between demand of the aerosol sprays and the associated increase in emissions of NMVOCs as a result. And from all the estimations conducted by the authors, the NMVOC emissions from the cosmetic category is the highest in Europe while the household category is higher in other regions, especially in Asia Pacific. The difference in these emissions can be attributed to the applications and the need for these aerosol products due to demographic dispositions.

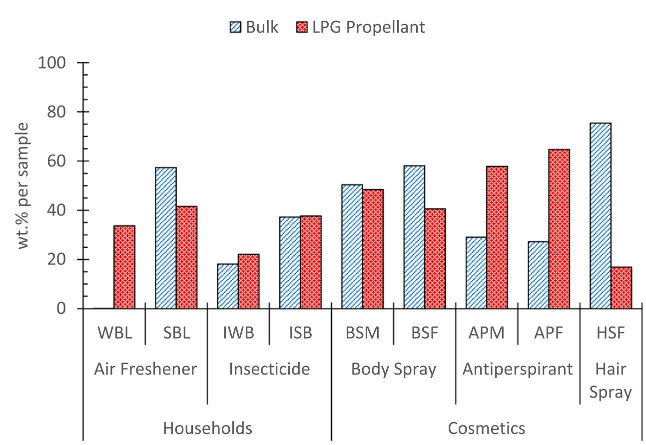

Fig. 6. NMVOCs (wt%) of product and propellant within aerosol samples with LPG propellant.

Table 5
The fraction distribution of NMVOCs within the selected aerosol sprays with LPG propellant.

Aerosol Sprays		Code	Non-Methane VOCs (wt%)					
			C_2	C_3	C_4	C_5	Ethanol (C_2)	TOTAL
Households	Air Freshener	WBL	0.1	10.9	21.8	0.9	0.1	33.8
		SBL	0.1	12.5	28.8	0.1	57.4	98.9
	Insecticide	IWB	0.1	6.8	14.4	0.9	18.3	40.4
		ISB	0.2	10.8	26.2	0.4	37.2	74.9
Cosmetics	Body Spray	BSM	0.1	14.9	32.6	0.8	50.4	98.8
		BSF	0.1	12.2	27.4	0.9	58.1	98.7
	Antiperspirant	APM	0.1	17.4	40.0	0.3	29.0	86.9
		APF	0.0	19.8	44.9	0.0	27.2	91.9
	Hair Spray	HSF	0.1	5.4	11.5	0.0	75.4	92.3

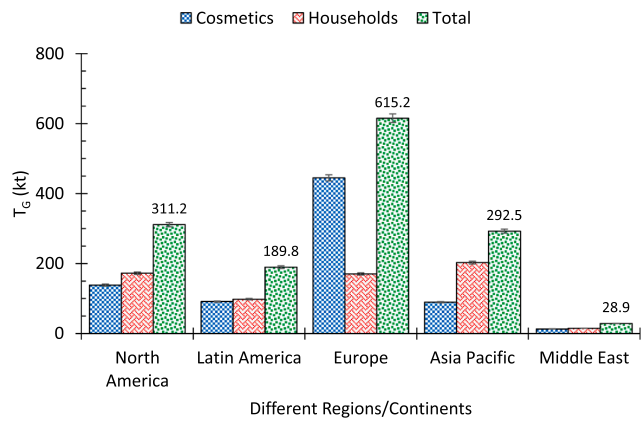

Fig. 7. Annual NMVOC emissions from selected domestic aerosol sprays in different continent.

3.4. Comparison of NMVOCs from selected domestic aerosol sprays (with LPG Propellant) and road transport in the UK

The magnitude and scale of these annual NMVOC emissions in the UK (about 61.2 kt), can be highlighted by comparing it to the NMVOC emissions from road transport in the UK. According to the vehicle licensing statistic of the UK (UK Department for Transport, 2018), there were about 37.7 million vehicles registered in the UK at the end of 2017. These include cars, light goods vehicle (LGV), heavy goods vehicle (HGV), motorcycles, and buses and coaches. Consequently, the cumulative NMVOC emissions resulting from these vehicles was about 29.9 kt at the end of 2017 (Office for National Statistics UK, 2019). Evidently, the total non-methane VOC emissions from domestic aerosol sprays is twice than those from the road. That means the level of NMVOC emissions from domestic aerosol sprays is equivalent to NMVOC emissions from about 77.2 million vehicles.

