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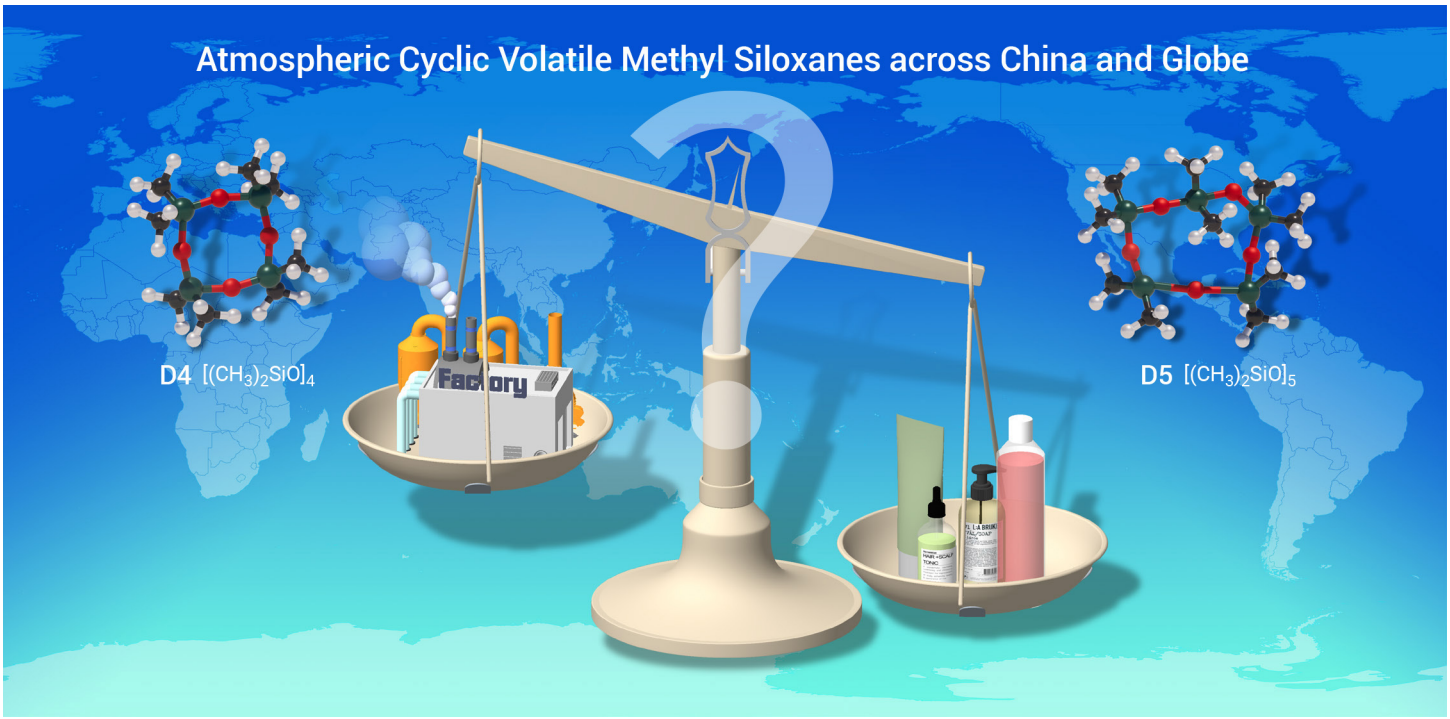
GRAPHICAL ABSTRACT



PUBLIC SUMMARY

- China is a global hotspot for atmospheric D4, a chemical primarily from industrial silicone production.
- D4 pollution stems mainly from industry, while D5 is linked to the use of personal care products.
- Higher personal income, not just population, better predicts D5 levels from consumer product use.
- To cut global D4 pollution, efforts must focus on industrial sources in major producing countries.

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Atmospheric occurrence and socioeconomic drivers of cyclic volatile methyl siloxanes across China

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Cyclic volatile methyl siloxanes (cVMS) are high-production volume chemicals, and their release into the atmosphere has raised increasing concerns regarding potential adverse effects on human health and the environment. China dominates global cVMS production, particularly octamethylcyclotetrasiloxane (D4) and decamethylcyclopentasiloxane (D5); but their atmospheric occurrence and socio-economic drivers are largely unknown. Here, we report initial findings from mobile and in situ measurements of atmospheric D4 and D5 across China using the Vocus proton-transfer-reaction time-of-flight mass spectrometer. We observed high concentrations of D4 (up to 139 pptv) and D5 (up to 176 pptv), induced by strong chemical factory emissions. The ambient mixing ratios of both chemicals increased from north to south and west to east, and D4 was one order of magnitude higher than D5. Moreover, we found that residential income better explained the domestic and global heterogeneity of cVMS compared to population density. These findings pave the way for exploring large-scale emission inventories and environmental impacts of atmospheric cVMS, highlighting China's substantial contribution to global atmospheric D4 levels primarily from industrial production and underscoring the need to consider regional economic factors in global D5 emission models.

INTRODUCTION

Silicones, also known as polysiloxanes, are composed of cyclic or linear backbone structures consisting of silicon (Si) atoms linked via oxygen (O) atoms. These structures have one or several side chains on each Si atom, which may form cross-links and influence the properties of the polymer. In the simplest silicone, poly(dimethylsiloxane), these organic side chains are merely methyl groups. Cyclic volatile methyl siloxanes (cVMS) are named according to the number of Si atoms. The main compounds, octamethylcyclotetrasiloxane (D4; $[(CH_3)_2SiO]_4$) and decamethylcyclopentasiloxane (D5; $[(CH_3)_2SiO]_5$), are frequently used together as a blended mixture called cyclomethicone, which may also include hexamethylcyclotrisiloxane (D3; $[(CH_3)_2SiO]_3$) as an impurity and the longer chain dodecamethylcyclohexasiloxane (D6; $C_{12}H_{36}O_6Si_6$).[1] cVMS possess a gamut of unique physical and chemical properties that confer upon them a myriad of consumer and industrial applications. They are commonly used as additives in cleaning and cosmetic products (e.g., shampoos, conditioners, hair care items, body lotions, and sunscreens) to provide specific features like ease of application, desirable sensory characteristics, and resistance to washing off.[2]

Due to their widespread use, cVMS have become high-production volume chemicals. D4 and D5 in this group share some similarities in their properties, such as high vapor pressure and low water solubility, and it is likely that more than 90% of the D4 and D5 consumed are emitted into the atmosphere.[3,4] However, there are some notable differences between these two compounds: D4 is considered as a persistent, bioaccumulative, and toxic substance, and measures have been taken to phase it out in personal care products,[5] whereas opinions on the environmental risks of D5 are divided.[1] Furthermore, while both D4 and D5 are used as raw materials in various industries, D5 is more commonly used in personal care products due to its lower viscosity and better spreading properties. Additionally, D4 and D5 are resistant to oxidation, reduction and photodegradation, but D4 (11 days) has a longer atmospheric lifetime than D5 (7 days) at an OH concentration of 10^6 molec. cm^{-3}.[6]

