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POLICY BRIDGE

Global emissions of VOCs from compressed aerosol products

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Disposable compressed gas aerosols have been a ubiquitous part of life since the mid-1950s. The signing of the Montreal Protocol in 1987 led to aerosol propellants changing from halocarbons to less damaging replacements; around 93% of current aerosol emissions by mass are volatile organic compounds (VOCs), with small contributions from compressed air (6.6%) and fluorocarbons (0.4%). The global consumption of aerosol units has increased significantly since the signing of the Montreal Protocol, increasing by an order of magnitude in some countries. In high-income countries, annual consumption increased through the 1990s and 2000s, typically reaching a plateau of approximately 10 ± 3 units person^{-1} year^{-1}, dependant on product preferences. The largest contributors of both units and mass emissions are personal care products (PCPs). Consumption of aerosols in lower- and upper-middle income countries are growing rapidly, for example, Brazil, Mexico, China, Thailand, all tripling reported consumption since 2006. Based on evidence drawn from national production estimates, product specifications and formulations, and interpolation of usage between countries of similar economic status, we estimate global emissions of VOC from aerosol propellants were approximately 1.3 ± 0.23 Tg year^{-1} in 2018. The fraction of anthropogenic VOC emissions accounted for by aerosols has in some countries increased significantly as emissions from vehicles and fuels have declined. For example, in the UK, 6.1% of anthropogenic VOC emissions were from aerosols in 2017, more than were released from gasoline passenger cars. Should low- and middle-income economies grow consumption per capita in line with recent trends, then we project global aerosol consumption may reach approximately $4.4 \pm 0.96 \times 10^{10}$ units year^{-1} in 2050. Should existing national and international policies on aerosol product formulation remain unchanged, and VOCs remain the dominant propellant, compressed aerosols could account for a global emission of approximately 2.2 ± 0.48 Tg year^{-1} in 2050.

Keywords: VOCs, Air quality, Emissions, Aerosols, Montreal Protocol

1. Introduction

Aerosols dispensers have been extensively used in professional and consumer products across the globe for over 70 years. Their ease of use and effectiveness for product application has aided their popularity, with 5.6 billion units of aerosols being manufactured in 2018 in Europe alone (European Aerosol Federation [FEA], 2019). Norwegian chemical engineer Erik Rotheim patented the first aerosol spray can and valve, which was capable of both holding and dispensing products in 1927 (National Aerosol Association, n.d.). However, it wasn't until the end of World War II that aerosol products began to be mass produced, with the American government creating the first insect repellent, known as the "bug bomb," to help protect servicemen from diseases such as malaria. After that, the commercial use of aerosols quickly expanded to include hair spray, air freshener, deodorant, and shaving foams, all manufactured for general public use. These products soon became available in Europe, creating the first boom in aerosol product popularity in the 1960s and their use has continued globally in the 21st century (FEA, 2016).

There are three chemical components to an aerosol product: the active ingredient, a solvent, and a propellant. The active ingredient is the portion of the product intended for application and is concentrated in the form of a solution, suspension, emulsion, or powder (UNC Eshelman School of Pharmacy, n.d.). The propellant dispenses the product while sometimes also acting as a cosolvent. The propellant can be a liquefied or compressed gas and can comprise anywhere from 5% to 90% of the total product mass depending on its intended use. The aerosol propellant forces the product out of the can when the nozzle is depressed. The difference in pressure between the propellant inside and the pressure of the outside air triggers product release. Additionally, propellants can act to disperse the product into a fine mist on evaporation as the active ingredients are broken up. These particles can be expelled in the form of droplets, foam, paste, or

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powder depending on the dispersing ability of the propellant and the force with which they are dispensed (FEA, n.d.).

Initially chlorofluorocarbons (CFCs) were used as aerosol propellants; their nonflammable and odorless properties and chemical stability made them ideal for use in consumer products. However, concerns associated with their environmental impact were catalyzed by Molina and Rowland's (1974) paper detailing the possible destruction of ozone arising from halogens released through CFC photodissociation in the stratosphere. The CFCs used in aerosols proved to be extremely potent ozone-depleting substances and having high global warming potentials (GWPs) between 1,100 and 14,000 for a 100-year time period (Greenhouse Gas Protocol, 2016). The establishment of the Montreal Protocol (signed 1987, effective 1989) led to all United Nations (UN) member states agreeing to regulate the production and consumption of almost 100 man-made chemicals including CFCs (United Nations Environment, n.d.). This was later amended (The Montreal Amendment 1997) to include the phasing out of hydrochlorofluorocarbons (HCFCs), which were also being used as aerosol propellants, and several other amendments shortening the time frame for the overall phase out of these chemicals from use. The most recent, the Kigali Amendment (signed 2016, effective 2019) goes further still in curbing ozone-depleting substances. All signing parties have agreed to reduce the production and consumption of hydrofluorocarbons (HFCs), another aerosol propellant, by more than 80% by 2047 (United Nations Industrial Development Organization, n.d.). The current status of the amendment is that 112 parties have signed out of 193 member states (United Nations, 2015).

The implementation of the Montreal Protocol was extremely successful and led to the rapid replacement of halocarbon-containing propellants with simple short lifetime volatile organic compounds (VOCs), replacement chemicals that were of considerably lower (although not zero) environmental impact. The role of VOCs in promoting tropospheric ozone and photochemical smog was established at the time of the Montreal Protocol; however, the air quality degradation arising from their use as aerosol replacements was considered as far less significant than the negative impacts of stratospheric ozone depletion. Since VOCs were introduced as halocarbon replacements in aerosols, there has been limited reassessment of the impacts of that policy decision from an air quality perspective or the future trajectory and appropriateness of VOCs as replacements for the coming decades.

Table 1. Compressed aerosol propellant properties. DOI: https://doi.org/10.1525/elementa.2020.20.00177.t1

	Propellant Properties			
Propellant Type	**Flammable?**	**Pressure**	**Purity[a]**	**Cosolvent Required?**
Hydrocarbons	Yes	High	Varies	Yes
DME	Yes	High	High	Rarely
Compressed gas	No	Low	Varies	Yes

DME = dimethyl ether.

BOC (2015), PURmate (2018), Diversified CPC International (n.d.).

[a]Odorless.

2. Aerosol propellants currently in use

The changes required by the Montreal Protocol were not technically problematic for aerosol manufacturers since CFC and HCFC alternatives were already being used in countries where pre-Montreal Protocol bans of ozone-depleting substances were in place, such as the United States and Sweden (Directorates General [DG] Enterprise and Industry, 2014). A majority of manufacturers elected to use hydrocarbon blends as a replacement propellant, and this remains the most common formulation today. Most hydrocarbon aerosol propellant (HAP) blends are made up of propane, *n*-butane, and *iso*-butane, with other hydrocarbons such as *iso*-pentane and *n*-pentane, sometimes included. There are no standard HAP formulae, and many different manufacturer-specific blends are found in consumer products, each with a different vapor pressure—a key factor controlling aerosol performance. Generally, a blend of propane and *n*-butane will have a lower vapor pressure than one of propane and *iso*-butane (Diversified CPC International, n.d.), and this will have an effect on its dispensing properties. Despite their range of uses, hydrocarbons are not always an appropriate propellant. Some PCPs, which can be defined as any cosmetic or hygiene product available to the public for personal use (Yeoman et al., 2020), require a very pure and odorless propellant for which dimethyl ether (DME) is the most common solvent used. Another alternative is compressed gas (e.g., N_2 or air), which is considered the most environmentally friendly option.