Whilst the NMVOC components and their toxicity for road transport and domestic aerosol sprays may differ in compositions, the presented data here intends to provide the quantitative landscape of the problem. As the road transport regulation is evolving very fast and the industry is moving towards "greener" pathway through electric vehicles, similar considerations are also needed in aerosol industry and regulatory bodies regarding the scale of NMVOC emissions from the utilisation of their products before it becomes more serious issues and dangers in future than they are at present. Prolong exposure to these potentially hazardous emissions is a leeway to a critical problem which can, thus, be tackled by adopting immediate stringent measures. Majority of cosmetic aerosol sprays are applied directly to the skin and have been shown to have a remarkably high dermal retention factor of 100% as evidenced by Comiskey et al. (2015). This extends to the inferences of previous studies (Schraufnagel et al., 2019a, 2019b, Uhde and Schulz, 2015) where they stated that air pollution, generally, is not local to the respiratory system of the human body but affects every organ.

Also, NMVOCs from these domestic aerosol sprays can be precursors to tropospheric ozone formation and further react with one another to form other forms of secondary organic aerosols (SOA) through ozonolysis (Niu et al., 2017). Each NMVOC constituent of domestic aerosol spray has an ozone formation potential (OFP) and global warming contribution with varying degrees of efficacy. The evaluation of this property at "source" is certainly important with respect to the concentration of each component which will further highlight the potential contribution to outdoor pollution resulting from the use of organic-based aerosol sprays.

3.5. Potential environmental impact of domestic aerosol products at "source"

Here, the environmental impact of aerosol sprays is discussed and analysed. The ozone formation potential (OFP) of the NMVOCs in the products and their global warming contribution were evaluated. For context, however, that the evaluation of these quantities was at "source". Thus, not all the NMVOCs from the aerosol products are emitted into the atmosphere as reported by Niu et al (2017), as some of these emissions fall/settle on furniture and flooring.

3.5.1. Ozone formation potential (OFP)

Using Eq. (7) and data for each component analysed in Table 5, the OFP was estimated based on the MIR of various VOC species provided by Carter (1994). The concentration of each species was expressed in g/L of product since the assessment of the OFP is at "source". The average OFP in each type of aerosol can thus be constructed as shown in Fig. 8.

As shown in Fig. 8, the household category has lower OFP values compared to the cosmetics. Air freshener solvent-based (SBL) has more OFP compared to air freshener water-based (WBL). This trend is also the same for insecticides as ISB has higher OFB compared to IWB and this is due to the fact the solvent-based products contain ethanol.

The similar results were also observed in Fig. 5, where the water-based samples showed lower NMVOCs compared to the solvent-based. In the cosmetic category, however, APM and APF had lower ozone formation potential compared to other samples in this category (body spray and hair spray). Body sprays contained more NMVOCs per can compared to hair spray (HSF). However, hair spray sample presented more OFP with about 105.1 g/L which is higher than both body sprays and antiperspirants. This is due to the fact that hair spray sample is attributed to higher volatile content (i.e. ethanol) as shown in Fig. 5. Dinh et al (2015) realised similar pattern where the hair sprays exhibited higher OFP values compared to the other products tested, albeit not at "source" but the abundance of ethanol within the aerosol spray plume at ambient condition was responsible. Spray products have the potential to be ozone forming compared to other anthropogenic sources of NMVOCs as a result of their dispersive properties.

Furthermore, the MIR values for various species of VOCs from Carter (1994) are directly proportional to the OFP values and range from 0.25 to 1.38 g O_3 per g VOC. The OFP values obtained using the MIR method is significant and this will unequivocally highlight the economic importance of the NMVOCs used in domestic aerosol sprays and their role in the contribution to ozone formation and indoor and outdoor air pollution wholly. To the authors' knowledge, there is no available literature which has tackled the quantification of NMVOCs in domestic aerosol sprays and also their corresponding ozone formation potential at

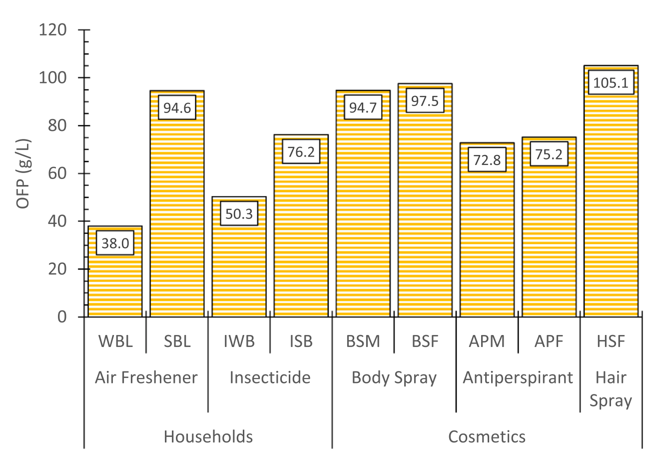

Fig. 8. Ozone formation potential (OFP) for selected domestic aerosol sprays with LPG propellant "source".