Extensive observations of atmospheric D4 and D5 have been conducted in Europe and North America,[7-11] with outdoor mixing ratios reaching as high as ~40 pptv (Table S1). However, the major methods employed in previous studies were offline collection and laboratory analysis, which are labor-intensive, time-consuming, and prone to artifacts.[12-14] These studies have demonstrated that urban atmospheric D5 concentrations are correlated with population density in urban areas, which can be used to distinguish personal care product emissions from other common urban emissions.[15-18] This is logical since environmental D4 and D5 in developed regions are primarily driven by the consumption rather than the production of silicones. China is the world's largest producer of silicones, accounting for 57% of the total global production with an output of 1.25 million metric tons. Meanwhile, China is a significant consumer of silicones, with medical and personal care products accounting for only 10.3% of the total consumption. Although some water bodies, indoor dust, aquatic animals, and soil sediments in China have been found to be seriously contaminated with cVMS,[19-26] the atmospheric concentrations and socioeconomic drivers of these pollutants are largely unknown.[27-30]

Here using a mobile laboratory equipped with the Vocus Proton-Transfer-Reaction Time-of-Flight Mass-Spectrometry (PTR-TOF-MS), we report on highly time-resolved and ultra-sensitive mobile measurements of atmospheric D4 and D5 concentrations over two exploratory pan-region transects in China. Moreover, in situ measurements of D4 and D5 were made at a ground site in Shanghai, the largest megacity in China. From these data, we investigate the distribution patterns and driving forces of atmospheric D4 and D5 across China.

MATERIALS AND METHODS

Field campaigns

The cVMS Observation Criss-Crossed China (COCC-China) project aimed to map atmospheric cVMS in China through three field campaigns (Figure 1). The first campaign occurred on 25 November, 2019, where we followed a north to south (N-S) route in northern China, starting from Dongying City in Shandong and traveling all the way south to the suburban area of Nanjing City in Jiangsu Province. During the N-S route, we primarily traveled through rural areas. The second campaign was conducted between November 29 and December 1, 2019, following a west to east (W-E) route in southern China. Our W-E route began in Xingyi, which serves as the capital of Qianxinan Prefecture in Guizhou, a relatively undeveloped province. We then traveled eastward, crossing almost the entire southern part of China until we reached Shaoxing in Zhejiang, which is considered one of the wealthiest provinces in the country. The third campaign took place in downtown Shanghai, specifically on the rooftop of Shanghai Academy of Environmental Sciences (SAES;[31,32] 121.4364 °E, 31.1759 °N; 18 m a.g.l.), between October 25 and November 23, 2021. This campaign was part of the air quality protection action for the 2021 China International Import Expo. With a population of 24.9 million and a land area of 6341 km^2, Shanghai is China's largest city with the most populous urban areas.

Mobile laboratory

The mobile measurement platform utilized in this study is the Vocus PTR-TOF-MS (TOFWERK AG, Thun, Switzerland) Mobile Laboratory, which is an IVECO van with a height of 3.8 m. The van's rear contains an air conditioning system that can be controlled from the driver cab and maintains a constant

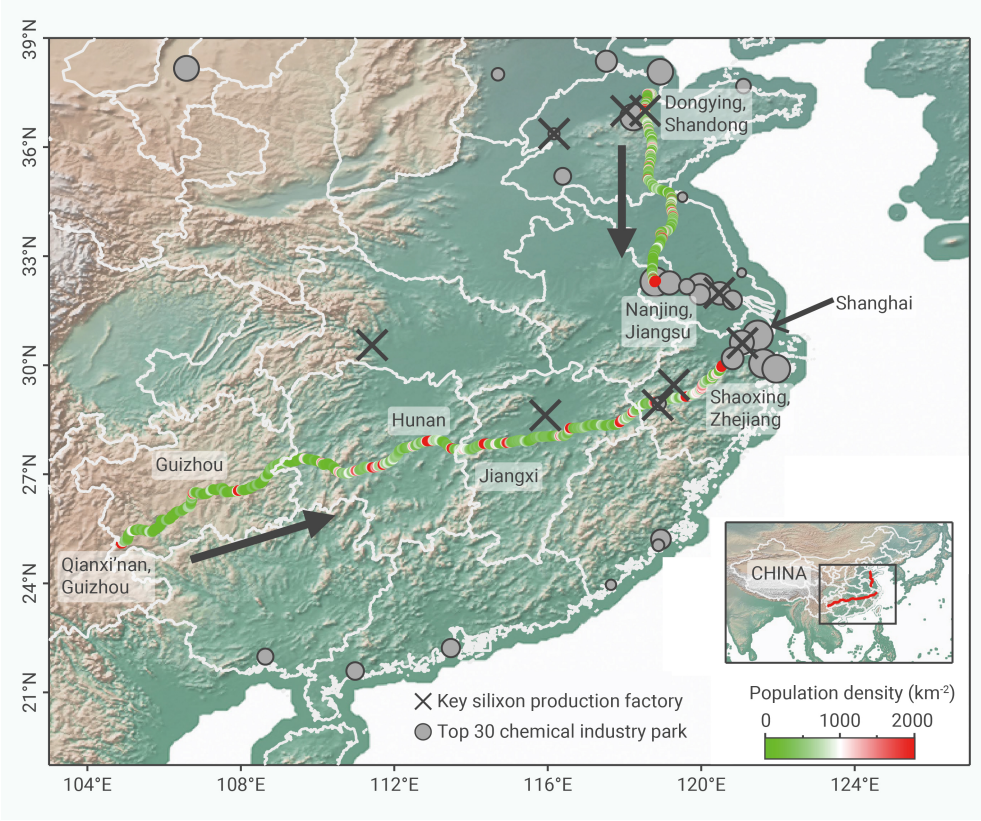

Figure 1. Locations of mobile and in-situ measurements.

temperature in the rear of the van, overcoming any instrumental heating. Here, D4 and D5 were determined as $C_8H_{24}Si_4O_4H^+$ (*m/z* 297.08244) and $C_{10}H_{30}O_5Si_5H^+$ (*m/z* 371.10123), respectively, with the Vocus PTR-TOF-MS utilizing H_3O^+ as the reagent ion. This instrument has been demonstrated to be a powerful tool for both field and laboratory deployments.[33,34] While the Vocus PTR-TOF-MS measured a wide range of other volatile organic compounds simultaneously, this study focuses specifically on the distribution and drivers of D4 and D5 due to their significance as high-production volume silicones with distinct environmental profiles and previously unknown large-scale patterns in China. Additionally, while advanced source apportionment techniques like Positive Matrix Factorization (PMF) are valuable, their application to mobile measurements across such large and heterogeneous areas is challenging due to the rapidly changing source environment and difficulties in obtaining consistent tracer profiles, and was therefore considered beyond the scope of this initial large-scale mapping study.

