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Atmospheric amines are a crucial yet missing link in Earth's climate via airborne aerosol production

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Atmospheric amines, derivatives of ammonia, play a unique yet not fully understood role in air quality, climate and public health. Sub-5 parts per trillion Volume (pptV, $<10^{-12}$ in volume) mixing ratios of amines facilitate the physical and/or chemical transformation of aerosols in the atmosphere, enhancing aerosol formation and growth rates, aerosol hygroscopicity, and the activation of cloud condensation nuclei. This serves as the initial step for cloud droplet formation and, consequently, influences cloud properties and the hydrological cycle. Ambient observations demonstrate more than a thousand-fold particle formation rates in the presence of amines as compared to ammonia. Yet, the challenges related to detecting minute levels of amines, the paucity of ambient amine measurements, and the limited process-based understanding of airborne aerosol production have resulted in amines being underrepresented in global climate models. Therefore, advanced techniques with extremely low detection limits and highly spatially and temporally resolved ambient amine measurements globally in diverse environments are essential.

The interaction between the Earth's ecosystem and the atmosphere is a non-linear and dynamic process that has been evolving differently in recent decades¹. This is due to changes in atmospheric factors, including levels of greenhouse and reactive gases, water vapor, and the uneven distribution of aerosol concentrations and sizes over time and space. Additionally, variations in atmospheric dynamics, the state of the oceans, and the cryosphere contribute to these changes. Therefore, the pressing issue of climate change – a major challenge of the 21st century, is driven by the need for a thorough process-level understanding of the diverse environmental factors that add to its intricacy. Despite advancements in regional to global climate models incorporated with process-based parameterizations based on controlled laboratory experiments, ambient measurements and satellite observations, the process-level understanding of the climate system remains elusive. One such complex driver is the interaction between aerosols and clouds, which remains the largest source of uncertainty in the climate system due to its dynamic and non-linear relationships². This is mainly due to limitations in model assimilation and validation, exacerbated by the uneven geographical distribution and spatial heterogeneity of measurement networks, asynchronous monitoring, and inconsistencies in data collection methods of various atmospheric variables. This emphasizes the need for advanced climate prediction models with fewer uncertainties to reduce climate risk to the Earth's ecosystem. Therefore, it is paramount to improve our current process-level understanding – specifically, how aerosols form, grow,

activate, and interact with clouds and radiation, thereby affecting air quality, the hydrological cycle, climate, and human health.

Models estimate that up to 90% of aerosols are airborne via the condensation of low-volatility vapors, such as sulfuric acid (SA)³. However, the formation of initial molecular clusters in the planetary boundary layer would not be possible without binding agents like ammonia (NH₃) and amines^{4,5}. These agents promote the growth of molecular clusters by preventing their evaporation, making them essential for aerosol formation. One exception is a study that showed ion-induced nucleation of pure biogenic volatile organic compounds (VOCs) without SA in a chamber experiment under atmospheric conditions⁶. Atmospheric amines are ubiquitous organic bases that are emitted into the atmosphere from a wide range of natural sources, such as marine organisms, vegetation and forest fires, and anthropogenic sources, such as biomass and fossil-fuel burning, animal husbandry, waste incineration, sewage treatment and residential cooking^{7–9}. Gas-phase concentrations of amines are typically reported to be 10 to 1000 times lower compared to NH₃ concentrations^{10,11}, but have a thousand-fold higher enhancement factor for particle nucleation rates than NH₃ at low concentrations^{4,12}. In laboratory settings, dimethylamine (DMA) has been shown to replace NH₃ in molecular clusters with SA, making it an interesting target compound for ambient particle formation studies at the molecular level. It should be noted that near emission sources, amine mixing ratios can reach as high as hundreds of parts per billion in Volume (ppbV),

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as one study reported butylamine (BA) in the range from 27.3 to 187.7 ppbV in areas near livestock operations¹³. But, reports on amine speciation and ambient concentrations at the level relevant to new particle formation (NPF) are rare and discrepant^{14,15}.