"source".

3.5.2. Global warming contribution

Using Eq. (8) and data from Table 5 to obtain the mass of each propellant component, the amount of carbon dioxide equivalency (CDE or CO_2e) was calculated for the selected domestic aerosol sprays in this investigation and results are presented in Table 6.

From Table 6, antiperspirants, within the cosmetic category, and air fresheners, within the households' category, have the highest global warming contribution per can, since they contain a higher level of LPG propellant. So, there is a direct correlation between the global warming contribution and the amount of LPG. This is also alarming, as LPG accounts for 80% of all aerosol dispensers as earlier stated. According to the report by Department for Business, Energy, and Industrial Strategy (Department for Business Energy and Industrial Strategy, 2020), the level of the UK greenhouse gas emissions from the residential sector (including combustion for heating and cooling, garden machinery, and fluorinated gases from aerosols and metered dose inhales) in 2018 were about 69.1 $MtCO_2e$. Thus, the results from this analysis, without considering ethanol as the most abundant constituent in aerosol spray, showed domestic aerosol sprays contribute 0.18% of the total CDE from the UK residential sector. Considering the total annual consumption of the domestic aerosol sprays in the UK in 2018, the total global warming contribution from cosmetic and household categories can be estimated to be 129.8 $ktCO_2e$. For organic compounds used in everyday products that have received passive attention in the past, this is alarming as they have the potential to not only affect the indoor air quality but the outdoor as well in the form of global warming.

Fig. 9 presents the CDE of each continent from propellant of domestic aerosol sprays from a global viewpoint. As it shows, the total CDE around the world is about 3154.8 $ktCO_2e$ in which Europe contributes to approximately 41% followed by North America (~22%) and Asia Pacific with about 21%.

Therefore, in order to reduce NMVOC emissions and improve the indoor air quality, a revisit to the use of organic compounds in domestic aerosol product must be made. This can be through the replacement of the LPG propellant with the clean compressed air or alternative gases which do not pose a danger to the environment.

3.6. Potential benefits of replacing LPG propellant with compressed gas for domestic aerosol sprays

The total amount of NMVOC emissions measured for the different domestic aerosol sprays have already been established and shown in the previous Sections. However, this section now demonstrates the analysis of replacing this potentially harmful and hazardous LPG propellant with the compressed gas such as nitrogen under three different scenarios (see Appendix C). In the *first scenario,* it was assumed that all LPG propellant is removed and replaced with nitrogen whilst the same level of the bulk in the aerosol can were maintained. In the *second scenario,* the LPG propellant was completely removed and replaced with nitrogen but the level of the bulk in the can was increased to 60% by volume. The *third scenario* shows the current commercial attempt to replace the propellant.

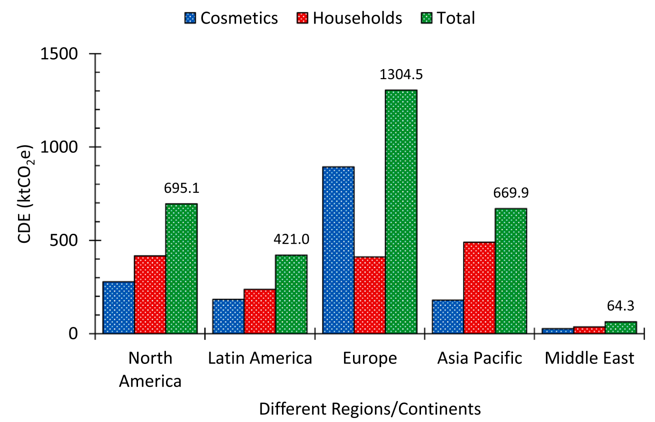

Fig. 9. Worldwide carbon dioxide equivalency of propellant from domestic aerosol sprays.