During mobile measurements, ambient air was drawn into a perfluoroalkoxy sampling tube (1/8" OD) mounted at the front of the van (0.5 m above the roof, i.e. 4.3 m a.g.l.) at a flow rate of ~2 L min^{-1}. Tests showed neglectable field blank and the wall loss of Teflon tubes had minimal effect (with relative deviations averaging less than 5%) on the quantification of viscous species (such as amines).[31,34] The instrument's acquisition rate was set at 1 second, and its responses to D4 (10344 cps $ppbv^{-1}$) and D5 (10079 cps $ppbv^{-1}$) were directly calibrated by the measurements of a certified gas-phase calibration mixture in ultra-pure nitrogen (D4: 500 ppb, D5: 484 ppb; Apel-Riemer Environmental, Inc., Miami, Florida) at RH 55%. The detection limits (3σ) for D4 and D5 were also determined as 1.60 pptv and 1.67 pptv, respectively. Regular zero air measurements were performed during the campaigns by overflowing the inlet with ultra-high purity nitrogen to monitor instrument background levels and ensure baseline stability. The instrument's mass calibration was checked daily using known ions present in ambient air. Full details of all instrument operation, calibration, and QA/QC for cVMS measurements are provided in the Supplemental Information Text S1.

RESULTS

Data overview

Figure 2 shows the log-scaled time series of atmospheric D4 and D5 concentrations measured during three field campaigns, with their statistical results summarized in Table 1. Mobile measurement results of D4 and D5 along the N-S and W-E routes are accompanied by population density (1 km^2), which is extracted from a 100-m-resolution population map (https://www.worldpop.org/). In-situ observations in Shanghai were conducted with simultaneous measurements of benzene. Notably, the original time resolution of D4 and D5 measurements, taken every second, were averaged to every minute and every hour for the mobile measurements and in-situ measurements, respectively. From a macro perspective, it is evident that D4 and D5 both gradually increase from north to south (Figure 2A) and from west to east (Figure 2B). In addition, taking all the data as a whole, Table 1 demonstrates that ambient concentrations of D4 and D5 measured along the W-E route in southern China are higher than those measured along the N-S route in northern China. Therefore, the distribution pattern of atmospheric D4 and D5 in China shows a decreasing gradient from the southeast towards other regions. While diurnal variations undoubtedly influence local concentrations (as seen in Figure 2C), the mobile measurements aimed to capture broad spatial trends across extensive regions. The observed consistent gradients from north-to-south and west-to-east, largely based on daytime measurements when traversing key areas, suggest that regional differences in emission strengths are the primary drivers of these large-scale patterns, overriding localized temporal variability for the purpose of this geographical analysis.

For the mobile measurements mainly conducted in non-urban areas, D4 was the dominant cVMS species, with much higher concentrations than D5. However, the gap between D4 and D5 could narrow or overlap in certain sections of mobile measurements, and in-situ observations in downtown Shanghai even have an opposite result, indicating that the factors influencing atmospheric abundance of D4 and D5 may vary by location. Upon closer inspection of the N-S route, the mean concentrations of D4 and D5 in the vicinity of the Nanjing Chemical Industrial Park (NCIP; highlighted in green in Figure 2A) were 20.0 pptv and 12.9 pptv, respectively, while in the non-industrial section they were much lower at only 11.9 pptv and 2.6 pptv, respectively. It is difficult to definitively separate industrial production from personal consumption or local traffic emissions based solely on mobile data and population density in these mixed environments. However, the magnitude of these directly measured concentration peaks observed when traversing known industrial areas like NCIP significantly exceeds typical ambient levels (e.g., Figure 2A, NCIP), unequivocally pointing to industrial activities as the primary driver of these major hotspots. Unlike the pulsed concentration peaks observed in the N-S route, D4 and D5 along the W-E route fluctuate relatively moderately, but their high concentrations are still concentrated in the highly industrialized Zhejiang section. Nevertheless, the population density in the NCIP area and Zhejiang section are also much higher, making it difficult to determine whether the high levels of D4 and D5 in southeastern China are caused by industrial production or personal consumption.

Factors influencing the variabilities of atmospheric D4 and D5

The observed atmospheric D4 and D5 must be influenced by both anthropogenic and natural factors. Undoubtedly, meteorological conditions could potentially influence ambient concentrations. However, obtaining comprehensive, route-specific, high-resolution meteorological data for quantitative analysis alongside mobile measurements presents considerable challenges. Our campaigns were conducted in late autumn/early winter, which could