Ge et al.^{7,16} provided the first and the most comprehensive fundamental assessment of amines sources, fluxes, dynamics, and health effects. Gas-phase amines in the atmosphere primarily follow two reaction pathways: gas-phase reactions with oxidants and participation in heterogeneous reactions with acidic or carbonyl compounds, subsequently partitioning into nanoparticles. Lee and Wexler¹⁷ delineated the gas-phase reaction rates of amines with hydroxyl (OH) radical, ozone (O₃), nitrate (NO₃) radical, chlorine (Cl) atoms, and photolysis, as well as the products from these reactions. Previous studies also synthesized laboratory experiments of the multiphase chemistry of amines, including acid-base neutralization, carbonyl-amine interaction and particle-phase oxidation¹⁸, and measurements of reduced nitrogen-containing compounds in both the gas and particle phases¹⁹. Shen et al.⁸ recently summarized a decade of progress in understanding emission sources, detection methods, and oxidation reaction mechanisms to elucidate their direct or indirect effects on air quality and climate.

To obtain a holistic view of the role of amines in atmospheric chemistry, particle formation, air quality and climate, multiple laboratory experiments, quantum chemical calculations, and field observations have been conducted. These studies show that amines contribute to atmospheric particle formation and growth^{4,5,20–25}, the formation of organic brown carbon (BrC)^{26,27}, cloud water droplets^{28,29}, aqueous fog and rain droplets³⁰, and plays a crucial role in the biogeochemical nitrogen cycle³¹. Low molecular weight amines, like trimethylamine (TMA), not only have higher basicity than NH₃ but also account for 14–35% of the concentrations of NH₃ in aerosol particles³², yet amines are underrepresented in global climate models. The major uncertainties in the global climate model arise from limitations in measurement technology and model inaccuracies, which must be better constrained to accurately quantify the role of amines in air quality and climate^{33–36}. Additionally, many amines pose risks to public health due to their toxicity and allergenic effects, with some capable of forming carcinogenic compounds, such as nitrosamines when they react with nitrites^{7,17}. As carbon capture and storage (CCS) technologies increasingly rely on organic solvents such as monoethanolamine (MEA) or methyldiethanolamine (MDEA) to reduce carbon emissions from fossil-based energy and industry sources, ambient concentrations of amines will likely rise in the future³⁷. While the Intergovernmental Panel for Climate Change (IPCC) highlights that accelerating CCS technologies could help the world to surpass climate tipping points², the potential impacts of rising amines concentrations on the environment and climate are under-recognized in the IPCC report. Field observations and modelling proxies reveal decreasing emissions of sulfur dioxide (SO₂, precursor of SA and the main sink of amines via acid-base neutralisation), condensation sink, particle formation rates, and NPF frequencies³⁸, as a result, we should expect a longer atmospheric lifetime of amines and NH₃, allowing their further transport to remote areas, including marine and polar areas. Furthermore, there is a risk of drinking water contamination by nitrosamines and nitramines³⁹. Hence, it becomes increasingly important to monitor the ambient concentration of amines⁴⁰ and study their gas-phase oxidation products to assess environmental impacts and health risks associated with amines from CCS facilities. Consequently, scientific research on atmospheric amines is rapidly growing globally, encompassing field observations, theoretical and numerical calculations, laboratory experiments, and advanced state-of-the-art techniques for detecting minute levels of amines in the air.

Why do we need accurate monitoring of atmospheric amines?

Prior to the 1980s, research on atmospheric amines primarily focused on regions with high emissions, such as livestock farms and waste treatment facilities, motivated by the potential carcinogenic risks of their derivatives,

especially nitrosamines⁴¹. In addition to the potential negative effects of amines on human health, there has been colossal interest in amines and their derivatives in aerosols, their impact on air quality and climate. Laboratory experiments and field studies have both demonstrated the substantial contribution of amines to initial cluster formation. While NH₃ effectively stabilizes clusters, amines (e.g., DMA, the strongest stabilizer among amine molecules²²) can multiply the particle formation rate by up to 1000 times compared to NH₃^{4,20,42,43}. Further, studies have demonstrated that the combined presence of amines and NH₃ can lead to more efficient particle formation with SA and water compared to cases where NH₃ is absent⁴⁴. Since multiple particle formation pathways can be active simultaneously in the atmosphere, identifying dominant cluster formation pathways from field measurements is challenging. However, both atmospheric field measurements and laboratory experiments have highlighted the importance of SA, organic compounds and amines for NPF^{5,45–49}. Previous studies have identified amines as an important organic component in sub-micron aerosols^{28,50–55}, either via acid-base reactions with gas-phase inorganic acid or acidic aerosols⁵⁶ or via condensation products following oxidation reactions with OH, O₃, and NO₃^{57–59}, with yields from 8 to 15% from oxidation of amines⁶⁰. But, the exact contribution of amines, along with acids (e.g., SA, iodic acid), other bases (e.g., NH₃) and organics (e.g., highly oxygenated organic molecules, HOMs), to the growth of sub-3 nm clusters in diverse environments, is still elusive^{61–64}.

