Elementa: Science of the Anthropocene

Global emissions of VOCs from compressed aerosol products.

--Manuscript Draft--

Manuscript Number:	ELEMENTA-D-20-00177R2
Full Title:	Global emissions of VOCs from compressed aerosol products.
Short Title:	Global emissions of VOCs from compressed aerosol products.
Article Type:	Policy Bridge
Section/Category:	Atmospheric Science Domain
Manuscript Classifications:	Atmospheric Science; Human Impacts on the Atmosphere; Air Quality
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Manuscript Region of Origin:	UNITED KINGDOM
Suggested Reviewers:	
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Global emissions of VOCs from compressed aerosol products.

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Abstract

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 ~10 ± 3 units person^{-1} yr^{-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 e.g., 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 ~ 1.3 ± 0.23 Tg yr^{-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 all VOC emissions were from aerosols in 2017, more than was 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 ~ 4.4 ± 0.96 x 10^{10} units yr^{-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 ~ 2.2 ± 0.48 Tg yr^{-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 hairspray, 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 (European Aerosol Federation (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 co-solvent. The propellant can be a liquefied or compressed gas and can comprise anywhere from 5% - 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 powder depending on the dispersing ability of the propellant and the force with which they are dispensed (European Aerosol Federation (FEA), n.d.).

Initially chlorofluorocarbons (CFCs) were used as aerosol propellants; their non-flammable and odourless properties and chemical stability made them ideal for use in consumer products. However, concerns associated with their environmental impact were catalysed by Molina and Rowland's 1974 paper detailing the possible destruction of ozone arising from halogens released through CFC photodissociation in the stratosphere (Molina and Rowland, 1974). The CFCs used in aerosols proved to be extremely potent ozone depleting substances, as well as having high global warming potentials between 1,100 to 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 timeframe 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 life-time 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.

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 USA and Sweden (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 vapour pressure — a key factor controlling aerosol performance. Generally, a blend of propane and *n*-butane will have a lower vapour 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 personal care products (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 odourless propellant, for which dimethyl ether (DME) is the most common solvent used. Another alternative is compressed gas (for example 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 vapour pressure and purity required (although cost may be an influencing factor). The higher the vapour pressure, the higher the degree of dispersion from the aerosol can, and the finer and drier the product mist. High vapour pressure products include air freshener, flying insect spray, and spray paint, whereas lower vapour pressure products include shaving cream, gels and mousse, and perfume (DG Enterprise and Industry, 2014). A medium vapour pressure product, for example hairspray, 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 vapour pressure products having a low ratio of propellant to active ingredient. These are important factors considered in the following section on emissions (see Table 3).

Table 1: Compressed aerosol propellant properties

	Propellant Properties			
Propellant Type	**Flammable?**	**Pressure**	**Purity***	**Co-Solvent Required?**
Hydrocarbons	Yes	High	Varies	Yes
DME	Yes	High	High	Rarely
Compressed Gas	No	Low	Varies	Yes

*Odourless

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

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 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 HAPs and DME make up 93% of emissions, which we describe collectively henceforth as `VOC emissions`, labelled 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 ~1-6% of VOC in most countries), in the remainder of this paper 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 global warming potential (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 either of the other propellants as a consequence of the Kigali Amendment. In terms of their GWP, VOCs are significantly more environmentally acceptable (e.g., DME compared to HFCs in Good et al. 1998) and whilst they do not have a large direct effect on radiative forcing, they do undergo photo-oxidation that generates tropospheric ozone, itself a greenhouse gas. As for the compressed gas propellants, carbon dioxide (CO_2) 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 pressurised cans are the direct health risks which 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 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). Whilst 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 (EU), or World Health Organisation (WHO) 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 organisations/agencies, as presented in Table 2.

Table 2: Recommended exposure limits

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]

TLV: Threshold limit values, AEGL: Acute exposure guideline levels (Over 1 hour), STEL: Short-term exposure limit, TWA: Time weighted average (over 8 hours)

[a] (BOC, 2005)

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

[c] (Health and Safety Executive, 2020)

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 non-methane hydrocarbons in the atmosphere, with atmospheric lifetimes of ~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 are 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 vapour pressure product will have a high mass dispense rate and will therefore emit more propellant and smaller particles over a short period of time (European Aerosol Federation (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 (European Aerosol Federation (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 recognised 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, hairspray).

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 industrialised 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 onwards, 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 ~ 70% of the propellant of a high-pressure product, and a low-pressure product ~ 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, ~ 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.

Table 3: Simplified aerosol product volume and pressure assumptions

Product	Average Size (mL)	Pressure
Air fresheners	300	High
Automotive	500	Medium
Colognes/Perfumes	75	Low
Deos/Body sprays/APS	250	Medium
Hairspray products	300	Medium
Hard surface cleaners	750	Low

Industrial	300	High
Insecticides	300	Medium
Medical (excl 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)

The upwards 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 are in decline, such as hairspray, whereas others show increasing trends, such as aerosol deodorants and air fresheners. Overall personal care products (PCPs), and in particular deodorants, body sprays, antiperspirants and hairspray, 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 kilotonnes of propellant, overwhelmingly as VOCs in the form of either simple hydrocarbons or DME, is estimated to be 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 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). Whilst 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 three 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 Impact Assessment Study on the Adaptation to Technical Progress of the Aerosol Dispensers Directive, Figure 2-2 (DG Enterprise and Industry, 2014).