Scenario-I:

This scenario was adopted for simplicity given that to replace LPG with nitrogen, the volume of the bulk must be increased to compensate for the shelf life of the aerosol spray, as nitrogen does not have the compression and expansion properties of LPG (Nourian et al., 2014, Nourian et al., 2015, Nourian et al., 2016). The bulk volume alteration of the aerosol formulation depends on the targeted application of aerosol spray and is not within the scope of this work. Fig. 10 shows the potential reduction in NMVOC emissions from the selected domestic aerosol sprays by the replacement of LPG propellant with nitrogen.

For household category, the water-based samples have significant reduction compared to the solvent-based counterparts. This is because most of the volatile components come from the propellants whereas, in

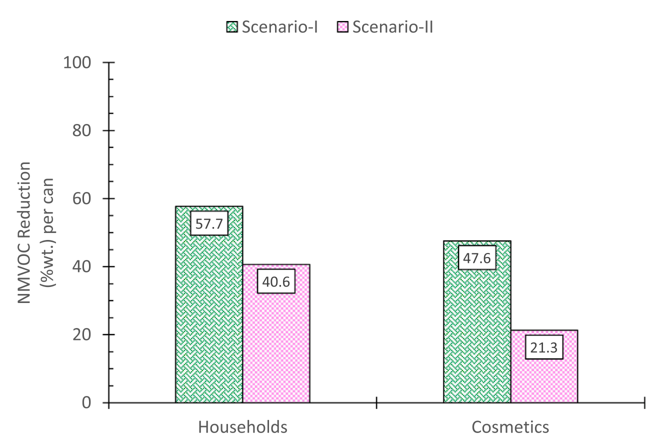

Fig. 10. Reduction of NMVOC emissions by replacing LPG propellant with nitrogen.

Table 6
Global warming contribution of propellant from selected domestic aerosol sprays.

Aerosol Sprays		Code	g CO_2 Equivalent				
			Ethane (C_2)	Propane (C_3)	Butane (C_4)	Total Propellant	Average
Households	Air Freshener	WBL	2.5	92.0	224.7	319.2	288.85
		SBL	2.8	96.7	236.8	336.3	
	Insecticide	IWB	2.9	74.8	155.3	233.0	
		ISB	2.9	80.3	183.7	266.9	
Cosmetics	Body Spray	BSM	1.2	47.7	122.5	171.4	234.66
		BSF	0.4	23.8	63.9	88.1	
	Antiperspirant	APM	1.6	94.0	246.4	342.0	
		APF	0.2	98.8	274.6	373.6	
	Hair Spray	HSF	1.0	54.4	142.8	198.2	

solvent-based samples, the product contains significant NMVOCs as shown in Table 4. Conversely, given the ethanol content in both male and female deodorants (BSM and BSF) in the cosmetic category, a far less reduction was seen compared to the antiperspirants (APM and APF) where there was less ethanol and more propellant. Hair spray had the lowest reduction as it has the highest ethanol content in all the test products which infers that the bulk of the NMVOC emission comes from the bulk and not the propellant.

Scenario-II:

By replacing the total volume of the LPG in a can by an equivalent amount of compressed gas, the total weight of the can will inevitably be reduced. This may thus be contentious by the aerosol industry since such conversion could have an adverse effect on the customer perceptions and hence impact on the overall product sales. Therefore, in this scenario, there is an assumption that (i) the LPG propellant is completely replaced by compressed gas and at the same time the volume of the product increased to 60%, (ii) the proportion of the active ingredients and solvent kept same.

On the average, about 53% reduction in NMVOCs can be realised by replacing the LPG propellant with compressed gas (i.e. nitrogen) in domestic aerosol sprays with a direct replacement. Furthermore, even with an increase of 60% of the total volume of the product in the cans to augment the effect of the replacement of LPG with nitrogen, there will still be a reduction in total NMVOC emissions by about 31%.

Scenario-III:

In view of the above scenarios, it is worth noting that there have been attempts by a few FTSE aerosol companies in replacing the LPG by nitrogen for air fresheners. As shown in Table 4, by replacing the LPG propellant with nitrogen for water-based air freshener, there is a possibility of reducing the NMVOC emissions by 99.8%. It, also, seems that the aerosol industry has begun noticing the necessity and the benefits of converting to cleaner eco-friendly aerosols. The potential benefits of this, however, have not been completely preserved. Fig. 11 shows the results of two more air freshener samples (see Table 1) filled with nitrogen propellant (WBN and SBN) and comparison of the NMVOC emissions with LPG filled air freshener samples (WBL and SBL).