Figure 1a shows the geographic distribution of locations where ambient concentrations of gas-phase and/or particle-phase amines have been reported in the literature. This highlights the global scarcity of such observations, especially in the Southern Hemisphere, Southeast Asia and the Arctic, as well as discrepancies in reported amine concentrations in both the gas- and particle-phase. Although over 150 amines have been identified in the atmosphere¹⁷, only about 10 of them have been widely reported. The reported concentrations of gas-phase amines exhibit variability of over ten thousand-fold (ranging from <0.1 to >10⁵ pptV) across different seasons, sources, and detection methods (Table 1). Particle-phase amine concentrations also show large variability, ranging from 0.02 to 425.9 ng m⁻³ (Table 2). The emission of amines from a particular source also shows a thousand-fold variability in both the gas- and particle-phase concentrations. For example, gas-phase amines from agriculture activities range from <0.5 to >100 pptV^{65–68}. Moreover, the different techniques employed exhibit large variability in the measured levels of gas-phase and particle-phase amines. For example, gas-phase DMA concentrations ranged from <0.15 to >80 pptV in Hyttälä boreal coniferous forest, Finland during spring or summer^{15,69–71}, while particle-phase DMA concentrations in the particulate matter (particles with aerodynamic diameter of 2.5 µm or less, PM_{2.5}) ranged from 2.4 to 280 ng m⁻³ in coastal Qingdao, China during winter^{72,73}. Figure 1b, c depict the variability in different gas-phase and particle-phase amines based on reported ambient measurements (Tables 1 and 2). Livestock operations show the highest levels of different gas-phase amines, whereas other sources exhibited erratic behaviour in amines, encompassing data points across the entire range. Nevertheless, the accurate detection of amines, especially in the gas-phase, is lacking for several reasons; (i) current mass spectrometers cannot separate individual amine species and can only detect sums such as C2 or C4 amines due to their identical elemental composition, (ii) measurements are affected by contamination and memory effects, which increases detection limit, and insufficient resolution in mass spectrometers, resulting uncertainty in peak identification. The lack of analytical standards and zero samples exacerbates these issues, and (iii) there are discrepancies between laboratory/theoretical and ambient observations, such as the unobserved replacement of NH₃ in clusters by DMA under ambient conditions^{5,74,75}. Thus, the critical knowledge gap in amine measurement technology concerns whether the reported amine mixing ratios are subject to low or high bias from instrumental detection issues or by the natural variability stemming from varying source strengths. This indicates that the development of global gridded emission inventories for amines is currently not feasible. Notably, the oversimplified regional gridded inventories of amine emissions have been shown to hinder the simulation