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 industrialised countries, dominated by emissions from road transport, fossil fuels and the associated extractive and refining industries. For example, see a recent analysis of multi-year sectoral VOC emissions trends in Lewis et al., 2020 for the UK. Whilst 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 onwards focused predominately on sources such as gasoline vehicle exhaust and fugitive emissions, and significant reductions occurred in many industrialised 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 USA 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 downwards trends in estimated emissions are also reflected in ambient data (e.g., Dollard et al., 2007 and Lewis et al., 2020). The UK National Atmospheric Emissions Inventory, from which the data in Figure 5 is 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 behaviour 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. Whilst 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 (~60 kt pa) were greater than the total VOC emissions arising from all passenger cars in the UK (estimated as ~30 kt pa in 2017 (Office for National Statistics UK, 2019)).

7. Global consumption and future projections

Industry reported data on aerosol products is 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 USA, 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 one 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, does not exist.

Expressed per capita, there are notable absolute differences between countries, the USA being the highest per capita user of aerosols. The average annual per capita production from high-income countries is ~10 ± 3 aerosol units person^{-1} yr^{-1}. The per capita use data for Argentina has been calculated as 55% of production, as it has been reported 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, 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} yr^{-1} value. Figure 7 is annotated with a line marking this high-income limit. We predict the year in which this will occur assuming that future rate of production follows recent past trends. This per capita forecast data has been calculated using population predictions and is corrected for the expected large population growth in some emerging economies. First to reach 10 units person^{-1} yr^{-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 timeframe. 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} yr^{-1} plateau, production and consumption remains 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 categorised 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). 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 ± 3. The current average annual units per person for upper-middle income countries is estimated as 5 ± 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 extrapolation of past trends, that they will reach the maximum of 10 units per person per year ~ 2050. Lower-middle income countries have been estimated to currently consume 2 ± 1 units per person annually, and will reach 3 ± 1 by 2050. Low-income countries are assumed to currently have no annual consumption, and will not increase consumption on the 2020-2050 timeframe. 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 (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 USA and Europe by 10% to account for export. (Industry data suggests that the majority of European and USA 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 over-estimate 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 kilotonnes 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 kilotonnes 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 United Nations 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 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 3), 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 sub-product 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 does not exist (and that is most countries), gives an estimate of current global VOC emissions from compressed aerosols of 1.3 ± 0.23 Tg yr^{-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 ~ 4.4×10^{10} units yr^{-1}, generating an emission of 2.2 ± 0.48 Tg yr^{-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 yr^{-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.

8. Conclusions

The annual per person consumption of aerosols has broadly stabilised 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 ~ 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 ± 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 (1437.8 kt, no uncertainties given, estimate based on a market report of the number of aerosol valves sold in one year). We note that our calculation has taken aerosol can size, fill pressure and percentage 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 ± 0.48 Tg per year in 2050.

To assess the scale of downstream impact of aerosol VOC emissions would need a complex and comprehensive modelling 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 paper. 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 three-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 initialised 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, 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, 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 three-day UK summertime photochemical trajectory reducing the initialising 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 hours. 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 multi-day timescales.

Whilst at present aerosol VOC propellants make up ~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. Whilst 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 ~37.9 Tg yr^{-1} in 2040. If aerosols consumption follows the patterns shown here and the propellant remain as of today, then they would represent ~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 aerosolisation 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 the contribution of VOCs 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. Whilst 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 appear 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 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.

Whilst this paper 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 de-aerosolization. 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 non-aerosol alternatives exist is simply down to 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 amongst air quality professionals there is limited knowledge of the scale of aerosol impact. Approaches to emissions reduction from personal care products could potentially be communications-led, with individuals encouraged to switch to non-aerosol alternatives, or moderate consumption. Product labelling 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 labelling of paints and varnishes. Whilst behavourial 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.

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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

Acknowledgements.

AMY is in receipt of funding for a PhD from the Natural Environment Research Council, and ACL acknowledges their support for the NCAS National Capability underpinning programme. The authors are grateful to Prof. Lucy Carpenter and Dr Pete Edwards for helpful discussions and advice that have informed this paper.

Funding Information

Natural Environment Research Council. Grant Number: NE/M021513/1

Competing Interests

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

Supplementary Information

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.

[a] Included within the grouping of Europe

[b] Included in China's total

File type: Microsoft Word (.doc)

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 dataset on the University of York data repository.

Figure Titles and Legends

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 *Pentane **Propane ***Butane. HAPs and DME have been combined as total VOC.

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 (FEA), 2013). The grey circles are sized to be proportional to the amount of aerosol propellant release per usage.

Figure 3: **United Kingdom aerosol filling statistics by product class for the period 1965-2019.**

(British Aerosol Manufacturers' Association (BAMA), data aggregated and combined from reports in 2019, 2018, 2015)

Figure 4**: Estimated United Kingdom aerosol emissions in kilotonnes 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, pressure) from Table 3.

Figure 5: **Total annual UK anthropogenic VOC emissions (excl. 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 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

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 (FEA), 2020, 2019, 2018, 2017; Spray TM, 2017a, 2017b, 2016a, 2016b, 2015).