As can be seen, the highest NMVOCs is for SBL air freshener with about 155.71 g per can and the lowest one is for WBN air freshener with about 12.70 g per can. This shows almost 92% reduction in NMVOCs by replacing the LPG propellant with nitrogen propellant. Comparably, this is about 7% lower than what the present authors previously recommended (about 99.8% reduction). This is due to the additional solvent component (i.e. ethanol) that mixed with the active ingredients of the bulk.

As can be seen in Fig. 11, the level of NMVOCs was found to be higher for the second commercial air freshener (SBN) compared to WBN air freshener. This level of increase in NMVOCs are mainly due to the size

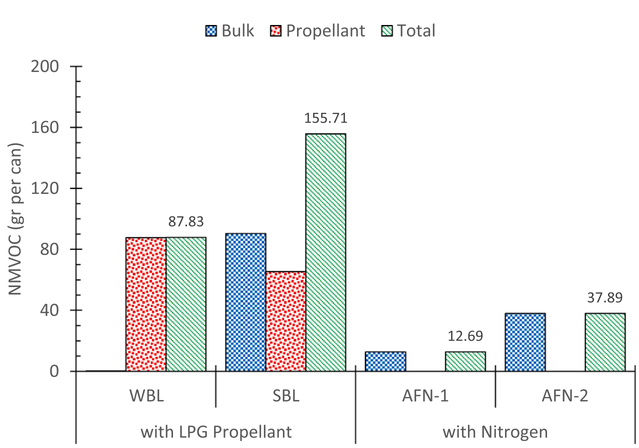

Fig. 11. Comparison of NMVOC emissions for air freshener using LPG propellant vs compressed gas (commercially available).

differences of the corresponding sample that were used. However, compared to the similar air fresheners filled with LPG, there is a reduction of 57% compared to WBL and 76% compared to SBL.

It is clear from these analyses that the replacement of LPG propellants with compressed gas (i.e. nitrogen), can have a significant effect on the NMVOC emissions. These products can also contain chemical such as formaldehyde which can damage health and combine with outdoor pollution create harmful particles. The reduction to 'Near-ZERO' emission for aerosol sprays is also immensely possible on the proviso that the chemical formulation products to be converted, at the gradual pace, to the water-based products.

4. Conclusion

A different approach to the quantification NMVOC emissions from the different categories of the domestic aerosol product was carried out through laboratory testing and analyses of the emission at "source". From the results obtained, certain insights were for the first time demonstrated, and the following conclusions can be drawn:

- Results indicated that cosmetic samples contained high concentrations of NMVOCs with male (BSM) and female (BSF) body sprays having 98.8 and 98.7 wt% respectively. Hair spray (HSF) contained 92.3 wt% of NMVOCs and male and female targeted antiperspirants (APM and APF) comprising of 86.9 and 91.9 wt% respectively. For household products, the solvent-based air freshener with 98.9 wt% NMVOC was the highest.
- Water-based products had less NMVOCs in all the analyses and it is due to the absence of organic solvents such as ethanol which was present in all the other samples tested.
- The majority of the NMVOCs identified in this work are ethane, propane, *iso*-butane, butane, *iso*-pentane; which are predominantly LPG components, and ethanol. This LPG propellant contributes to about 52% of the total NMVOCs in domestic aerosol sprays and replacing it with compressed gas propellant (i.e. nitrogen) will lower annual NMVOCs emissions by 30 to 52% which is beneficial to both consumers and the industry at large.
- Any action in reducing the emission in the aerosol sector can lead to an immediate impact on Indoor Air Quality (IAQ) as well as the public health since most of the aerosol sprays are utilised within the indoor environment
- The annual NMVOC emission in the UK emanating from the use of domestic aerosol sprays was evaluated to be 61.2 kt/year. This magnitude of the NMVOCs emission was reaffirmed by evaluating the road transport emissions to provide a scale of reference and an equivalent NMVOC emission from 77.2 million cars in the UK alone was realised. Globally, 1437.6 kt, which is equivalent to 1.8 billion cars.
- The ability of the NMVOCs from aerosol spray product to form tropospheric ozone was also highlighted. Hair spray has the highest ozone forming potential with about 105.1 g of Ozone per litre of the product.
- LPG propellants from domestic aerosol sprays also have global warming potential effects with a cumulative carbon dioxide equivalent of 129.8 $ktCO_2e$ in the UK which is which is around 0.2% of the total greenhouse gas emissions of the UK residential sector. This value globally is equivalent of 3154.8 $ktCO_2e$ as a result of using LPG propellants.
- The issue of NMVOC emissions from domestic aerosol sprays cannot be overemphasised. It is, therefore, vital to expedite the development of alternative technologies utilising 'clean air' such as compressed air or nitrogen and move to water-based product formulation. This enables circumventing the impending danger posed by current and conventional technologies in the aerosol industry.
- The replacement of LPG propellants with compressed gas (i.e. nitrogen), can have a significant effect on the reduction of NMVOC