Table 1 gives an outline of the three main propellant types, highlighting the properties which manufacturers consider in their selection. Choosing an appropriate propellant ultimately depends on two things: the vapor pressure and purity required (although cost may be an influencing factor). The higher the vapor pressure, the higher the degree of dispersion from the aerosol can, and the finer and drier the product mist. High vapor pressure products include air freshener, flying insect spray, and spray paint, whereas lower vapor pressure products include shaving cream, gels and mousse, and perfume (DG Enterprise and Industry, 2014). A medium vapor pressure product, for example, hair spray, deodorant, or furniture polish, has a wet application and moderate dispersion. Product dispersion is also controlled by the percentage of propellant in the can, with lower vapor pressure products having a low ratio of propellant to active ingredient. These are important factors considered in the following section on emissions (see **Table 2**).

The breakdown of the types of propellants used for aerosol products is shown in **Figure 1**, with hydrocarbon-based propellants dominating the mass of emissions, and

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Table 2. Simplified aerosol product volume and pressure assumptions. DOI: https://doi.org/10.1525/elementa.2020.20.00177.t2

Product	Average Size (mL)	Pressure
Air fresheners	300	High
Automotive	500	Medium
Colognes/perfumes	75	Low
Deos/body sprays/anti-perspirant	250	Medium
Hairspray products	300	Medium
Hard-surface cleaners	750	Low
Industrial	300	High
Insecticides	300	Medium
Medical (excluding inhalers)	300	Medium
Miscellaneous	300	Medium
Other household	300	Medium
Other personal	300	Medium
Oven cleaner	300	Medium
Paints/lacquers	400	High
Shaving products	200	Low
Shoe/leather cleaners	250	Low
Starches	300	Medium
Suntan/bronzing products	200	Medium
Veterinary/pet care	300	Medium
Waxes/polishes	400	Medium

DG Enterprise and Industry (2014).

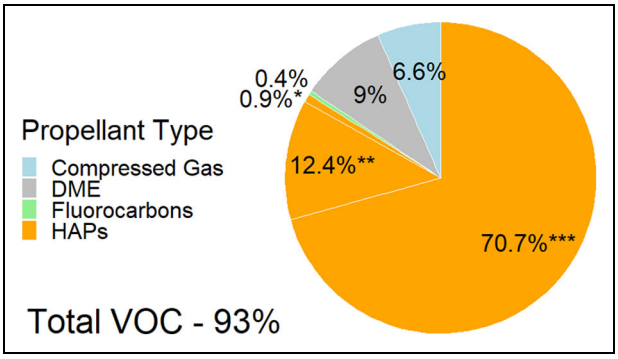


Figure 1. Consumption of aerosol propellants by type (2012; DG Enterprise and Industry, 2014). HAP speciation has been made using the reported composition of aerosol products included within the UK National Atmospheric Emissions Inventory available at www.uk-air.defra.gov.uk. HAPs and DME have been combined as total VOC. HAP = hydrocarbon aerosol propellant; DME = dimethyl ether; VOC = volatile organic compound. *Pentane. **Propane. ***Butane. DOI: https://doi.org/10.1525/elementa.2020.20.00177.f1

particularly the C_4 alkane isomers (DG Enterprise and Industry, 2014). Although these data are from 2012, we assume in this work, and later calculations, that there has not been a significant change in distribution between these broad chemical classes over the last decade. We assume that without any major policy change on aerosol composition and usage, this would also hold for the foreseeable future. In combination, hydrocarbon-based propellants and DME make up 93% of emissions, which we describe collectively henceforth as "VOC emissions" labeled on **Figure 1** as "Total VOC" Products using compressed air makes up 6.6% of mass, and 0.4% is from products using fluorocarbons (largely medical devices such as inhalers). Since compressed air has no direct environmental impacts once emitted, and HCFCs make up an insignificant amount of mass of emissions (e.g., 0.4% of approximately 1–6% of VOC in most countries), in the remainder of this article, we discount this mass in subsequent calculations and report mass emissions from aerosols as they relate to VOCs.

3. Atmospheric impacts and health effects

To assess the atmospheric impact of aerosol propellant emissions beyond stratospheric ozone depletion, which was considered in the Montreal Protocol, two major additional impacts from propellants are examined: (i) the GWP of the gas once released and (ii) impact on health (toxicity), either directly or *via* the contribution of those emissions to secondary air pollutants such as ozone or $PM_{2.5}$. Indirect impacts on the atmosphere can also arise from the manufacturing activities that produce the propellant and other supply-chain impacts from raw material extraction, through production to distribution. The fluorocarbons, made up primarily of HFCs as they have only recently been targeted by the Montreal Protocol, are undoubtedly the most impactful of the four propellant groups by the metric of GWP. However, HFCs are only used in a very small range of products, typically health-related, and many are likely to be converted to alternative propellants as a consequence of the Kigali Amendment. In terms of GWP, VOCs are significantly more environmentally acceptable (e.g., DME compared to HFCs in Good et al. 1998), and while they do not have a large direct effect on radiative forcing, they do undergo photooxidation that generates tropospheric ozone, itself a greenhouse gas. As for the compressed gas propellants, carbon dioxide and nitrous oxide are both well known as greenhouse gases, but the mass associated with their aerosol use is insignificant compared to other anthropogenic sources.

The health impacts of VOCs that are used as propellants are either through direct inhalation toxicity or through a contribution to degraded ambient air quality. Solvent/inhalant abuse (Williams et al., 2007), the triggering of asthma attacks (Lovén et al., 2019), and fire risks from flammable propellants and pressurized cans are the direct health risks that usually coincide with short-term, high-concentration releases or exposure. When propellants are inhaled, they can take the place of oxygen in the lungs, causing nausea, vomiting, rapid breathing, and in severe cases, comas and death (Canadian Centre for Occupational Health and Safety, 2020). These hazards are

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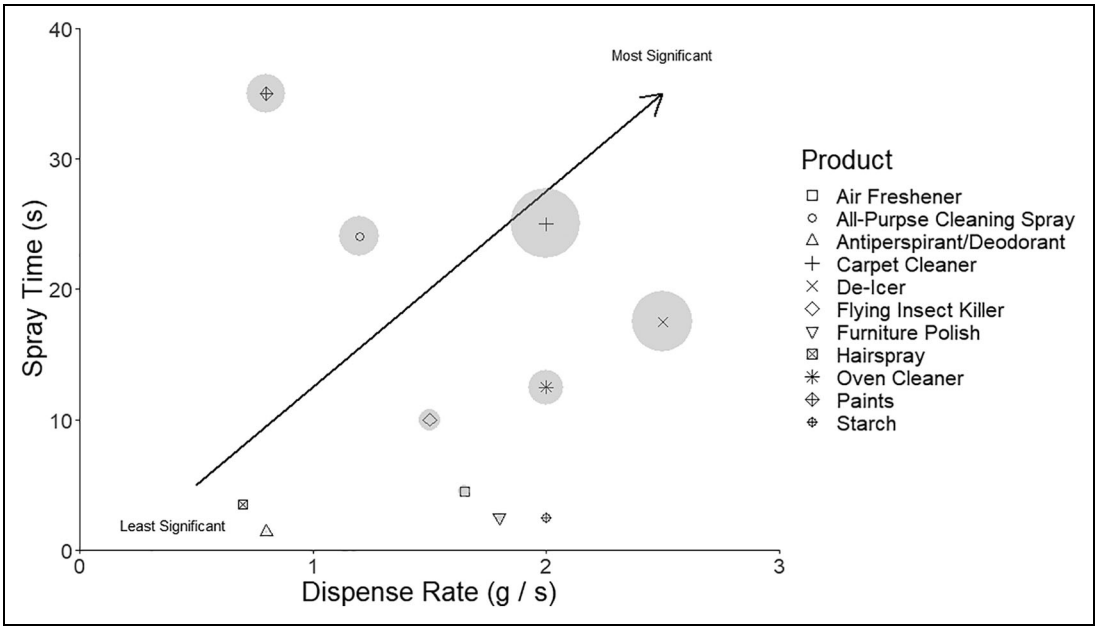