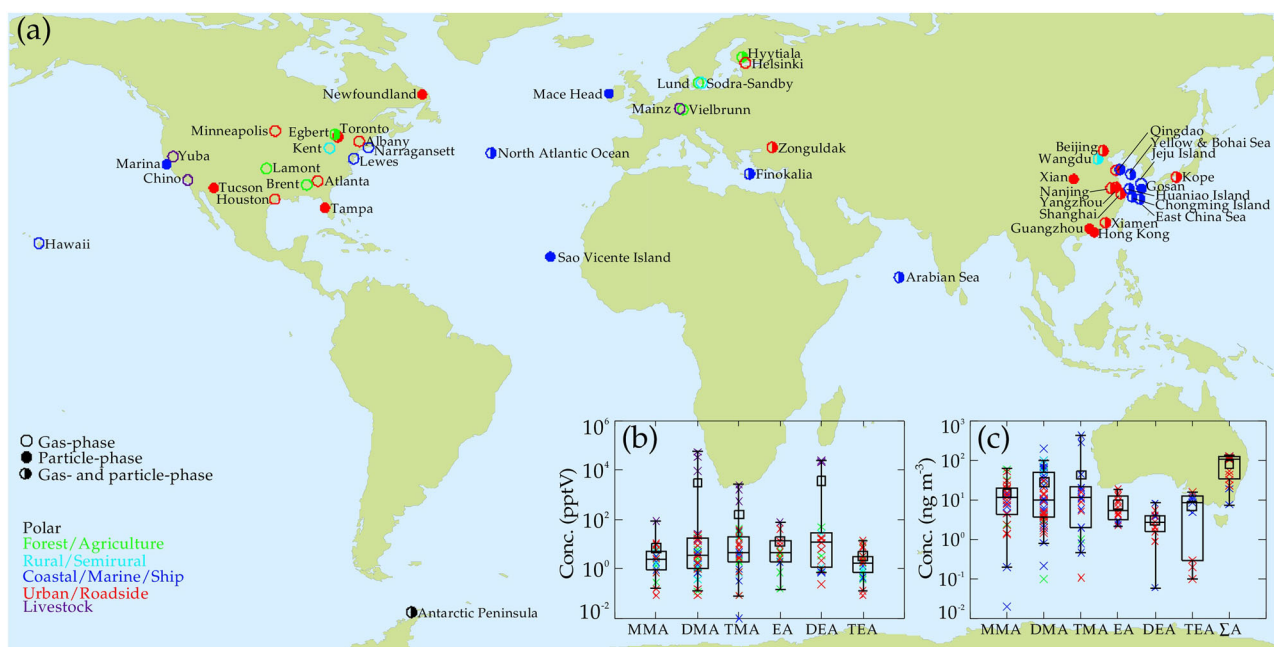


Fig. 1 | Overview of ambient gas-phase and particle-phase amines across the globe. **a** Geographical distribution of locations where ambient observations of gas-phase amines (open circles), particle-phase amines (filled circles), or both (open left half and filled right half circles) are reported. The colour of the symbol indicates the type of environment (see legend). **b** Box-whisker plot of gas-phase concentrations of widely reported amines (Table 1). **c** Same as (b) but for particle-phase amines (Table 2). The black open square indicates the mean, the horizontal black line indicates the median, the bottom and top of the box represent the 25th and 75th

percentiles, and the bottom and top of the whisker represent the 5th and 95th percentiles. The cross symbols indicate all individual data points, with the colour referring to the type of environment. ΣA refers to the sum of particle-phase amines. Details of amine measurement technology, location and time periods are presented in Tables 1 and 2. MMA Monomethylamine, DMA Dimethylamine, TMA Trimethylamine, EA Ethylamine, DEA Diethylamine, TEA Triethylamine, ΣA total mass of aliphatic amines.

accuracy of atmospheric numerical models⁷⁶. Although minute quantities of acidic compounds dominate aerosol numbers, resolving the role of amines in particle formation and growth has implications for aerosol-cloud-radiation-climate interaction research. Previous laboratory and atmospheric studies have observed high NPF rates involving precursor vapors (usually SA) stabilized by amines^{24,43,61}, and these clusters subsequently grow to large sizes by condensing vapors which are typically low-volatile organic and inorganic compounds^{49,77,78}. Several studies showed the vital contribution of NPF to cloud condensation nuclei (CCN) concentrations, thus affecting the cloud properties, especially in pristine areas^{3,79–82}. However, amines influence CCN activity by either enhancing particle hygroscopicity through the formation of aminium salts³² or suppressing it through photochemical reactions with OH radicals⁸³. Tang et al.⁸³ demonstrated that secondary organic aerosols (SOA) formed from the reactions of TMA and BA with OH radicals consist of organic material with low hygroscopicity, characterized by a single hygroscopicity parameter ($\kappa \leq 0.25$). Although BA is less abundant in the atmosphere compared to TMA, it has been detected at high concentrations near dairy facilities, reaching concentration levels up to 187 ppbV¹³. This indicates that CCN activity for amines depends on the reaction pathways of specific amines and their magnitude, indicating climate impacts associated with amines are complex. Therefore, the accurate detection of a variety of amines and their chemical pathways are crucial for improving our process-level understanding of the climate system which could open new opportunities for Earth system modelling.

Amines impact airborne aerosol production

Airborne aerosol production is a non-linear dynamic process involving multiple components in the formation of aerosols, with amines facilitating the fastest base-neutralisation mechanism, thereby decreasing the evaporation of nucleated clusters⁴. Field studies on atmospheric acids have become more prevalent with the development of mass spectrometric methods that have low detection limits (sub-pptV, $<10^{-12}$ in volume) and

high time and mass resolution. However, observations of base compounds (e.g., amines) in the field are almost solely carried out by estimating their abundance in acidic molecular clusters (e.g., SA)^{24,66,84,85}. Direct measurements of amines at sub-5 pptV concentrations are difficult to reach, even in ultra-clean chamber facilities¹⁴. Further, simultaneous real-time measurements of gas-phase amines and their particulate counterparts in the marine and polar remote atmospheres remain challenging (Fig. 1), although this is not the case in the continental atmosphere⁵⁴.