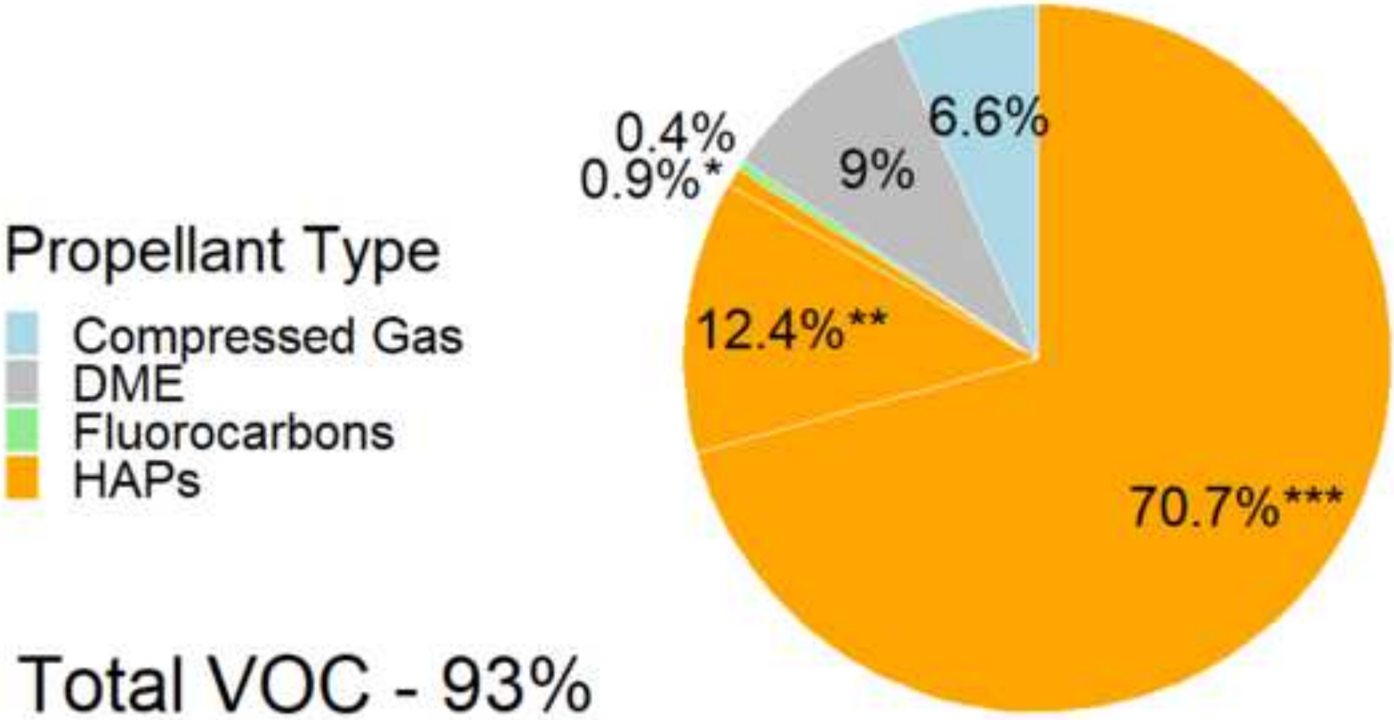
Figure 7: **Aerosol production per capita.**

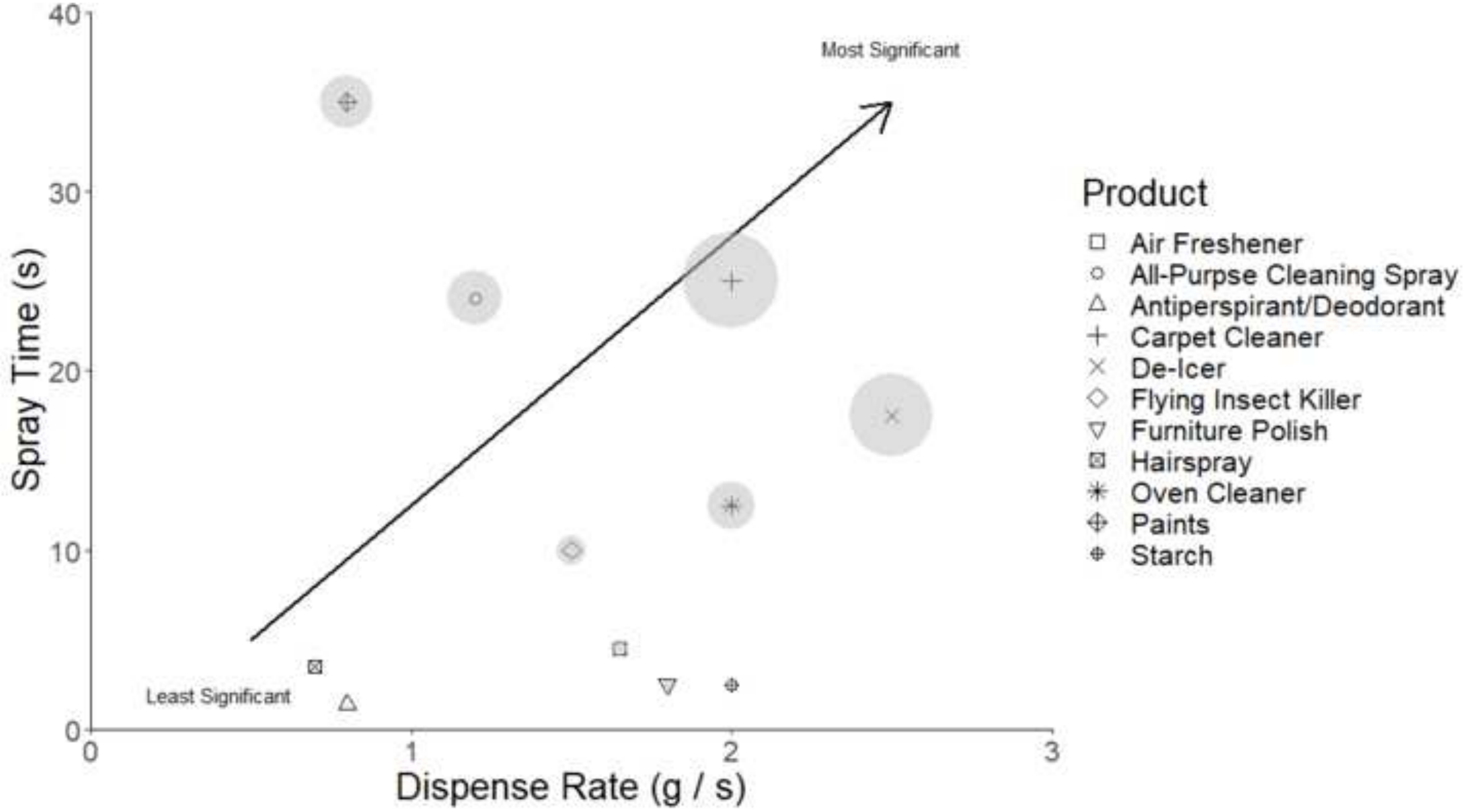
Points in black have been estimated using the same linear regression as to predict future production figures. Unit data are taken from Figure 6 and population data used to create per capita values from The World Bank (The World Bank, n.d.). A typical high-income consumption rate of 10 unit per person per year is marked with the black solid line.