Figure 2. Propellant emission potential for a range of domestic products per unit of application by a user. Plot based on discharge rates and typical spray time, showing the median value where a range was given (European Aerosol Federation, 2013). The gray circles are sized to be proportional to the amount of aerosol propellant release per usage. DOI: https://doi.org/10.1525/elementa.2020.20.00177.f2

associated with inappropriate and unsafe use by users, yet even with careful and appropriate use, there remains potential for health impacts. The physiological response to hydrocarbon propellants was first studied in 1978 (Stewart et al., 1978). Although acute, single exposure (250, 500, and 1,000 ppm) to propellants, *iso*-butane and propane were shown to have "no untoward physiological effects" on pulmonary and cognitive function or cardiac rhythm, repetitive exposure to 1,000 ppm did cause minor cognitive decline. Additionally, participants had detectable traces of propellants in their blood and on their breath. Possible long-term toxic health effects could be caused by propellants making their way into the respiratory and cardiovascular systems; however, there have been no long-term studies on the inhalation of propellants from the continuous use of aerosol consumer products to evaluate this.

There are no current UN, European Union, or World Health Organization regulations or exposure limits for any of these compounds in ambient outdoor or indoor air. However, there are several workplace exposure limit guidelines from a range of organizations/agencies as presented in **Table 3**.

The more significant health impact of VOCs is their contribution to poor air quality. Propane and *n*-butane are the second and third most abundant nonmethane hydrocarbons in the atmosphere, with atmospheric lifetimes of approximately 13 and 7 days, respectively (Hodnebrog et al., 2018). In the presence of sunlight and NO_x, tropospheric ozone can form (Haagen-Smit and Fox, 1954). The contribution of VOCs to ozone is well-described (e.g., Derwent et al., 1996), as is the formation of other more harmful secondary aldehydes (Rosado-Reyes and Francisco, 2007). Significant attention has been paid to reducing emissions as part of air quality management at national and continental scales (as an example EC Directive 1999/13/EC, The Council of the European Union, 1999). The formation of secondary organic aerosols (SOAs) is also possible from propellants, although the by-products from their oxidation generate species with relatively limited SOA potential compared to other classes such as monoterpenes and aromatic compounds (Wang and Wang, 2016).

4. Emissions of VOCs from individual aerosol products

Since aerosols are used for a very wide variety of applications, unsurprisingly the emissions arising from each product are highly variable. For example, a high vapor pressure product will have a high mass dispense rate and will therefore emit more propellant and smaller particles over a short period of time (FEA, 2013). The overall emission of propellant is therefore the result of the propellant concentration, dispense rate, and the time in use or "spray time." **Figure 2** shows the relationship between dispense rate, typical spray time, and the amount of propellant released per application (FEA, 2013). The products in the "most significant" segment of the plot emit more per aerosol application, combining high dispense rates and longer spray times. This figure shows only the potential from each product type, the absolute emitted by each product class is dependent on how frequently each product is used and by how many people, and it is recognized that some items on this plot would be used only infrequently (e.g., oven, carpet cleaner) compared to others that may be used multiple times per day (e.g., deodorants, hair spray).

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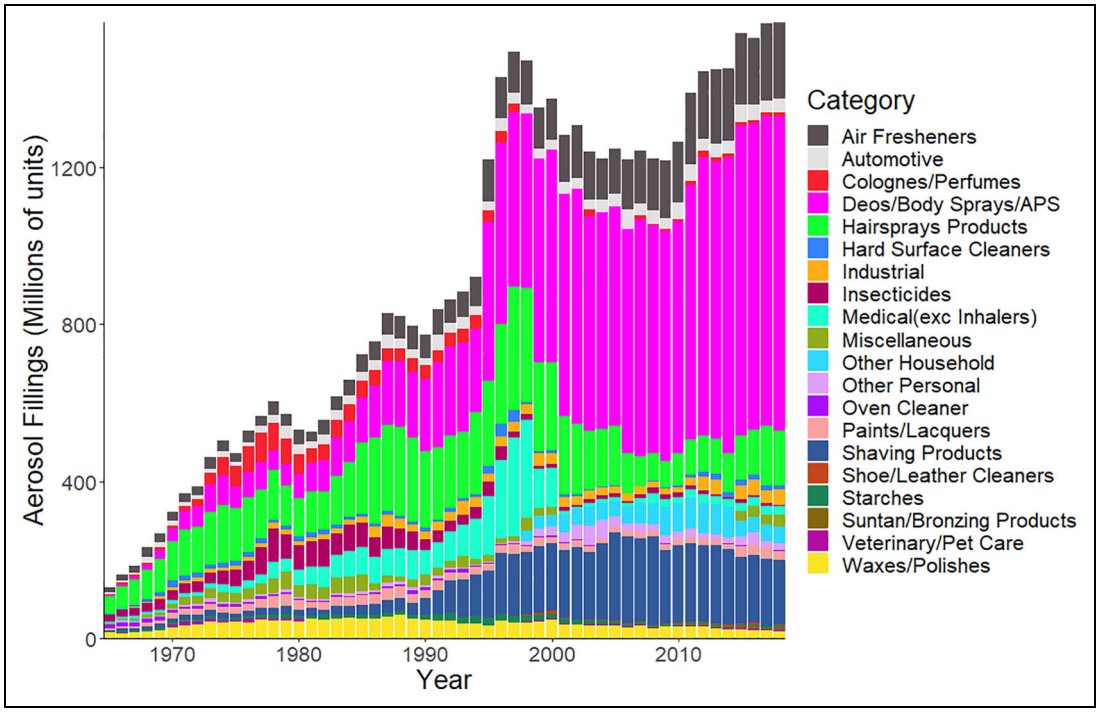


Figure 3. UK aerosol filling statistics by product class for the period 1965–2019 (British Aerosol Manufacturers' Association, data aggregated and combined from reports in 2015, 2018, 2019). DOI: https://doi.org/10.1525/elementa.2020.20.00177.f3

5. VOC propellant emissions at a national scale

A more complete assessment of the emissions from aerosol products can be derived from consumption statistics, and this can be evaluated using a combination of manufacturing and sales data along with information on the aerosol products themselves. In this section, we examine in detail aerosol propellant emissions for the UK, using this as a reasonable case study that is likely to be broadly representative of other high-income industrialized countries. In later sections, we look more widely at trends in other countries. We note however the significant heterogeneity in how data are reported between countries and that it is very difficult to generate exact like-for-like comparisons. **Figure 3** shows UK aerosol fillings from the year 1960 onward, with each unit of aerosol filling representing one can of product. Each unit is not however of a standard size/mass or volume of propellant; instead the graph shows only unit consumption rather than trends in mass of propellant emissions. For the avoidance of doubt, in this study, we are considering only emissions from individual disposable aerosol canisters. This is a quantity of VOC that is distinct from the total emission of VOCs that might be used as a propellant, for example, industrial car paint spraying may use hydrocarbons from bulk tanks.

To convert data on aerosol unit fillings into values representative of an atmospheric mass emission of propellant requires assumptions to be made about the average chemical composition of each unit, size, and pressure (which determines the amount of propellant in each).