Amines are emitted into the atmosphere from diverse natural and anthropogenic sources in both continental and marine environments⁷. Anthropogenic activities predominantly contribute to amine emissions, although approximately 30% of amines in the atmosphere are believed to originate from the decomposition of organic matter in the oceans^{4,7} that can be transported to remote continental locations. Since amines from continental and marine sources can be transported regionally across atmospheric environments, the reported acid-amine neutralisation mechanisms can be broadly categorized into continental and coastal/marine atmospheres.

Continental atmosphere

Amines in continental atmospheres come from both natural (vegetation and forest fires) and anthropogenic sources (biomass/fossil-fuel burning, animal husbandry, waste incineration, sewage treatment, and residential cooking). Once in the air, amines can rapidly react with SA to form stable salt particles through acid-base reactions. Whether amines form aminium salts depends not only on temperature but also on the specific amine involved, its concentration, and the type of acidic species present. In addition, the presence of NH_3 competes with amines for acidic molecules⁵⁶. The formation rate of nanometre-sized particles reaches saturation with amine mixing ratios as low as 5 pptV in atmospherically relevant SA concentrations^{4,86}. The CLOUD chamber experiment showed that the addition of only 5 pptV DMA enhances the nucleation rate of SA by more than six orders of magnitude at 10 pptV NH_3 , 278 K and 38% relative

Table 1 | Summary of measurement location, time period, technique and concentrations of gas-phase amines in diverse locations