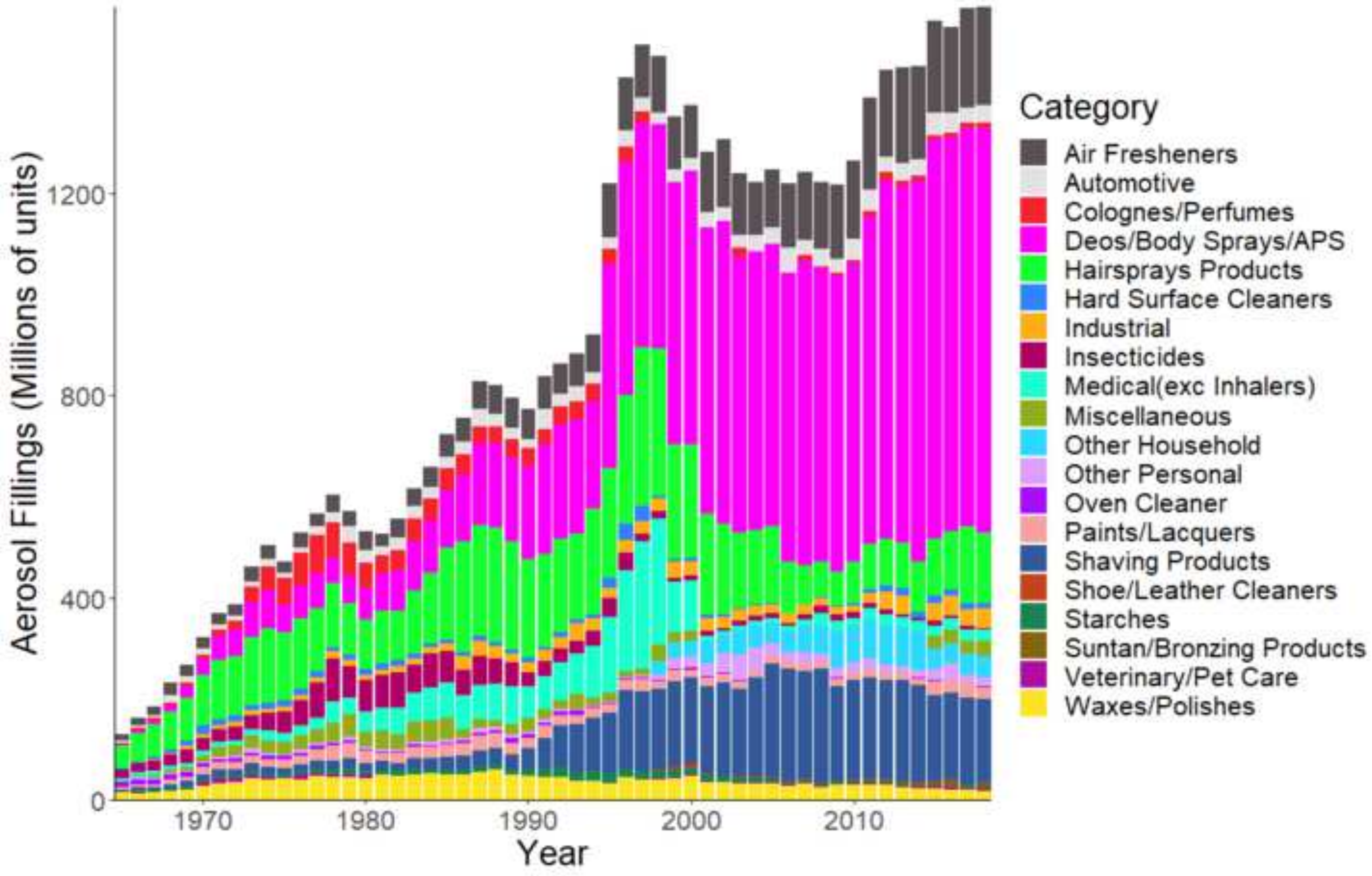
Figure 8: **Estimated trends in global aerosol consumption**