Using recently published laboratory test data from Nourian et al., (2021) as a guide for our calculations, a 300 mL high-pressure aerosol product contains on average 83 g of propellant; differences in both volume (average product size) and pressures can then be scaled accordingly. A medium-pressure product will contain approximately 70% of the propellant of a high-pressure product and a low-pressure product approximately 25%. For example, air fresheners are typically sold in a 250 mL canister and are a medium-pressure product. Therefore, they will contain, on average, approximately 48 g of propellant per can (69 g in 250 mL, scaled down to 70%). **Table 3** shows the estimated aerosol product ratings to support a conversion of national unit–consumption statistics into a national atmospheric emission of propellant. Aerosol VOC emissions by mass and by-product for the UK are then shown in **Figure 4**.

The upward trends in **Figure 4** reflect several different factors, some related simply to population growth in the UK (from 52 million in 1960 to 67 million in 2020, the World Bank, n.d.) and some to consumer trends and habits. The consumption of some aerosol products is in decline, such as hair spray, whereas others show increasing trends, such as aerosol deodorants and air fresheners. Overall PCPs, and in particular deodorants, body sprays, antiperspirants, and hair spray, are consistently responsible for the largest portion of the aerosol market by both filling number and total mass of VOC emissions. In total, using this bottom-up methodology, an estimated ~80 kt of propellant, overwhelmingly as VOCs in the form of either simple hydrocarbons or DME, is emitted from the UK each year. Statistics and long-term data at this level of granular product detail are not available on a global scale, or indeed regional scale, but we consider these

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trends are likely broadly reflective of patterns in other European countries. Recent industry reported aerosol consumption patterns for Europe are shown in the supplementary material (Figure S1). Although European data are not reported using the same aerosol product taxonomy as the UK data, they show a similar pattern with PCPs forming the largest class of aerosol products. Although only covering 3 years of production, this demonstrates that the year-to-year demand and production for each product is reasonably constant, a fact that is further highlighted in the Impact Assessment Study on the Adaptation to Technical Progress of the Aerosol Dispensers Directive, **figure 2**.2 (DG Enterprise and Industry, 2014).

Table 3. Recommended exposure limits. DOI: https://doi.org/10.1525/elementa.2020.20.00177.t3

Propellant Type	Recommended Exposure Limits
Propane	TLV 1,000 ppm[a]
	AEGL 5,500 ppm[b]
Butane	STEL 750 ppm[c]
	TWA 600 ppm[c]
	AEGL 5,500 ppm[b]
DME	STEL 500 ppm[c]
	TWA 400 ppm[c]

DME = dimethyl ether; TLV = threshold limit values; AEGL = acute exposure guideline levels (over 1 h); STEL = short-term exposure limit; TWA = time weighted average (over 8 h).

[a]BOC (2005).

[b]Committee on Acute Exposure Guideline Levels (2012).

[c]Health and Safety Executive (2020).

6. Contributions to national VOCs emission budgets

During the period following the Montreal Protocol and the phase out of CFCs and HCFCs from aerosols, anthropogenic VOC emissions were, in most industrialized countries, dominated by emissions from road transport, fossil fuels and the associated extractive and refining industries. For example, see a recent analysis of multiyear sectoral VOC emissions trends in Lewis et al. (2020) for the UK. Although the emissions of VOCs from aerosols have generally been accounted for in emissions inventories, until the early 2000s propellant VOCs made up only a small fraction of any individual country's national emissions. Policies designed to reduce photochemical ozone pollution from the 1980s onward focused predominately on sources such as gasoline vehicle exhaust and fugitive emissions, and significant reductions occurred in many industrialized countries in the 1990s and 2000s. As transport and fossil fuel VOC emissions declined, other sectors such as solvents from household products have grown in significance in terms of their fractional contribution to VOC emissions (e.g., for the United States, see McDonald et al., 2018).

As an example, the emissions of VOCs from anthropogenic sources in the UK is shown in **Figure 5**, showing the total VOC from all sources, with aerosols highlighted individually as a source. These downward trends in estimated

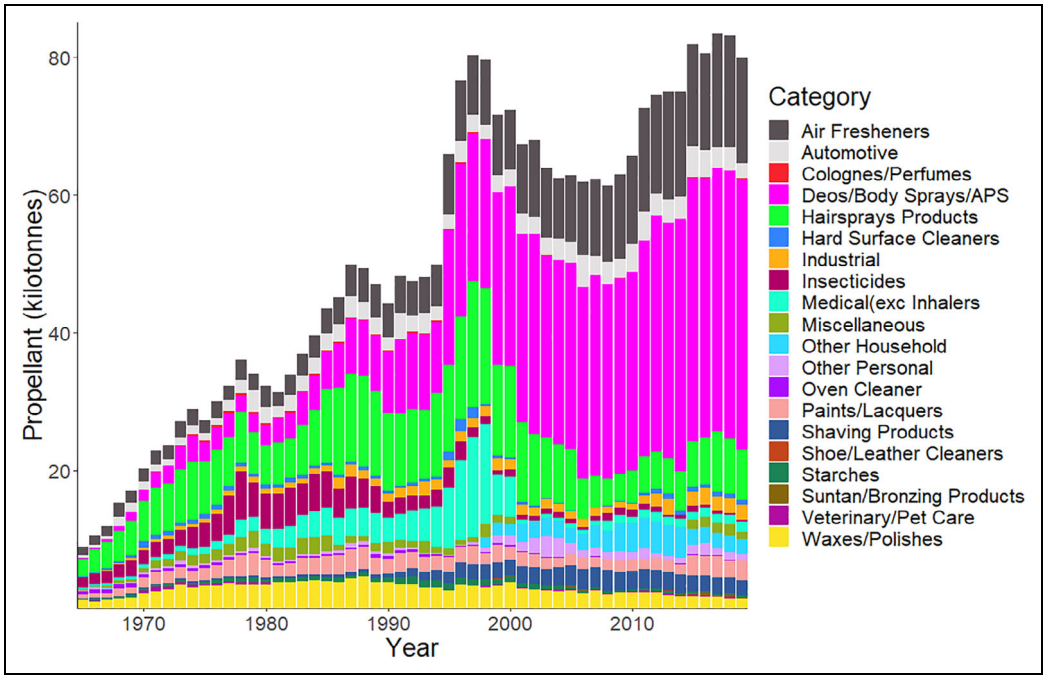


Figure 4. Estimated UK aerosol emissions in kilotons for all propellant types, constructed using industry fill reporting statistics from **Figure 3** and corrected for mass emissions specific to individual products based on fill assumptions (volume and pressure) from **Table 2**. DOI: https://doi.org/10.1525/elementa.2020.20.00177.f4