Location	Type	Lat. (deg.)	Lon. (deg.)	Time Period	Technique	Concentrations (pptV)					Reference	
						MMA	DMA	TMA	EA	DEA		TEA
Hyytiälä, Finland	Forest	61.51	24.17	May–Jun 2013	Bisulfate-cluster Cl-API-TOF		<0.150					Sipilä et al. ¹⁵
				Mar 2001	H ₃ O ⁺ (H ₂ O) _n CIMS		<32	34–80			Sellegrì et al. ⁶⁹	
				Mar–Dec 2015	MARGA-MS	<1.9–8.8	<1.7–4.1	<0.1–6.1	<0.19–8.2		Hemmiälä et al. ⁷⁰	
				May–Oct 2011	LC-MS		42 ± 30 ^a				Kieloaho et al. ⁷¹ #	
Egbert, Canada	Agriculture	44.23	–79.78	Oct–Nov 2010	AIM-IC		1–10 (6.5 ± 2.1)				<1.0 ^k	VandenBoer et al. ⁶⁵
Brent, AL, USA	Rural Forest	32.94	–87.18	Jun–Jul 2013	Ethanol CIMS	<0.37	<1.51	1–10	<8.09	<5.44	<4.05	You et al. ²⁰¹ y
Vielbrunn, Germany	Agriculture	49.71	9.09	May–Jun 2014	Nitrate Cl-API-TOF	1–5	~1	1–5	1–5		1–5	Kürten et al. ⁶⁶
Mainz, Germany	Livestock	49.99	8.24	Apr–Sep 1993	GC	173.2	50.4	60.4				Schade & Crutzen ¹⁰
Lund, Sweden	Agriculture	55.71	13.19	Jan–May 1991	GC		0.5 ± 0.3	5.2 ± 0.3		1.4 ± 0.3	<0.5	Grönberg et al. ⁶⁷
Lamont, OK, USA	Agriculture	36.60	–97.48	Apr–May 2013	DMSO AmPMS	4	14	35	150	98	20	Freshour et al. ⁶⁸
Wangdu, China	Rural	38.71	115.16	Dec 2018–Jan 2019	H ₃ O ⁺ (H ₂ O) _n Vocus PTR-TOF-MS		14.6 ± 14.9					Wang et al. ²⁰²
Södra Sandby, Sweden	Rural	55.71	13.34	May–Jul 1991	GC		1.8 ± 0.6	41 ± 14		1.7 ± 0.4	0.7 ± 0.5	Grönberg et al. ⁶⁷
Lewes, DE, USA	Coastal	38.78	–75.16	Jul–Aug 2012	DMSO AmPMS	5	28	6	3	<1.0	2	Freshour et al. ⁶⁸
Oahu, Hawaii, USA	Coastal	19.89	–155.66	Nov 1985	GC		2.27	0.43				Van Neste et al. ¹⁷⁶
Finokalia, Greece	Coastal	35.33	25.67	2023–2016	HPLC-MS		7.8 ± 12.0 ^a	7.5 ± 12.4		1.1 ± 3.5	0.6 ± 0.4	Tzitzikalaki et al. ²⁰³
Arabian Sea	Ship	12.25	64.33	Aug–Oct 1994	FIGD-IC	2.91	0.97	0.01				Gibb et al. ⁵⁰
Narragansett, RI, USA	Coastal	41.45	–71.44	Jul–Aug 1985	GC	1.34	5.97	1.43				Van Neste et al. ¹⁷⁶
Kent, OH, USA	Semirural	41.15	–81.36	Nov 2011	Acetone/Ethanol CIMS	<12	8 ± 3	16 ± 7				Yu and Lee ²⁰⁴
Zonguldak, Turkey	Urban	41.31	31.85	Aug–Sep 2013	Ethanol CIMS	1–4	<0.5	5–10		10–50	<1.4	You et al. ²⁰¹
				Oct 2006–Apr 2007	GC-MS	1.02	1.41					Akytüz ²⁰⁵ *
				Mar–Sep 2007								
Houston, USA	Urban	29.72	–95.35	Ocr-2022	Ethanol CIMS	4 ± 2	6 ± 2	31 ± 9	79 ± 30	33 ± 12	12 ± 4	Tiszenkel et al. ²⁰⁶
Albany, NY, USA	Urban	42.65	–73.75	Sep–Nov 2001	HPLC-MS			67.65				Huang et al. ²⁰⁷
Shanghai, China	Urban	31.29	121.50	Mar–Sep 2007	Ethanol HR-TOF-CIMS	15.7 ± 5.9	40 ± 14.3	1.1 ± 0.6	15.4 ± 7.9	3.4 ± 3.7	3.5 ± 2.2	Yao et al. ⁶⁷ **
Nanjing, China	Urban	32.20	118.70	Aug–Sep 2012	H ₃ O ⁺ (H ₂ O) _n HR-TOF-CIMS	0.1–18.9	0.1–29.9	0.1–9.3				Zheng et al. ⁹
Beijing, China	Urban	40.56	116.5	Oct 2018–Mar 2019	TOF-CIMS		2.7 ^s					Deng et al. ²⁰⁸
Atlanta, GA, USA	Urban	33.80	–84.4	Oct 2018–Jan 2019	H ₃ O ⁺ (H ₂ O) _n HR-TOF-CIMS		5.2 ± 4.3 ^b					Zhu et al. ²⁰⁹
				Jul–Aug 2009	DMSO AmPMS	<0.2	<0.2	4–15	~4	10–30	3–25	Hanson et al. ²¹⁰
Minneapolis, MN, USA	Urban	44.96	–93.24	Oct 2012	DMSO AmPMS	4	41	19		20	5	Freshour et al. ⁶⁸
Toronto, Canada	Urban	43.66	–79.39	Jun–Jul 2009	AIM-IC		<2.7				<2.7	VandenBoer et al. ⁶⁴
Helsinki, Finland	Urban	60.20	24.96	Aug 2011	AIM-IC		23.6 ^b			0.3–1.3	0.1–0.16	Hellén et al. ²¹¹
Chino, California, USA	Livestock	33.97	–117.60	Aug–Sep 2013	IC			1.3–6.8 ^a				Dawson et al. ¹¹
Yuba, California, USA	Livestock	39.26	–121.35	Oct–Nov 2001	GC-MS		26.4–181.5 ^a			61.6–74.8 ^a		Rabaud et al. ¹³

Table 1 (continued) | Summary of measurement location, time period, technique and concentrations of gas-phase amines in diverse locations

Location	Type	Lat. (deg.)	Lon. (deg.)	Time Period	Technique	Concentrations (pptV)					Reference
						MMA	DMA	TMA	EA	DEA	TEA
Antarctic Peninsula	Polar	−62.66	−60.39	Jan–Feb 2019	Nitrate CI-API-TOF		1.25×10^{-11}		1.15×10^{-11}		Bréan et al. ⁸³