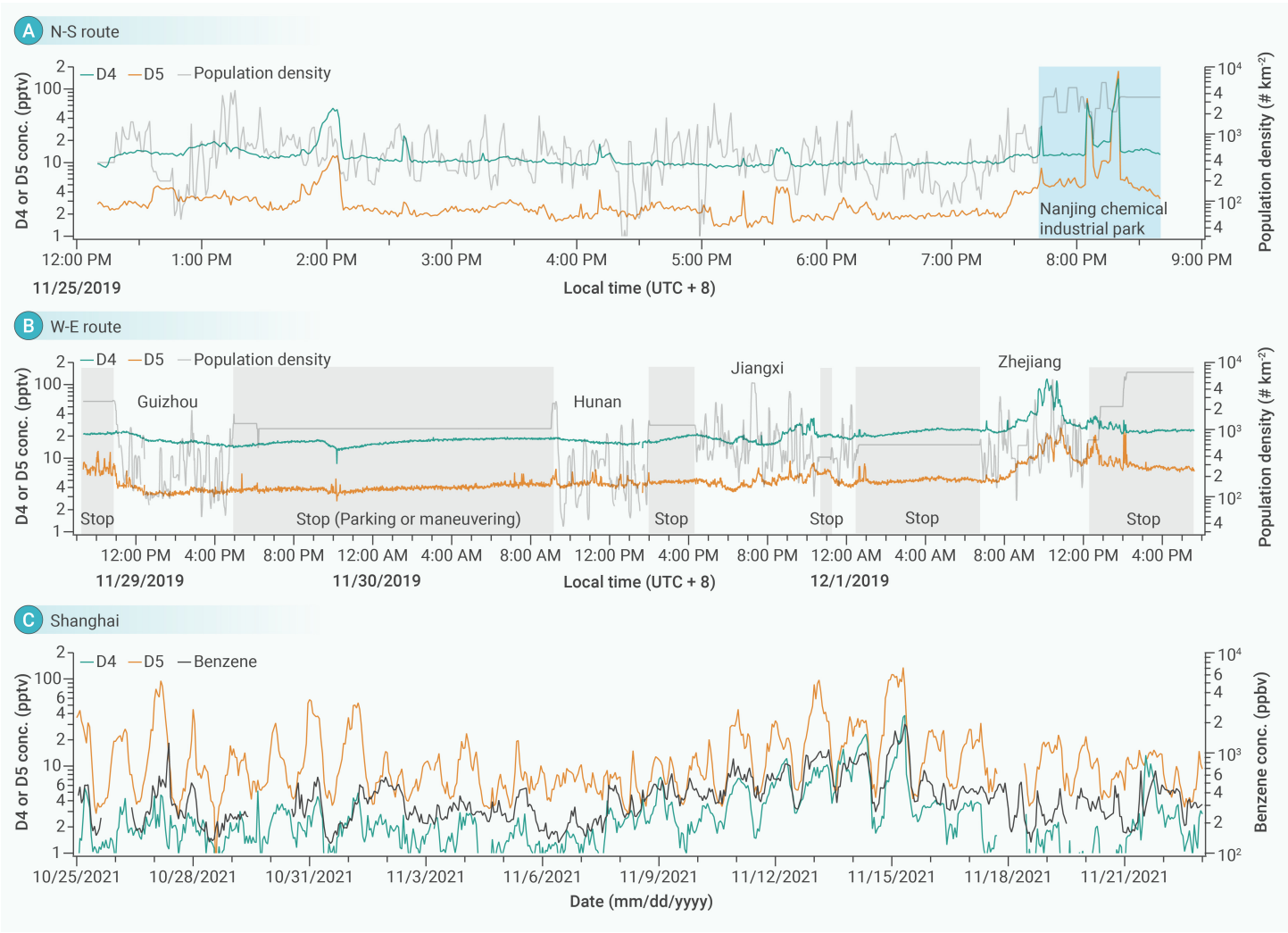

Figure 2. Time series of atmospheric D4 and D5 and associated parameters (A) Observed in the N-S route. (B) Observed in the W-E route. (C) Observed in downtown Shanghai.

Table 1. Summary of atmospheric D4 and D5 concentrations measured from three field campaigns.

			D4 (pptv)					D5 (pptv)			
			N	Mean ± 1σ	Median	Min	Max	Mean ± 1σ	Median	Min	Max
N-S route	All		511	13.1 ± 9.5	10.7	8.4	139.1	4.0 ± 10.6	2.4	1.3	175.7
	Industrial section		70	20.0 ± 20.6	13.6	11.7	139.1	12.9 ± 26.9	5.0	3.2	175.7
	Non-industrial section		441	11.9 ± 5.5	10.4	8.4	54.8	2.6 ± 1.3	2.3	1.3	12.5
W-E route	All		3363	21.2 ± 10.0	18.5	8.4	119.3	5.4 ± 2.7	4.6	2.6	26.0
	Non-stop section	All	1440	23.1 ± 14.3	18.4	13.7	119.3	5.7 ± 3.4	4.6	3.0	26.0
		Guizhou section	359	17.6 ± 2.5	16.8	14.1	23.7	3.7 ± 0.8	3.5	3.0	8.5
		Hunan section	291	16.8 ± 1.0	16.6	13.7	19.2	4.6 ± 0.6	4.5	3.4	7.2
		Jiangxi section	461	19.6 ± 3.8	19.0	14.4	35.3	5.0 ± 0.9	4.8	3.5	9.3
		Zhejiang section	329	39.6 ± 22.8	30.2	21.9	119.3	10.0 ± 4.8	9.1	4.5	26.0
Shanghai	All		702	3.2 ± 3.6	2.1	0.2	28.2	14.8 ± 17.2	8.8	0.6	135.3

favor accumulation. Nevertheless, the sheer magnitude of the observed regional concentration differences strongly suggests that the spatial distribution of emission sources is the primary determinant of the large-scale patterns reported, even as meteorology undoubtedly modulates concentrations. Figure 3 shows the potential association between atmospheric D4 and D5 observed from mobile measurements and their relationship with population density. D4 and D5 are closely related as mentioned above, but they also have some differences. Obtaining through hydrolysis and cracking of dimethyldichlorosilane monomers, D4 is the primary building block of other silicone materials (including D5).[2] In comparison, D5 is mainly used as an ingredient in personal care and consumer products and in dry cleaning.[1] Therefore, the D4/D5 ratio may indicate the relative importance of cVMS production versus consumption. While population density has been considered a predictor of atmospheric D5 in some urban studies due to its link with