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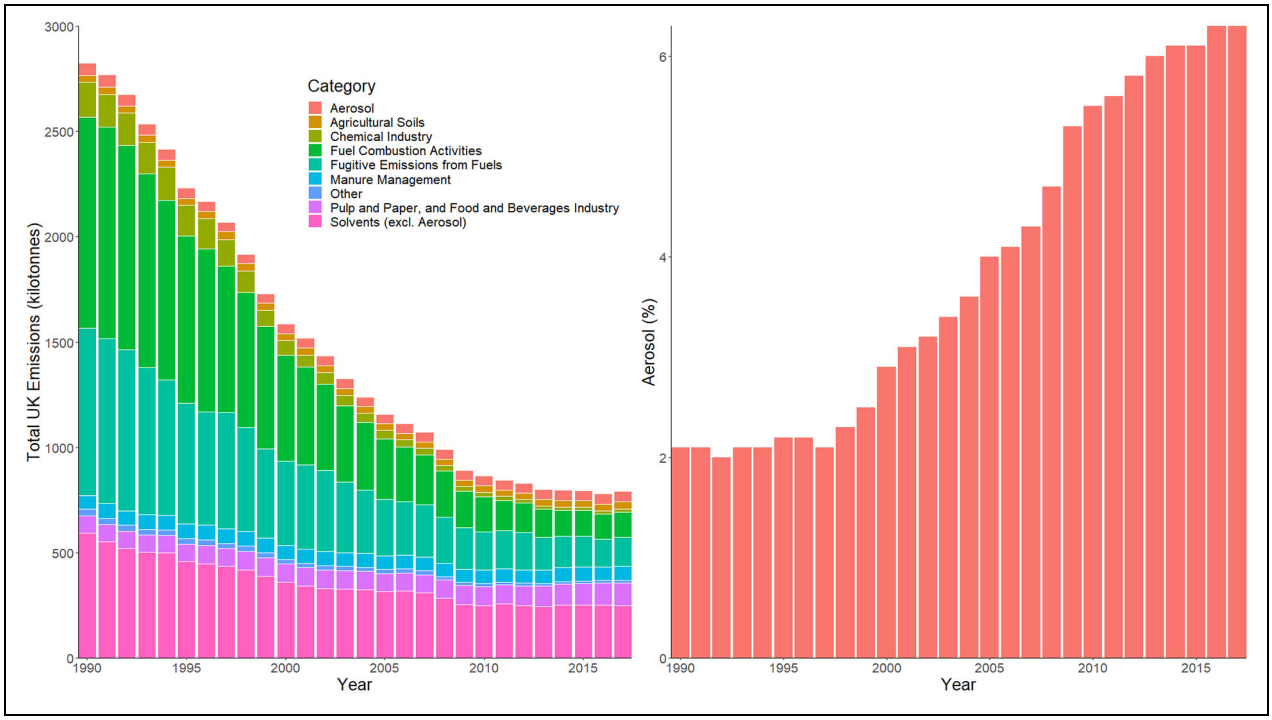


Figure 5. Total annual UK anthropogenic VOC emissions (excluding aerosols and biogenic) and VOCs from aerosols. Presented on the left-hand plot is the contribution of aerosols to total UK anthropogenic emissions, and on the right-hand plot, the percentage of the total it represents. VOC speciation and sectoral analysis have been made using the reported composition of aerosol products included within the UK National Atmospheric Emissions Inventory available at www.uk-air.defra.gov.uk. VOC = volatile organic compound. DOI: https://doi.org/10.1525/elementa.2020.20.00177.f5

emissions are also reflected in ambient data (e.g., Dollard et al., 2007; Lewis et al., 2020). The UK National Atmospheric Emissions Inventory (NAEI), from which the data in **Figure 5** are drawn, is highly detailed by sector and also by VOC species. Further details are described in Passant (2002). Taking the UK as likely representative of other countries in Europe in terms of consumer behavior and habits, the fraction of UK national emissions represented by aerosol propellants grew from around 2.0% of national emissions in 1990 to around 6.1% in 2017. Although this may superficially appear to be still a relatively modest contribution, put in perspective, the official inventory estimated UK emissions of VOCs from aerosol use in 2017 (approximately 60 kt pa) were greater than the total VOC emissions arising from all passenger cars in the UK (estimated as approximately 30 kt pa in 2017, Office for National Statistics UK, 2019).

7. Global consumption and future projections

Industry reported data on aerosol products are generally from trade bodies and at a national level. In high-income countries, this shows frequently that a broadly stable and consistent rate of production/consumption was reached in the mid-2000s. Countries for which long-term and internally consistent production data are available include the United States, Australia, Japan, China, Argentina, Mexico, Thailand, Brazil, Mexico, South Africa, and the continent of Europe grouped. We assume here that at a national level, production is a reasonable surrogate for consumption and emissions in high-income countries, although it is likely that some component of national production may be exported outside of the country of production. Reported data on aerosol unit production from a range of countries are shown in **Figure 6** along with a projection of the trend for each to 2050. There is evidence that since the start of the time series in 2005, most high-income countries have seen relatively little change in aerosol use. In **Figure 7**, the same data are expressed as a per capita value, correcting for population change over time in 1 year time steps. We produce per capita estimates by including the whole population (all ages) of a country since more specific data, for example, breaking this down as use by age demographic, do not exist.

Expressed per capita, there are notable absolute differences between countries, the United States being the highest per capita user of aerosols. The average annual per capita production from high-income countries is approximately 10 ± 3 aerosol units person^{-1} year^{-1}. The per capita use data for Argentina has been calculated as 55% of production, as it has been reported that they export 45% (Camara Argentina Del Aerosol, n.d.). With this adjustment, Argentinian per capita consumption rate is comparable with that of high-income countries and, as such, is an example of how an upper-middle income country can reach this value of 10 aerosol cans per person per year consumption rate.

Both figures also show the industry reported trends in aerosol production for a number of expanding middle-income economies (Argentina, Brazil, China, Mexico,

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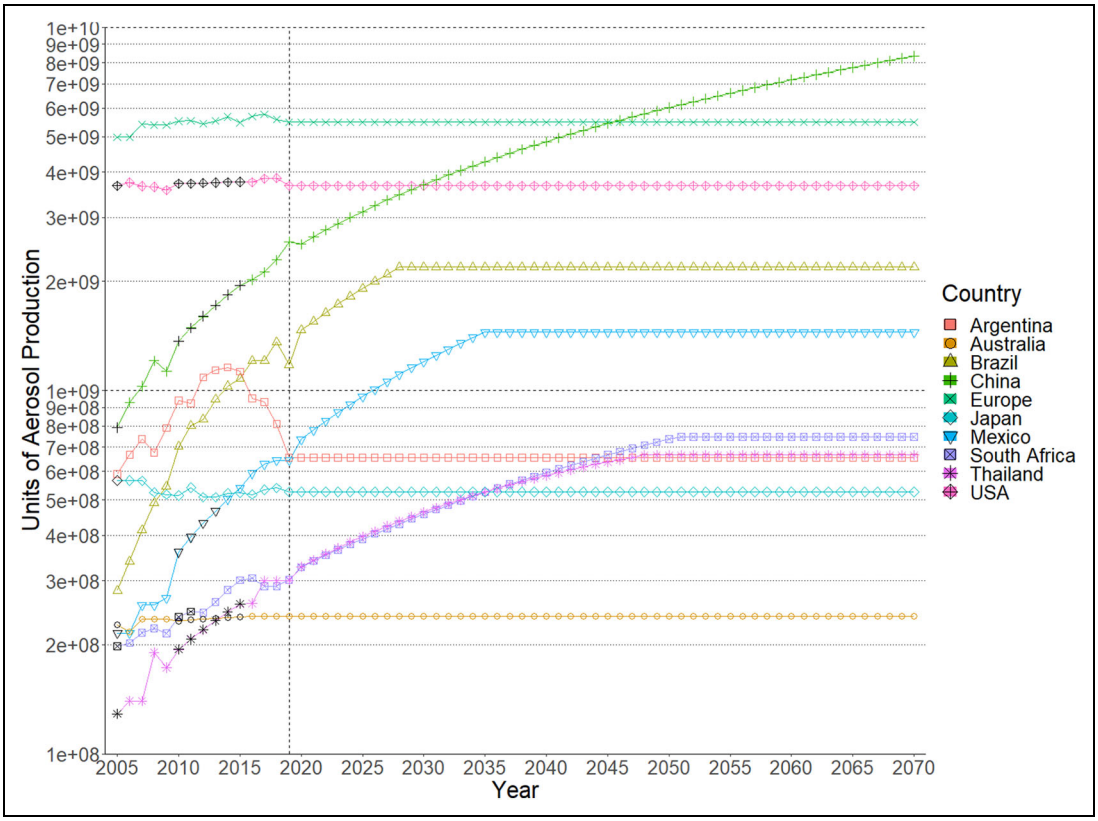