^aMean of reported concentrations for summer and winter; ^b8 h (4 am to 8 pm) average; ^cmean of throughout the measurement period and its standard deviation; ^dmedian value; ^eppbV; ^fions/s; ^gDMA + EA; ^hTMA + DEA; ⁱC₁; ^jdetection limit integration time 5 sec
MMA Monomethylamine, DMA Dimethylamine, TMA Trimethylamine, EA Ethylamine, DEA Diethylamine, TEA Triethylamine, DMSO dimethyl sulfoxide, CI-API-TOF Chemical Ionization Atmospheric Pressure interface Time-Of-Flight mass spectrometer, CIMS Chemical Ionization Mass Spectrometer, MARGA-MS Measuring Aerosols and Gases in Ambient Air coupled with mass Spectrometer, LC-MS Liquid Chromatography-Mass Spectrometry, AM-IC Ambient Ion Monitor-Ion Chromatography system, GC Gas Chromatography, AmPMS Ambient pressure Proton transfer Mass Spectrometer, FIGD-IC Flow injection gas diffusion coupled to IC, PTR-TOF-MS Proton-Transfer-Reaction Time-Of-Flight Mass Spectrometer, HR-TOF-CIMS High-Resolution Time-Of-Flight CIMS, HPLC High-Pressure Liquid Chromatography.

humidity, but the addition of further DMA up to 140 pptV produces a negligible increase in nucleation rate⁴. Other studies showed that for SA concentrations $<10^7$ molecules cm^{-3} and DMA >10 pptV, nucleation proceeds at or near the kinetic limit, indicating that collision between SA molecules and clusters associated with DMA minimizes evaporation compared to NH_3 ^{5,20}. The formation of the SA-base cluster and their subsequent stabilization by the presence of amines (e.g., DMA) and high NH_3 concentrations at lower temperatures have been shown to drive NPF in polluted environments^{49,87–89}. Studies in Chinese megacities showed that neutral SA-water-DMA nucleation was the dominant pathway for the formation of sub-3nm particles^{24,89}. The molecular level ternary nucleation mechanism involving SA, DMA and TMA showed that TMA can accelerate the SA-DMA-based NPF by 50–100%, with its contribution up to 43% to the particle formation rate, in Beijing⁹⁰. In contrast, measurements at the Finnish Antarctic research station (Aboa), approximately 130 km inland from the Southern Ocean coast, indicated that ion-induced SA- NH_3 nucleation drives particle formation⁹¹ indicating an insufficient source of amines at the site.

Terrestrial vegetation is a dominant source of VOC emissions to the atmosphere⁹². Several studies have shown that organic compounds derived from various precursor VOCs are involved in particle formation with SA^{62,93–96} and that they are a dominant source of SOA⁷⁷. According to a theoretical study by Zhao et al.⁹⁷, 2-methylglyceric acid (MGA), an isoprene derivative, can efficiently form heterodimers with SA and MSA, thereby contributing to NPF. However, it remains unclear whether adding a base molecule (such as NH_3 or amines) to this binary system would enhance or inhibit the formation of stable clusters. Kulmala et al.⁹⁸ showed that the growth rate of particles in Beijing did not correlate with organic compounds and SA concentrations summed, indicating that either multiphase chemical reactions assist the growth rate or higher volatility compounds condense onto nanoparticles. On the contrary, there exists only one study showing pure biogenic nucleation from HOMs in the nucleating cluster without base molecules (NH_3 or amine) at a high-altitude free-tropospheric site (3580 m a.s.l.) in Jungfrauoch, Switzerland⁸⁵. Nevertheless, organic compounds are important for nanoparticle growth and hence for the survival of newly formed particles in the atmosphere^{47,78}. It is well recognized that atmospheric SA concentrations are often insufficient to explain the observed growth of nanoparticles^{99,100} and organic compounds are believed to play an important role in the growth of nucleation mode particles^{101,102}. It is plausible that the particle-phase reactions between amines and organic acids, along with a synergistic effect involving carbonyls, may contribute to nanoparticle growth. Organic molecules containing carbonyl functional groups, such as glyoxal and methyl glyoxal, are formed through the oxidation of both biogenic^{103,104} and anthropogenic^{105,106} compounds. These organic molecules can react with primary and secondary amines, such as monomethylamine (MMA) and DMA to form imine and enamine compounds, as well as polymerized products^{107,108}, suggesting particle-phase reactions between amines and organic acids. Furthermore, Chen et al.¹⁰⁹ reported significant enhancements in diethylamine (DEA) in particles, attributed to increased aerosol water content and aerosol acidity in an urban area of Chongqing, China, during winter.