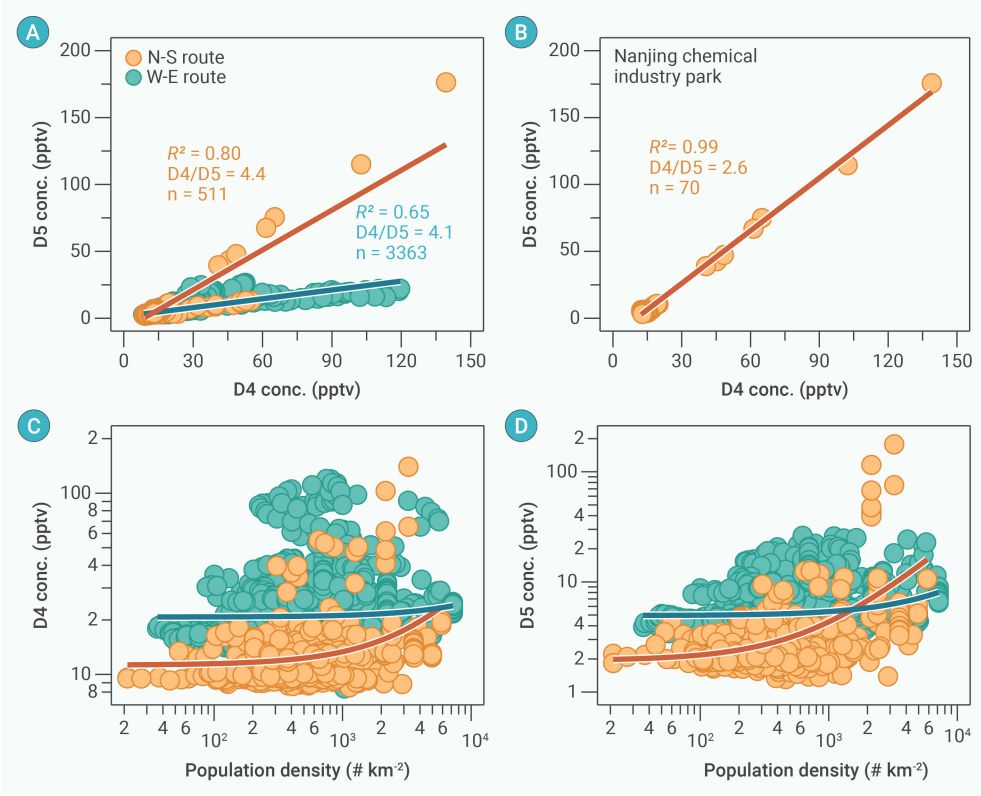

Figure 3. The correlation between atmospheric D4 and D5 (A) Observed from mobile measurements along the N-S and W-E routes. (B) Observed in the vicinity of Nanjing Chemical Industry Park. (C) The correlation between population density and atmospheric D4 observed from mobile measurements along the N-S and W-E routes. (D) The correlation between population density and atmospheric D5 observed from mobile measurements along the N-S and W-E routes.

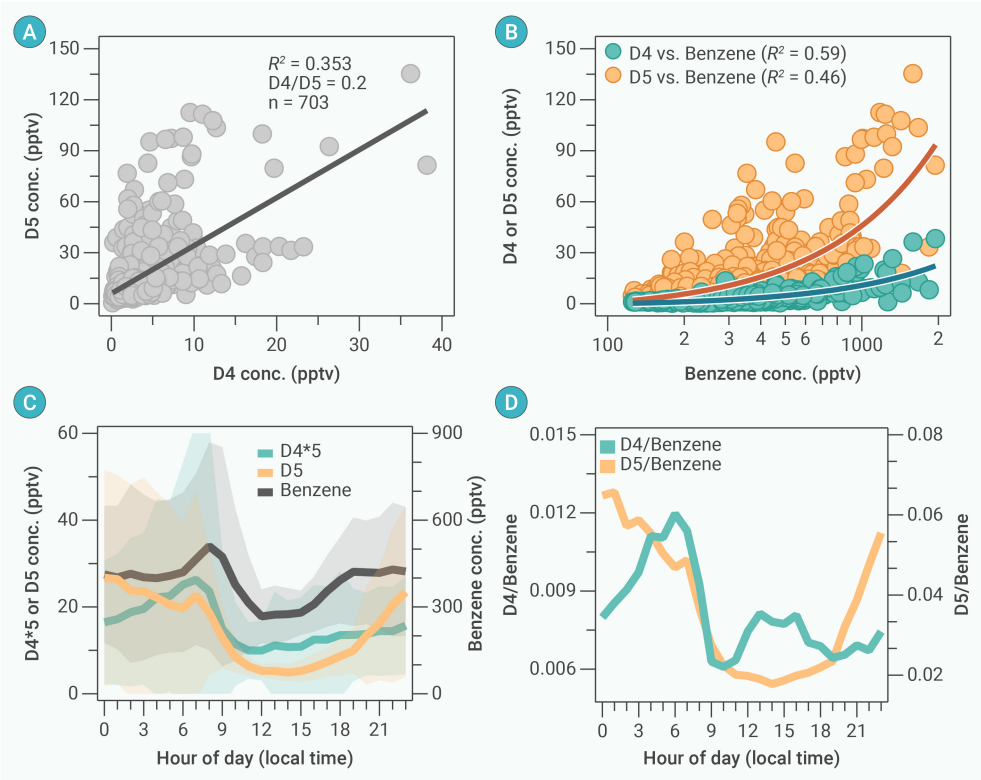

Figure 4. Observations in downtown Shanghai (A) The correlation between atmospheric D4 and D5 observed from in-situ measurements. (B) The correlation between benzene concentration and D4 or D5 concentration. (C) Diurnal variations of D4 (multiplied by 5), D5, and benzene concentrations. (D) Diurnal variations of D4/benzene and D5/benzene ratios.

In Figure 3A, we note that D4 and D5 measured along the N-S route has a stronger correlation. Besides, in Figure 3B, the average D4/D5 ratio near the NCIP dramatically shrinks to 2.6. There are multifold factors. Firstly, during the manufacturing process, D4 can be mostly consumed as an intermediate.[2] Therefore, D4 may not increase as rapidly as D5 near industrial point sources. Secondly, D4 typically has a longer atmospheric lifetime (~11 days) than D5 (~7 days).[6] It means that the D4/D5 ratio generally tends to be higher as the distance from industrial emission sources increases. Lastly, the atmospheric removal of D4 and D5 is largely determined by their reactions with OH radicals.[6,10] Our mobile measurements were performed in late autumn, and the concentration of OH radicals would significantly decrease as one moves northward, which leads to higher atmospheric stability of D4 and D5 in northern China. This can subsequently result in higher ratios and stronger correlations between D4 and D5 in the N-S route compared to the W-E route.

Figure 4 illustrates the characteristics of D4 and D5 measured at hourly resolution from in-situ observations in downtown Shanghai, the most developed city in China. In the urban setting of Shanghai, D4 shows a moderate correlation with benzene, a tracer of vehicular emissions (Figure 4B), while D5 shows a weaker correlation, suggesting contributions from mobile sources primarily influence urban D4. This contrasts with the mobile measurements outside major urban centers, where large-scale industrial emissions appeared to be more dominant drivers for the observed regional cVMS patterns, particularly for D4. Overall, the concentration of D4 (3.2 pptv) is much lower than D5 (14.8 pptv), resulting in a very low D4/D5 ratio (0.2 on average). These characteristics are similar to those observed in developed countries (e.g., Chicago and New York in the US)[10,16,36], confirming that atmospheric cVMS in developed and population-dense cities is mainly driven by D5 emissions from the consumption of personal care products. However, in Shanghai, D5 peaks at midnight and doesn't show high values during the evening rush hour (Figure 4C), which is consistent with the influence of personal care product usage,[15,16,35] our mobile measurements across largely non-urban routes show no significant correlation between population density and either D4 (Figure 3C) or D5 (Figure 3D). This suggests that outside dense urban areas in China, population density alone is not a primary driver, and cVMS levels, particularly D4, are more strongly influenced by industrial production sources. The higher concentrations along the W-E route compared to the N-S route (Table 1) further support the conclusion that silicone production is more prominent in southern/eastern China (Figure 1).