Figure 6. Global aerosol use by country and future projections based on recent trends. Where national reporting statistics are available, corrected for projected population growth indicated by the dashed line. Points in black where there are gaps in reporting data have been estimated using a linear regression (Camara Argentina Del Aerosol, n.d.; European Aerosol Federation, 2017, 2018, 2019, 2020; Spray TM, 2015, 2016a, 2016b, 2017a, 2017b). DOI: https://doi.org/10.1525/elementa.2020.20.00177.f6

South Africa, and Thailand). In each case, there is a significant growth in consumption over the reference period both in absolute number and as a per capita value. These recent trends are extrapolated as simple forward projections to 2050 but do not exceed a high-income 10 units person^{-1} year^{-1} value. **Figure 7** is annotated with a line marking of this high-income limit. We predict the year in which this will occur assuming that the future rate of production follows recent past trends. This per capita forecast data have been calculated using population predictions and are corrected for the expected large population growth in some emerging economies. First to reach 10 units person^{-1} year^{-1} would be Brazil in 2028, followed by Mexico in 2035, Thailand in 2048, and South Africa in 2051. China is not predicted to reach 10 cans per person plateau point in this time frame. Despite this, China will become the largest consumer of aerosol products in the world in the 2040s. We have assumed that once a country has reached the 10 units person^{-1} year^{-1} plateau, production and consumption remain constant since there is no historical precedent for a population using aerosols substantially above this rate per person.

The potential scale of future global aerosol consumption can be evaluated by first assessing the gross national income (GNI) of all nations, which tracks their wealth on an annual basis and then applying representative aerosol usage assumptions and trends to each GNI group. Every country has been categorized into one of the four following groups by the World Bank based on their GNI per capita (2019 data) using the World Bank Atlas Method (the World Bank, n.d.); low-income (less than US$1,035), lower-middle income (US$1,036–US$4,045), upper-middle income (US$4,046–US$12,535), or high-income (more than US$12,536) countries. A breakdown of these economy classifications can be found in Table S1. The majority of European countries fall within the high-income category, and as such, the whole of Europe has been classified as being high-income. We assume that all high-income nations have reached a consumption plateau, and the current and future average aerosol units consumed annually per person at 10 $\pm$ 3. The current average annual units per person for upper-middle income countries is estimated as 5 $\pm$ 2, although we note that our estimate is based on a relatively small number of countries in that GNI group that report annual statistics. We exclude China and Argentina from this group and treat them individually. We apply this estimate to all upper-middle income countries that do not report their usage data for the years up to 2020 and follow the projection that shows based on the extrapolation of past trends, that they will reach the maximum of 10 units per person per year by approximately 2050. Lower-middle income countries have been estimated to currently consume 2 $\pm$ 1 units per person annually and will reach 3 $\pm$ 1 by 2050. Low-income countries are assumed to currently have no

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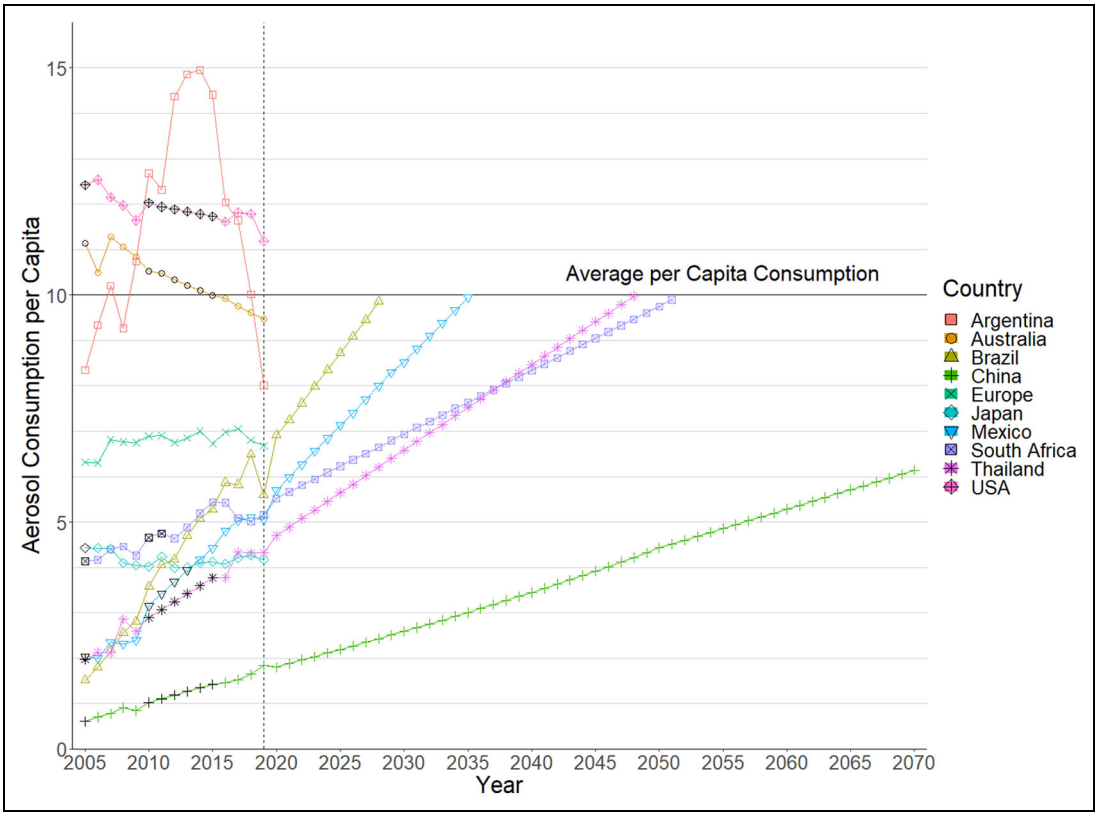


Figure 7. Aerosol production per capita. Points in black have been estimated using the same linear regression as to predict future production figures. Unit data are taken from **Figure 6** and population data used to create per capita values from the World Bank (n.d.). A typical high-income consumption rate of 10 unit per person per year is marked with the black solid line. DOI: https://doi.org/10.1525/elementa.2020.20.00177.f7

annual consumption and will not increase consumption on the 2020–2050 time frame. These last two assumptions mean our estimates of both current and future global use are potentially conservative.

Having classed each country by income level and having then assigned that country to an aerosol projection pathway, we then estimate absolute consumption by correcting for future population. Population projections taken from the World Bank (n.d.) have been combined with the production trends from **Figure 6** and are presented in **Figure 8** in units of aerosol consumption on the left-hand *y*-axis. We must assume that countries will remain in their GNI group and follow that aerosol trend over the next 30 years. Import and export must be taken into consideration as not all countries consuming aerosols will be producing them themselves, nor will high-producing countries consume all that they make. As such, the production value for Argentina has, again, been decreased by 45% and the United States and Europe by 10% to account for export. (Industry data suggest that the majority of European and United States production is consumed within those borders, hence the lower export value.) By focusing our calculations on aerosol usage per person, global estimates of VOC emissions are in essence unaffected by cross border trade, since all aerosols are at some point used by someone. One potential issue to account for would be the "banking" of remnant VOCs in used or partially used products in storage or sent for disposal. In general, aerosols are designed to dispense a large fraction of their contents, and if disposed of through recycling, when crushed, any remaining content would be released. For canisters sent to landfill (or stored very long term), it is possible that unreleased VOC would lead to an overestimate in our emissions in the short term (e.g., in year), but over a decadal timescale, those landfill units would ultimately degrade and leak out their contents.