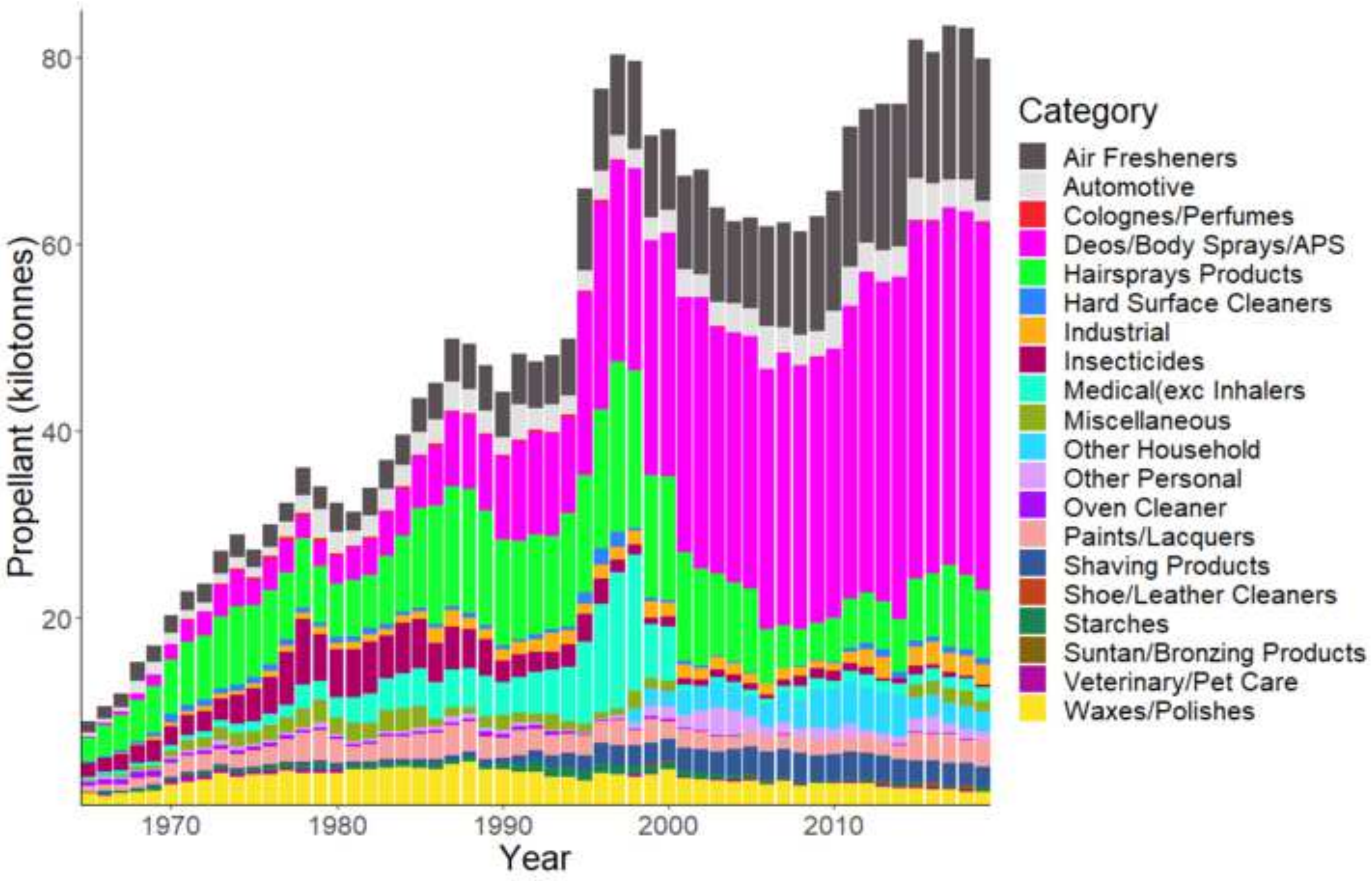
Expressed as units of aerosol cans consumed, and converted to mass of VOC propellant, based on the aerosol composition from Figure 1, the product templates shown in Figure 3, and growth curves extrapolated using recent trends seen in representative GNI groups where statistics were available. Shaded uncertainties incorporate population uncertainties and the range generated from a Monte Carlo simulation of possible per capita consumption rates and product distributions.

Figure 1


0.4%
0.9%*
9%
6.6%
12.4%**
70.7%***
Propellant Type
Compressed Gas
DME
Fluorocarbons
HAPs
Total VOC - 93%


Most Significant
Least Significant
Spray Time (s)
40
30
20
10
0
0
1
2
3
Dispense Rate (g / s)
Product
□ Air Freshener
○ All-Purpse Cleaning Spray
△ Antiperspirant/Deodorant
+ Carpet Cleaner
× De-Icer
◇ Flying Insect Killer
▽ Furniture Polish
⊠ Hairspray
✳ Oven Cleaner
⊕ Paints
⊕ Starch


Aerosol Fillings (Millions of units)
1200
800
400
0
1970
1980
1990
2000
2010
Year
Category
Air Fresheners
Automotive
Colognes/Perfumes
Deos/Body Sprays/APS
Hairsprays Products
Hard Surface Cleaners
Industrial
Insecticides
Medical(exc Inhalers)
Miscellaneous
Other Household
Other Personal
Oven Cleaner
Paints/Lacquers
Shaving Products
Shoe/Leather Cleaners
Starches
Suntan/Bronzing Products
Veterinary/Pet Care
Waxes/Polishes


Propellant (kilotonnes)
0
20
40
60
80
1970
1980
1990
2000
2010
Year
Category
Air Fresheners
Automotive
Colognes/Perfumes
Deos/Body Sprays/APS
Hairsprays Products
Hard Surface Cleaners
Industrial
Insecticides
Medical(exc Inhalers
Miscellaneous
Other Household
Other Personal
Oven Cleaner
Paints/Lacquers
Shaving Products
Shoe/Leather Cleaners
Starches
Suntan/Bronzing Products
Veterinary/Pet Care
Waxes/Polishes