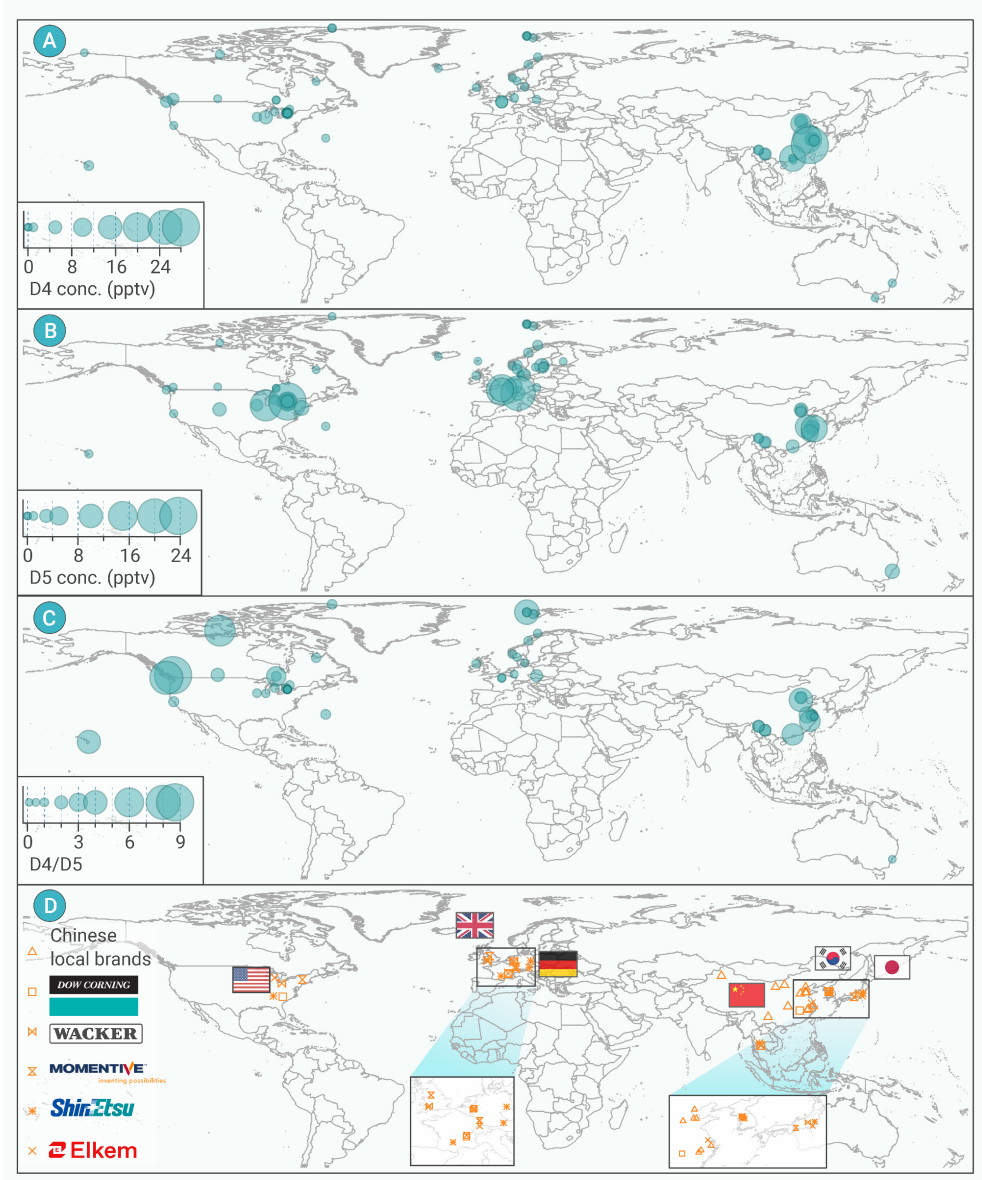

Figure 5. Global distribution of D4 and D5 in the Atmosphere Global distribution of atmospheric concentrations of D4 (A) and D5 (B). Global distribution of D4/D5 ratios (C). The locations silicone production factories and their manufactures global.

the urban boundary layer. Moreover, different from D4, the enhancement of D5 (indicative of D5/benzene in Figure 4D) maximizes during nighttime (< 0.1) and its amplitude is much smaller than those observed in developed countries (e.g., Boulder in the US (0.3)[17]). This suggests that personal care product emissions may still not be a major source of volatile organic compounds in Chinese cities.

Atmospheric D4 and D5 in China viewed from a global perspective

To provide a global context for our investigation into the emission sources, socioeconomic drivers, and long-range transport potential of atmospheric D4 and D5 in China, we have gathered relevant observational results from around the globe[8-11,13,16-17,28-29,37-42] in Table S1 and plotted them in Figure 5. It is crucial to recognize that direct quantitative inter-comparison must be approached with caution. Inherent variations in sampling strategies, analytical instrumentation, and prevailing local meteorological conditions can all contribute to inter-study variability. Nevertheless, by compiling and examining data from a wide array of locations, it is possible to discern broad geographical trends. The majority of these studies were conducted in the Northern Hemisphere, and the extensive spatial coverage of the compiled data reduces uncertainties caused by variations in sampling periods and analysis methods, thereby enhancing the reliability and comparability of our findings.

In Figure 5A, it is evident that China is a global hotspot of atmospheric D4. Throughout our three field campaigns, the average concentration of atmospheric D4 was 17.5 pptv, while other sites had atmospheric D4 concentrations mostly below 1 pptv, with the highest concentrations observed in Chicago, US (5.3 pptv)[10] and Paris, France (4.5 pptv).[37] Superficially, high-latitude regions such as the Arctic, where the population is sparse, also have very low atmospheric D4 concentrations. However, Figure 6A does not support the conclusion that population density and associated use of personal care products drive the global distribution of atmospheric D4. On the other hand, Figure 5D shows that global silicone production is highly concentrated in China, the US, Germany, the UK, Japan, France, South Korea, and Thailand. China accounts for nearly 60% of the total production (Figure 7B), while the silicone production of other countries is controlled by only five chemical giants. This matches the global distribution of atmospheric D4, confirming that atmospheric D4 is more likely to be driven by industrial production. We emphasize here that as the global hub of siloxane production, China's most severe atmospheric D4 pollution should be addressed from the production side rather than the consumption side. This positions China as a critical region for future global D4 emission inventory refinement and underscores the necessity of focusing mitigation strategies on industrial sources within major producing nations to effectively reduce global atmospheric D4 concentrations.