The approach described in **Figures 3** and 4 has been used as a template to convert from unit fillings (which is the metric for industry reported data) to a propellant emission by mass. We have used the detailed UK inventory and manufacturer reporting data (1,567 million cans filled with an estimated 83 kt of propellant in 2018) to derive an average of 53 g per aerosol filling across an averaged profile of all aerosol product types. As we are interested in the filling of VOCs specifically (and not compressed air), this figure has been scaled to 49 g per aerosol filling to account for the combined 93% HAP and DME consumption as seen in **Figure 1**. This "propellant factor" has been applied to the global fillings data (left-hand axis of **Figure 8**) to give data as kilotons of propellant on the right-hand *y*-axis.

The uncertainties have been assessed for both the projections of population and aerosol use and emission values. The UN Population Division provides data on uncertainties (United Nations, Department of Economic and Social Affairs, Population Division, 2019) in the form of 95% prediction intervals, reported as World Bank

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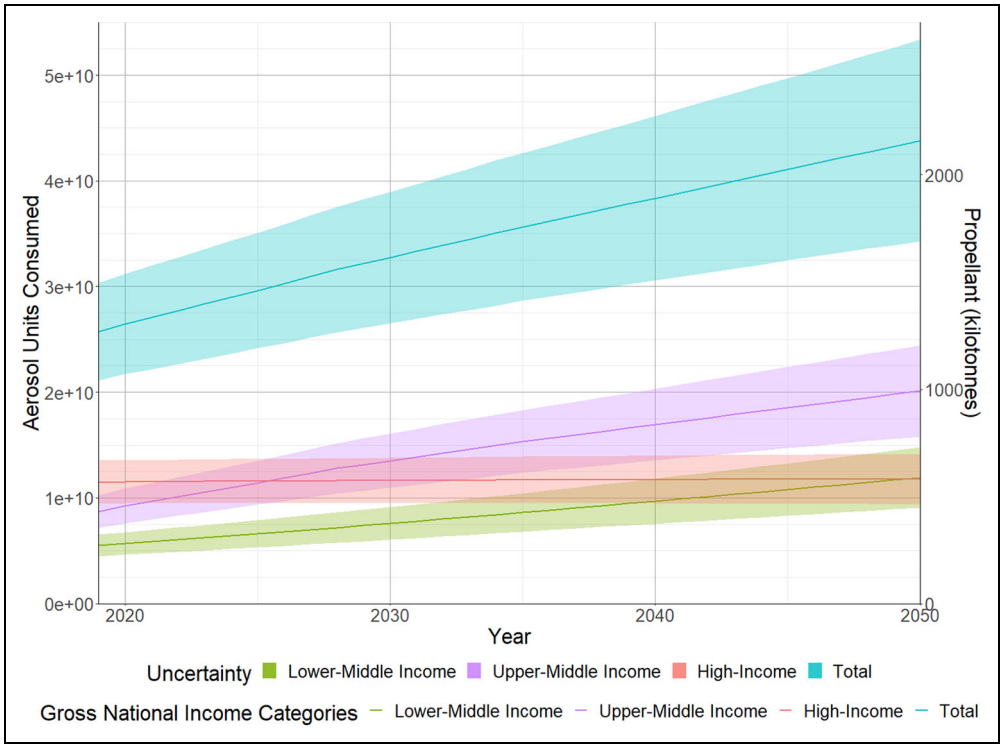


Figure 8. Estimated trends in global aerosol consumption. Expressed as units of aerosol cans consumed and converted to mass of VOC propellant based on the aerosol composition from **Figure 1**, the product templates shown in **Figure 3**, and growth curves extrapolated using recent trends seen in representative GNI groups where statistics were available. Shaded uncertainties incorporate population uncertainties and the range generated from a Monte Carlo simulation of possible per capita consumption rates and product distributions. VOC = volatile organic compound; GNI = gross national income. DOI: https://doi.org/10.1525/elementa.2020.20.00177.f8

income groups, which is converted to uncertainty using data for the "medium variant" population trajectory. As these data are only given in 5-year intervals, the intervening years were interpolated in a linear fashion. Percentage uncertainty in population growth is small for the high-income classification as census data are often more up-to-date and reliable and is unsurprisingly greater for the lower-middle income category. The prediction interval increases for all three income categories from 2020 to 2050 as population estimates become more uncertain the further into the future predictions are made.

There are many obvious uncertainties associated with the aerosol unit estimates and the conversion to VOC emissions. These are predominantly linked to the conversion of aerosol units into mass emissions (e.g., the process described in **Table 2**), and critically the likely final plateau usage in each country, since the use of aerosols (meaning which products and how many in total) in each country is a function of national habits and preferences. This does not follow any common variable like GDP, and notably the richest countries by per capita GDP do not necessarily have the highest aerosol usage. We assume that the plateau value varies around 10 ± 3 in an entirely random manner. The range here is based purely on the per capita values that have been reported in the past by individual high-income countries. We use a 10,000-step Monte Carlo simulation to evaluate a range of uncertainties in typical per capita usage in each country and the distribution of aerosol units between subproduct types. When combined with the uncertainties in the population projections, this inevitably leads to a spread in estimates, shown as the shaded bands on **Figure 8**.

Projecting per capita aerosol consumption across countries in the same GNI group if specific reporting data do not exist (and that is most countries) gives an estimate of current global VOC emissions from compressed aerosols of 1.3 ± 0.23 Tg year^{-1} for 2018. By applying projections of current usage trends into the future and including all countries in a GNI group, we estimate global aerosol production could reach approximately 4.4×10^{10} units year^{-1}, generating an emission of 2.2 ± 0.48 Tg year^{-1}, an increase of around 70% in VOC propellant emissions by 2050 from present day.

Estimates of the current global anthropogenic emissions of VOCs from *all* anthropogenic sources are also rather uncertain, but some recent estimates place total emissions in the range 98–156 Tg year^{-1}. Set against that global value (for 2013, taken from IPCC estimates), aerosol propellant VOCs currently represent around 1% of global anthropogenic emissions. If anthropogenic emissions reductions in middle-income countries follow those of high-income countries, with reducing emissions from gasoline vehicles and related fuel evaporation, then the fractional contribution to global emissions is likely to rise further, potentially approaching a value similar to that seen in a typical high-income country like the UK where propellants represent 6% of national VOC emissions.