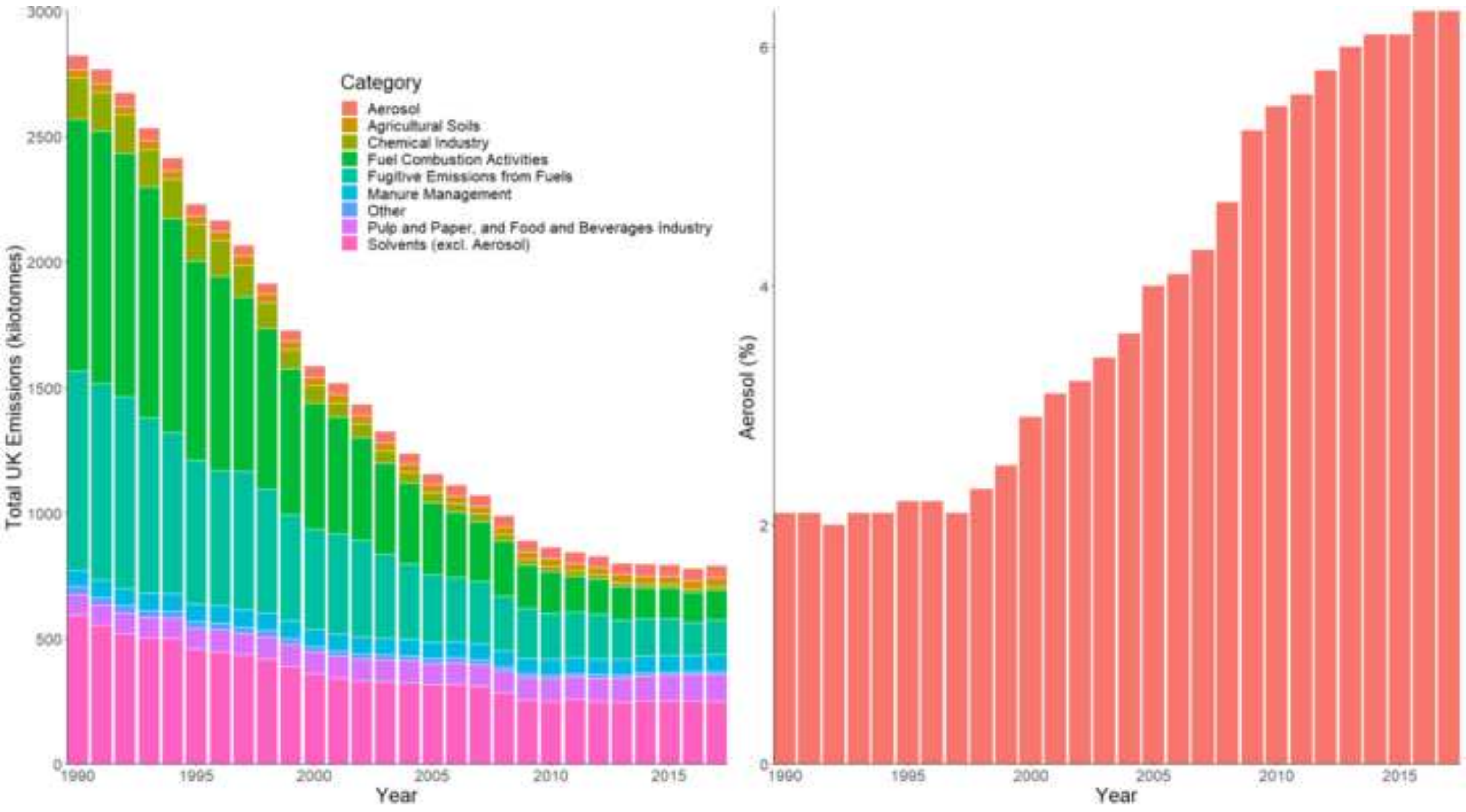
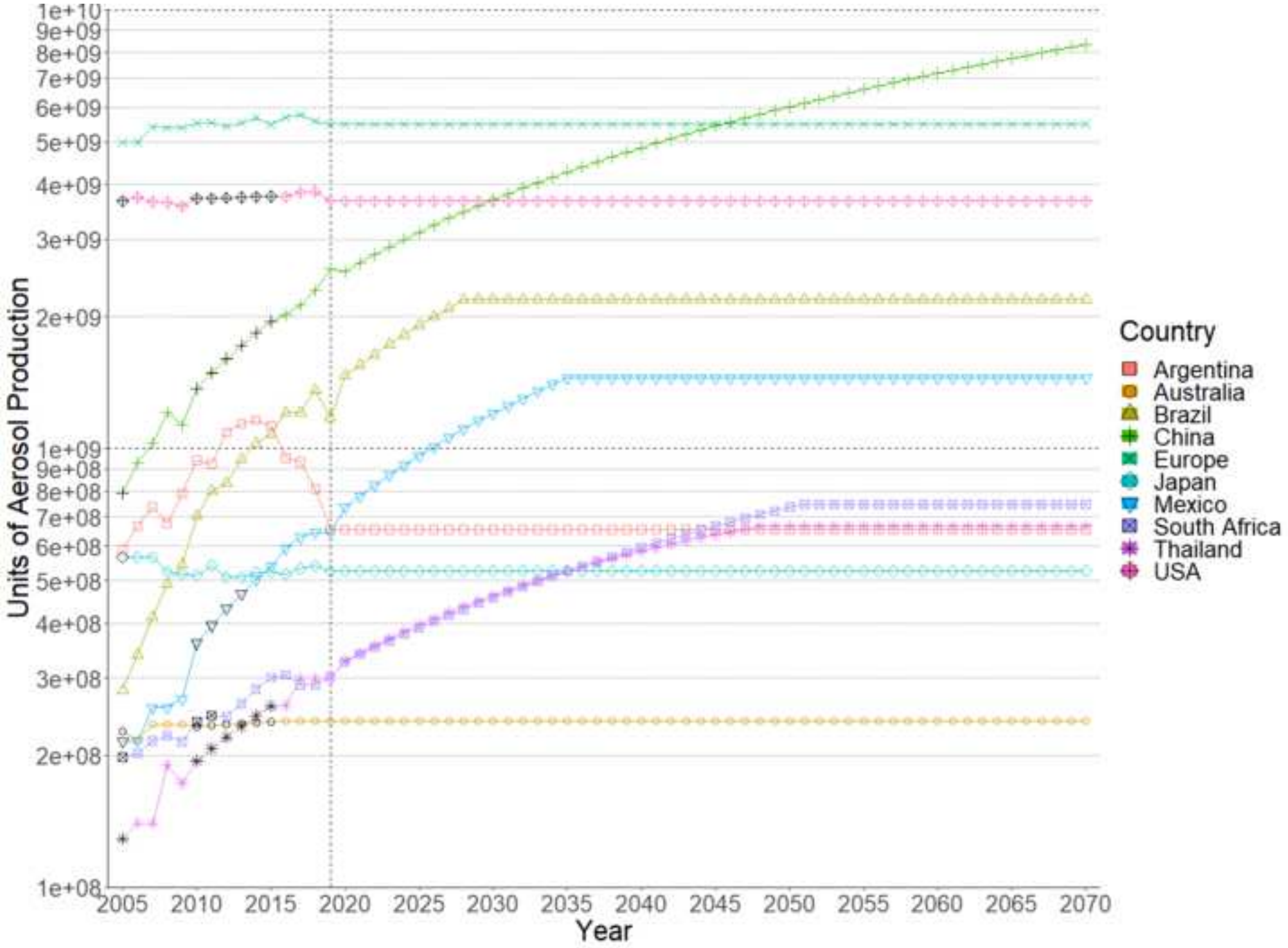