In Figure 5B, atmospheric D5 has three global hotspots: The middle and eastern US, Western Europe, and eastern China. Figure 5C shows that the D4/D5 ratios in remote areas (such as the Arctic, generally above 4) are significantly higher than those in population-dense regions, suggesting limited influence of long-range transport and atmospheric degradation on ambient D5 concentrations in cities. Unlike D4, Figure 6B suggests a potential role for population density in influencing ambient D5 concentrations in cities globally, likely reflecting emissions linked to personal care product usage (which occurs indoors but vents outdoors or enters wastewater streams). However, this relationship is not straightforward across all environments. As Coggon et al.[16] showed for the US and Europe, population density can be a factor in urban volatile chemical product emissions (including D5). Our analysis further suggests (Figure 6C) that D5 concentrations also correlate with D4 levels globally (though not strongly), indicating that silicone production retains an influence on ambient D5 patterns even in consumption-dominated areas. Furthermore, when comparing globally and within China's diverse economic landscape (Figure 7D), personal income emerges as a potentially more robust indicator of consumption patterns driving urban D5 variability than population density alone. This finding has significant worldwide implications, suggesting that socioeconomic parameters beyond simple population counts should be incorporated into models aiming to predict and create inventories for D5 emissions from personal care products on a global scale. Nevertheless, we cannot deny the great impact of personal care product consumption on atmospheric D5, as the highest concentrations of atmospheric D5 are found in Toronto[17] and Zurich,[11] where there is no silicone production in their respective countries (i.e., Canada and Switzerland).

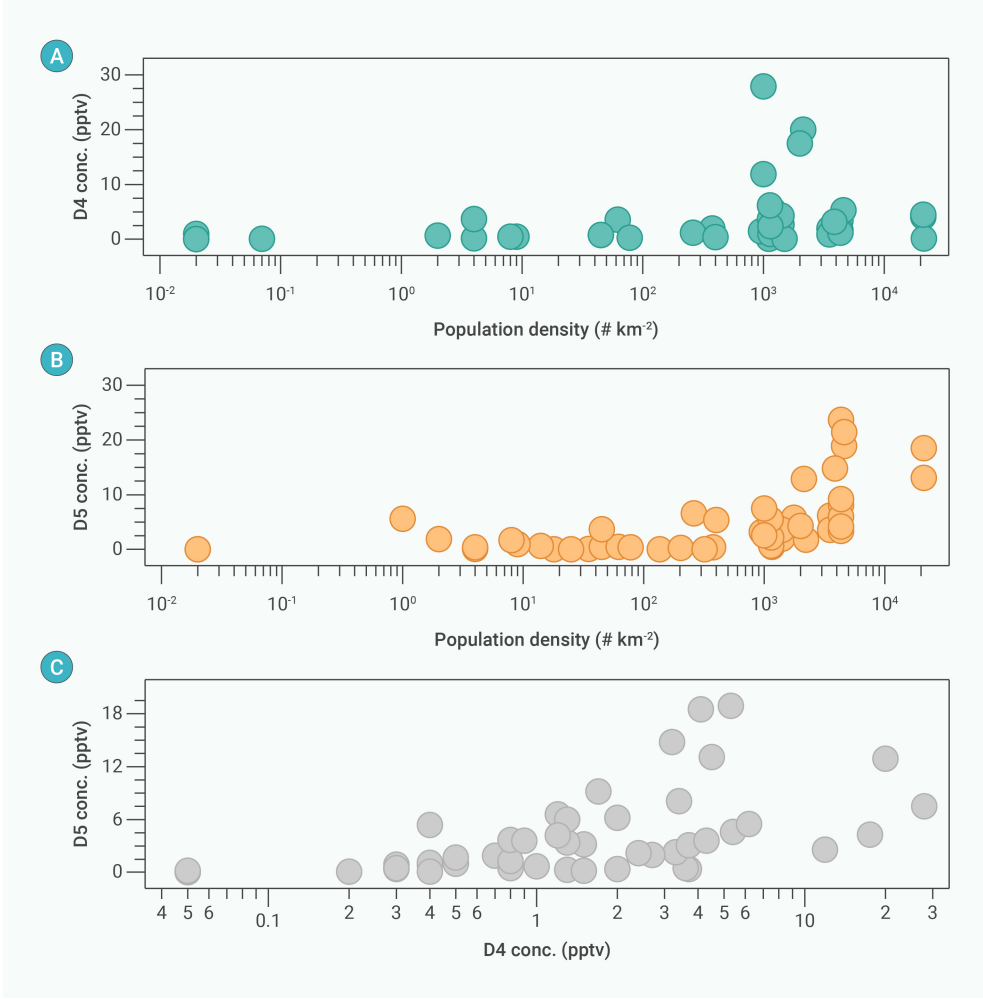

Figure 6. Relationships between global atmospheric D4/D5 concentrations, population density, and inter-compound correlations (A, B) Correlations between atmospheric D4 and D5 concentrations and population density across the investigated regions. (C) Scatter plot showing the correlation between atmospheric D4 and D5 concentrations.

Superficially, China's distinctive characteristics regarding D5 seem to complicate the global spatial pattern of atmospheric D5. Specifically, despite having the highest population density and silicone production in eastern China, atmospheric D5 concentrations in this region are still lower than in some developed cities in Europe and the US. While silicone production and consumption in China have been increasing year by year, they are unbalanced (Figure 7A). In 2019, China's share of global silicone production and consumption was 57% and 44%, respectively (Figure 7B). Looking more closely, silicone consumed in China's medical/personal care products accounted for only 10% of the total consumption (Figure 7C). Recently, Li et al.[43] found that higher D5 emissions from urban residential wastewater in China are associated with higher personal income. This study has not received enough attention, as unlike China, the economic differences among developed countries are relatively small. However, China is an emerging country with large domestic economic disparities. For example, in our study region, Shanghai had the highest GDP (Gross Domestic Product) per capita in 2019 (¥157 k), followed by Jiangsu (¥119 k), Zhejiang (¥116 k), Shandong (¥86 k), Jiangxi (¥56 k), Hunan (¥52 k), and Guizhou (¥39 k). This is generally consistent with the regional concentration gradient of our measured atmospheric D5 (Figure 2 & Table 1), indicating that areas with higher income levels may have higher personal care product consumption, leading to higher atmospheric D5 concentrations. Such rule appears to be applicable globally, as shown in Figure 7D, where silicon consumption per capita in different countries/regions is a function of GDP per capita. Our observation that personal income better explained D5 heterogeneity than population density represents an initial, encouraging step in this important area of research. While a full investigation using multivariate regression models to disentangle the complex interdependencies of GDP, industrial structure, population density, and income was beyond the scope of this initial large-scale mapping study, such analyses are certainly a valuable and necessary next step for future research aimed at developing robust predictive models for cVMS emissions. In conclusion, our findings suggest that personal income and associated use of personal care products could be a universal driving force behind the large variability of urban atmospheric D5. These results may help establish a parameterization scheme for developing a global D5 emission inventory.