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8. Conclusions

The annual per person consumption of aerosols has broadly stabilized in most high-income countries; however, there is evidence for a rapid rise in consumption in middle- and low-income countries. On current trends, it appears reasonable to assume that as economies and wealth grow that consumption patterns may converge on the historically stable figure of approximately 10 aerosol units per person per year, a value derived from past reporting in high-income countries. Accounting for the distribution of different aerosol products used allows for consumption statistics to be translated into amounts of propellant released, where that propellant is dominated by VOCs, a combination of simple hydrocarbons and DME. Not all countries report their aerosol use, but there is sufficient information across representative income levels to make some informed estimates of consumption in each of the four World Bank GNI categories. Based on this and projected growth in population in each country, some global estimates of aerosol use have been made. We estimate that globally around 1.3 $\pm$ 0.23 Tg of VOC propellant is currently released each year in the form of hydrocarbons and DME. The central value is lower than that estimated in Nourian et al. (2021; 1,437.8 kt, no uncertainties given, estimate based on a market report of the number of aerosol valves sold in 1 year). We note that our calculation has taken aerosol can size, fill pressure, and percentage of propellant into account. The method used here also makes use of a wider and more detailed breakdown of product consumption for annual global estimates. Assuming patterns of use continue forward on the trajectories seen in the recent past for middle- and low-income countries, then global VOC emissions from aerosols may reach around 2.2 $\pm$ 0.48 Tg per year in 2050.

To assess the scale of downstream impact of aerosol VOC emissions would need a complex and comprehensive modeling study since the effects would be dependent on the wider local and regional pollution conditions, geography, season, and so on. This is well beyond what we can include in this article. However, to give a scale of effect on ozone formation, we use a simple box model (MCM 3.1, www.mcm.york.ac.uk) run over a 3-day period and constrained to VOC, NO_x, and other supporting ambient data from the 2012 Clearflo air pollution research project in central London 2012 (Whalley et al., 2018). We use a baseline model initialized using the observed average ambient VOC, CO, HCHO, and NO_x concentrations, followed by a second counterfactual where we reduce the amount of ambient propane, *n*-butane, and *iso*-butane by the proportions reported as originating from aerosol propellants in the NAEI (Office for National Statistics UK, 2019). For reference, this uses conditions of NO: 3 ppb and NO_2: 10 ppb, along with a full range of VOCs (Top 10 were ethanol: 10.7 ppb, ethane: 8.3 ppb, acetone: 8.1 ppb, methanol: 6.8 ppb, *n*-butane: 5.2 ppb, propane: 4.9 ppb, *iso*-butane: 2.6 ppb, *iso*-pentane: 3.5 ppb, toluene: 2.6 ppb, and butanol: 2.5 ppb). For the counterfactual, 48% of *n*-butane emissions in the UK were estimated to be from an aerosol source, so this model was run with *n*-butane reduced from 5.2 ppb to 2.6 ppb. 14% of propane emissions were estimated to be from aerosol sources and so on. Over a 3-day UK summertime photochemical trajectory reducing the initializing ambient VOCs concentrations by the proportion accounted for by aerosol emissions in the NAEI resulted in a decrease in ozone of around 2.2–2.8 ppb after 72 h. We would suggest that in many other locations, the replacement of aerosol propellant with non-VOC alternatives would also lead to potentially meaningful reductions in surface ozone when measured over multiday timescales.

Although at present aerosol VOC propellants make up approximately 1% of global anthropogenic VOC emissions, their contribution as a fraction of emissions appears likely to rise. Substantial reductions in VOC emissions from road transport, gasoline vehicles, and evaporative losses have been reported in many high-income countries, and it seems likely that this will ultimately propagate through to middle- and low-income countries over time, particularly should transport fleet electrification become widespread by 2050. Although projections of VOCs in the future at a global scale are uncertain, in some more ambitious air quality and emission scenarios, for example, presented by Amann et al. (2020), global emissions of VOCs from all anthropogenic sources could decline to approximately 37.9 Tg $year^{-1}$ in 2040. If aerosols consumption follows the patterns shown here and the propellant remains as of today, then they would represent approximately 6% of all global VOC emissions—a value consistent with the current day UK contribution. We note that there are currently few downward pressures on the emissions of VOCs from aerosols specifically, indeed, there is some evidence that aerosolization is being applied to products that were previously dispensed as liquids—for example, suntan lotions and moisturisers.

Policy implications

The replacement of halocarbons with hydrocarbons in the 1987 Montreal Protocol was clearly a landmark environmental change. There may however be a case that the subsequent global growth in aerosol consumption was not foreseen at the time of the signing of the Protocol, when aerosol usage was lower per capita in high-income countries than today (roughly 50% of current use), and usage was very low in middle- and low-income countries such as China and India. Given that VOCs contribute to tropospheric ozone pollution, international policy revision may be required and the continued support of VOCs as the preferred replacement for halocarbons potentially not sustainable for aerosol products longer term. Although there are a few notable exceptions, such as the California Air Resources Board product regulations, the general absence of controls on aerosol formulation or consumption appears in tension with the often highly regulated nature of VOC emissions from other industry sectors. Road transport (both evaporative and tailpipe), buildings materials (e.g., timber, furniture), and decorative products (e.g., paints and vanishes) are all subject to specific emissions regulation in many countries. The cost–benefit of

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implementing new technologies to further reduce emissions of VOCs from gasoline vehicles may be disproportionately poor when compared to the equivalent air quality gains from VOC reduction that might be achieved more straightforwardly by a lowering of consumption of aerosols or the replacement of VOCs with less harmful compressed air/N_2 as the propellant.

Although this article is not intended to provide policy prescriptive solutions, we would stress the need for much improved collection of statistics on annual aerosol consumption by product type and by country. Without robust data of this kind, the full impacts of the Montreal Protocol remain uncertain to calculate as are the possible future benefits of replacement of hydrocarbons with alternatives. We are not experts in the manufacture of consumer products but would note that many technological options exist for the reduction of aerosol VOC emissions. As identified earlier, for some products, the use of compressed air or N_2 may be a viable alternative propellant. Perhaps more significantly for very many personal care and household cleaning products, a clear solution would be product deaerosolization. Many consumer products can be (and are) applied in their liquid or solid forms, for example, as roll-on deodorant, hair gel, solid furniture polish, bronzing lotion, room fragrance, to name but a few. In some cases, the continued use of aerosols when nonaerosol alternatives exist is simply down to the continuation of past consumer preferences and habits.

More generally the role played by aerosol VOC emissions in air pollution needs to be much more clearly articulated in messaging on air pollution and its management to the public. The association of VOC emissions with gasoline and vehicles is heavily entrenched, and even among air quality professionals, there is limited knowledge of the scale of aerosol impact. Approaches to emissions reduction from PCPs could potentially be communications-led, with individuals encouraged to switch to nonaerosol alternatives or moderate consumption. Product labeling of consumer products as high VOC emitting—and clearly linking this to poor air quality—may drive change away from aerosols to their alternatives as has been seen previously with the labeling of paints and varnishes. Although behavioral change appears to have considerable potential to reduce emissions, other more direct interventions could also be envisaged. Fiscal approaches such as variable taxation on aerosol products would be a more drastic measure for effecting change, as would regulatory phase out and banning of aerosols products containing HAPs or DME.

Data accessibility statement

Data used in this study are made publicly available via the Centre for Environmental Data and Analysis (www.ceda.ac.uk), an enduring research data repository. It is also mirrored as a data set on the University of York data repository.

Supplemental files

The supplemental files for this article can be found as follows:

Figure S1. European aerosol production breakdown. For the years 2016–2018, using data provided by the European Aerosol Federation (FEA). File Type: PNG Document (.png).

Table S1. Economic classifications according to the World Bank. Data determined using the World Bank Atlas method. File type: Microsoft Word (.doc).

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Competing interests

ACL is an associate editor of Elementa. He was not involved in any way in the peer review of the article.

Author contributions

Contributed to conception and design: AMY, ACL

Contributed to acquisition of data: AMY, ACL

Contributed to analysis and interpretation of data: AMY, ACL

Drafted and/or revised the article: AMY, ACL

Approved the submitted version for publication: AMY, ACL

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