emissions from aerosol sprays. The reduction to 'Near-ZERO' emission for aerosol sprays, as demonstrated in this study, can also be the assured choice for the aerosol industry to move to the water-based products' formulation.

CRediT authorship contribution statement

A. Nourian: Writing - review & editing, Methodology, Investigation, Data curation, Visualization, Supervision. **M.K. Abba:** Writing - original draft, Methodology, Investigation, Data curation. **G.G. Nasr:** Writing - review & editing, Data curation, Supervision, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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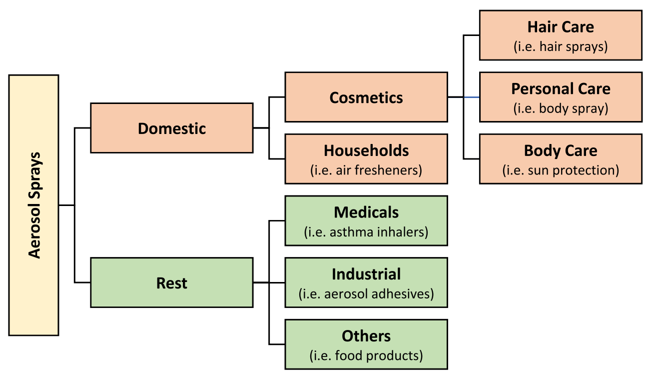
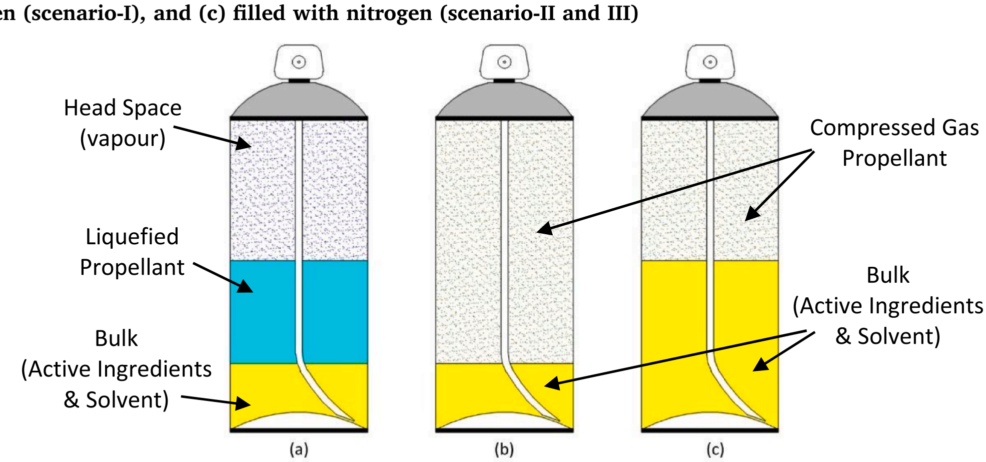
Appendix A. Different categories of aerosol sprays



Appendix B. Global aerosol sprays demand by different region in 2020 (million units)

	Domestic		Rest (Medicals, Industrials & Others)	TOTAL
	Cosmetics	Households		
North America	1185.5	1443.3	2685.1	5313.9
Latin America	781.9	822.3	589.8	2194.0
Europe	3804.7	1425.2	1346.9	6576.8
Asia Pacific	766.2	1696.8	582.3	3045.3
Middle East	114.9	129.1	79.8	323.8
TOTAL	6653.2	5516.7	5283.9	17453.8

Appendix C. Schematic illustration of the content's distribution within an aerosol spray samples, (a) filled with LPG propellant, (b) filled with nitrogen (scenario-I), and (c) filled with nitrogen (scenario-II and III)



Appendix D. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2020.106152.

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