CONCLUSION

In summary, here we report on highly time-resolved and ultra-sensitive mobile measurements of atmospheric cVMS concentrations over two pan-region transects in China, using a mobile laboratory equipped with the Vocus Proton-Transfer-Reaction Time-of-Flight Mass-Spectrometry. Additionally, we made in situ measurements of D4 and D5 at a ground site in Shanghai, the largest megacity in China. We found that (1) the distribution pattern of atmospheric D4 and D5 in China shows a decreasing gradient from the southeast towards other regions; (2) D5 had an overall concentration level similar to that of developed countries in Europe and North America, while D4 was one order of magnitude higher; (3) the sources of non-urban and urban atmospheric cVMS are mainly driven by their production and consumption, respectively, as primarily evidenced by their distinct, directly observed spatial distributions and the identification of significant emission hotspots co-located with industrial zones during our mobile campaigns. These direct observations are further supported by contrasting D4/D5 characteristics between industrial, non-urban, and urban settings, and by correlation analyses with relevant tracers or indicators; and (4) globally, personal income could be a universal driving force behind the large variability of urban atmospheric D5, which pave the way of establishing a parameterization scheme for developing a global D5 emission inventory. We make the important point that unlike developed countries, China may need to address its atmospheric cVMS problem more from the production side. Therefore, understanding and mitigating China's production-related cVMS emissions is not only a national imperative but also holds considerable importance for global atmospheric cVMS budgets, potential long-range transport, and international efforts to manage persistent and bioaccumulative substances. While this study offers unprecedented initial insights, we recognize that future investigations incorporating measurements across all seasons are crucial. Such studies would allow for a more complete understanding of cVMS temporal dynamics.

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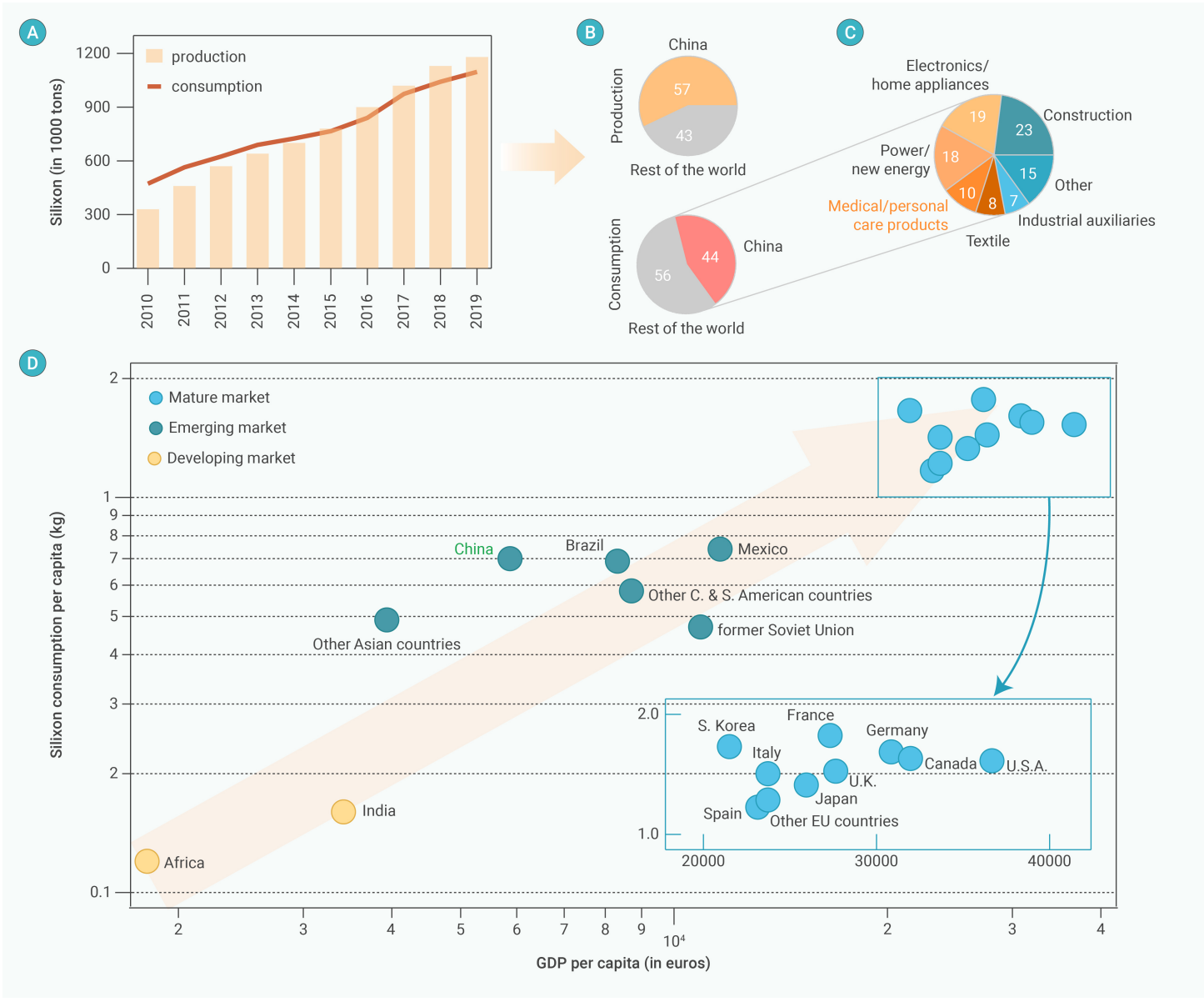

Figure 7. Silicone Development Trends (A) Ten years (2010-2019) of annual silicone production and consumption in China. (B) China's shares of silicone production and consumption in the world in 2019. (C) The structure of silicon consumption in China in 2019, with medical/personal care products accounting for only around 10%. (D) Silicone consumption per capita by country/region as a function of GDP per capita.

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AUTHOR CONTRIBUTIONS

Y.C. designed the research. Y.C., W.T. and L.Z. conducted the field campaign and data analysis. Y.C. wrote the manuscript with input from all co-authors. All authors contributed to the manuscript and approved the final version.

DECLARATION OF INTERESTS

The authors declare no competing interests.

DATA AND CODE AVAILABILITY

The population density in China in 2019 is available to download in Geotiff and ASCII XYZ format at a resolution of 30 arc (approximately 1km at the equator) from wordpop: https://www.worldpop.org/geodata/summary?id=44833. Other data are included in the article and/or Supplementary Information.

SUPPLEMENTAL INFORMATION

It can be found online at https://doi.org/10.59717/j.xinn-geo.2025.100155