Total UK Emissions (kilotonnes)
3000
2500
2000
1500
1000
500
0
1990
1995
2000
2005
2010
2015
Year
Category
Aerosol
Agricultural Soils
Chemical Industry
Fuel Combustion Activities
Fugitive Emissions from Fuels
Manure Management
Other
Pulp and Paper, and Food and Beverages Industry
Solvents (excl. Aerosol)
Aerosol (%)
6
4
2
0
1990
1995
2000
2005
2010
2015
Year

Figure 6


Units of Aerosol Production
1e+10
9e+09
8e+09
7e+09
6e+09
5e+09
4e+09
3e+09
2e+09
1e+09
9e+08
8e+08
7e+08
6e+08
5e+08
4e+08
3e+08
2e+08
1e+08
2005
2010
2015
2020
2025
2030
2035
2040
2045
2050
2055
2060
2065
2070
Year
Country
Argentina
Australia
Brazil
China
Europe
Japan
Mexico
South Africa
Thailand
USA

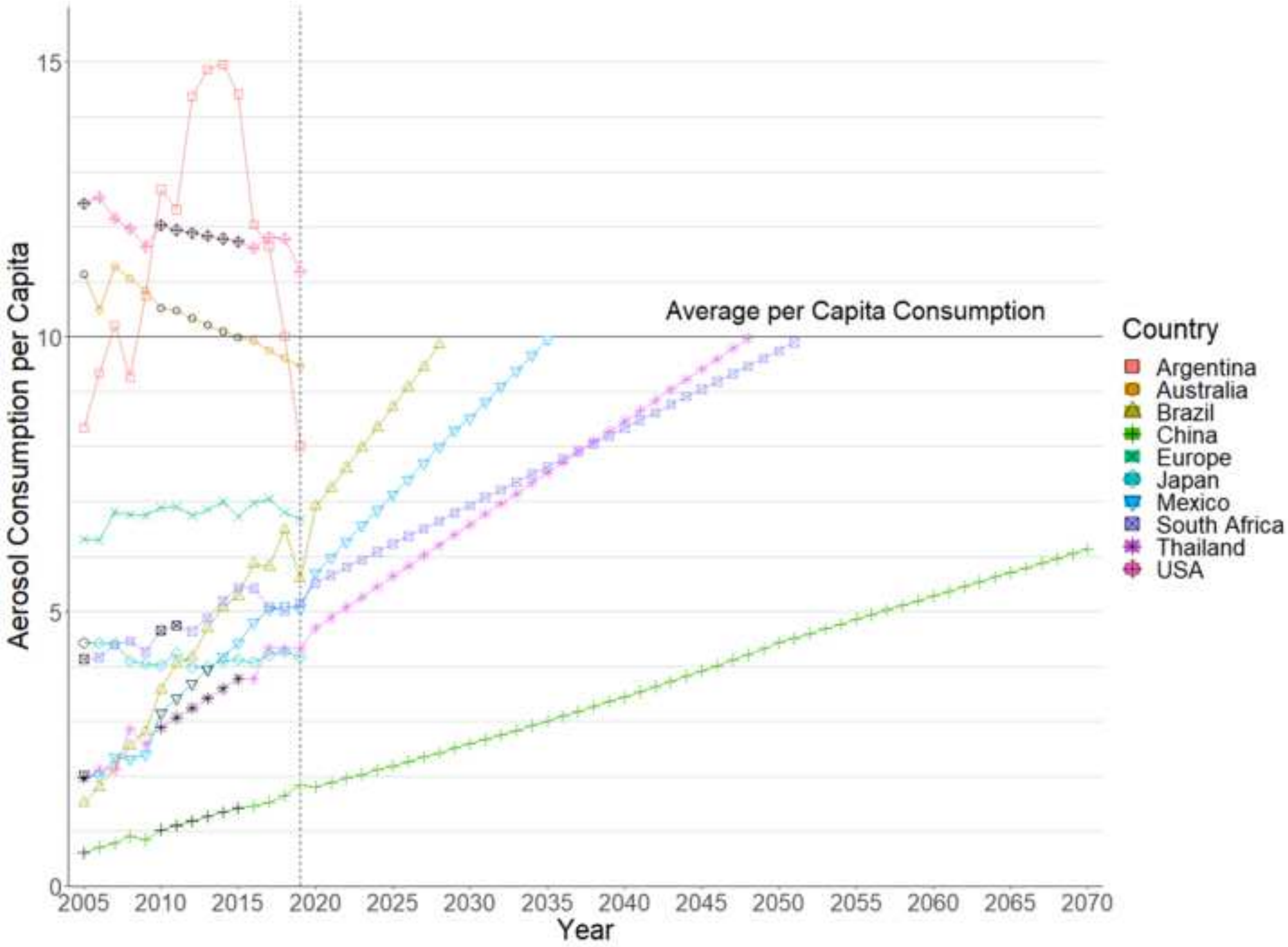
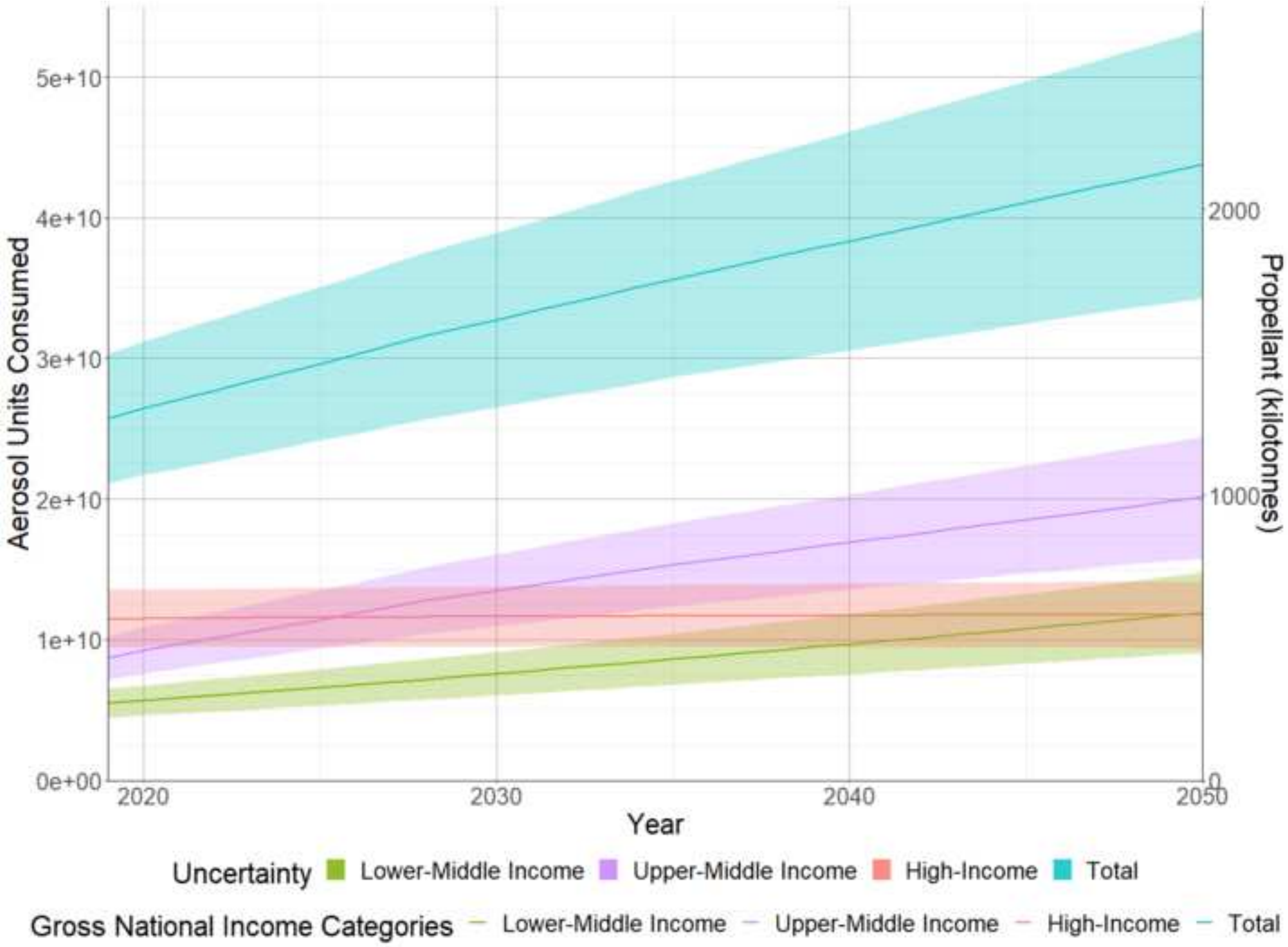

Aerosol Consumption per Capita
15
10
5
0
2005
2010
2015
2020
2025
2030
2035
2040
2045
2050
2055
2060
2065
2070
Year
Average per Capita Consumption
Country
Argentina
Australia
Brazil
China
Europe
Japan
Mexico
South Africa
Thailand
USA

Figure 8


Aerosol Units Consumed
5e+10
4e+10
3e+10
2e+10
1e+10
0e+00
Propellant (kilotonnes)
2000
1000
0
2020
2030
2040
2050
Year
Uncertainty
Lower-Middle Income
Upper-Middle Income
High-Income
Total
Gross National Income Categories
Lower-Middle Income
Upper-Middle Income
High-Income
Total