At present, no single organic compound has conclusively demonstrated strong neutral nucleation potential¹¹⁰, particularly in clustering with base molecules (amine or NH_3), except for HOMs containing an amino group that can contribute to methanesulfonic acid (MSA)-based NPF due to their high hydrogen bonding capacity¹¹¹. However, the effect of organics on the MSA-amine system is not well understood in the continental atmosphere. Such nitrogen-containing organics suggest the involvement of amines in cluster formation and warrant thorough investigations in future studies. The specific contributions of amines, NH_3 , and organic compounds to the growth of clusters larger than 3 nm diameter remain unclear in continental environments due to the lack of suitable methods for measuring trace amounts of these bases.

Table 2 | Summary of measurement location, time period, technique and concentrations of particle-phase amines in diverse locations

Location	Type	Lat. (deg.)	Lon. (deg.)	Time Period	Technique (particle range)	Concentrations (ng m ⁻³)					Reference	
						MMA	DMA	TMA	EA	DEA	TEA	ΣAmines
Hyttälä, Finland	Forest	61.51	24.17	Mar–Dec 2015	MARGA-MS (PM _{1.0})	<2.4–61.2	<3.1–55.5					Hemmlä et al. ⁷⁰
Egbert, Canada	Agriculture	44.23	–79.78	Oct–Nov 2010	AIM-IC (PM _{2.5})		0.1 ± 0.2	1 ± 0.6 ⁸				VandenBoer et al. ⁶⁵
Antarctic Peninsula	Ship	–60.0	–45.0	Jan–Feb 2015	ATOFMS (PM _{2.5})		2 ± 0.4	0.8 ± 0.3		3.8 ± 1.0		Dall’Osto et al. ¹¹⁶
North Atlantic Ocean	Ship	40.0	–40.0	Jun–Sep 1998	HPLC-MS (PM _{2.5})							7.6 Gorzelska & Galloway ¹¹⁴
Yellow Sea & Bohai Sea, China	Ship	35.49	123.79	May 2012	IC (PM _{1.0})		198.4	425.9				Hu et al. ²¹²
				Aug 2015	IC (PM _{1.0})		23.44 ± 12.62	17.73 ± 5.9				Xie et al. ²¹³
South Yellow Sea, China	Ship	34.79	123.72	Aug 2015	IC (PM _{1.0})		41.9 ± 7.2	44.9 ± 7.1				Yu et al. ²¹⁴
Arabian Sea	Ship	12.25	64.33	Aug–Oct 1994	FIGD-IC (PM _{1.0})		10.8	0.47				Gibb et al. ⁵⁰
Mace Head, Ireland	Coastal	53.32	–9.90	Apr–Sep 2006	IC (PM _{1.0})		9.91					Facchini et al. ⁵¹
Finokalia, Greece	Coastal	35.4	26.0	2005–06	IC (PM _{1.0})		9.01					Violaki & Mihalopoulos ²¹⁵
Sao Vicente Island, North Atlantic	Coastal	16.86	–24.86	Dec 2007	IC (PM _{1.0})	0.02	0.22			0.06		Müller et al. ¹⁴²
				Nov 2011, 2013	IC (PM _{1.0})	0.2	5.6			3.9		van Pinxteren et al. ⁵²
East China Sea, China	Ship	30.39	125.91	Jun 2016	IC (PM _{1.0})		30.20 ± 9.46	11.82 ± 6.5				Xie et al. ²¹³
				Apr–May 2018	AIM-IC (PM _{2.5})		<4 to 100	<2 to 21				Chen et al. ²¹⁶
Huaniao Island, China	Coastal	30.85	122.69	Jan 2013	PHLC/FLD (PM _{2.5})	4.76	0.82					Huang et al. ²¹⁷
Marina, California, USA	Ship	37.5	–123.0	Jul–Aug 2012	IC (PM _{1.0})		2.3–70.3					Youn et al. ²⁸
Jeju Island, South Korea	Coastal	32.48	126.26	Mar–Apr 2001	HPLC-MS (PM _{2.5})	13.5			3.1		19.6	Yang et al. ²¹⁸
Chongming Island, China	Coastal	31.73	121.21	May–Aug 2018	IC (PM _{2.5})	15.0 ± 15.0	6.3 ± 6.9	20.4 ± 30.1		4.0 ± 5.9		Du et al. ²¹⁹
Qingdao, China	Coastal	36.34	120.67	Nov–Dec 2019	UHPLC-MS (PM _{2.5})	6.9 ± 2.0	86.3 ± 20.2	5.9 ± 4.8	2.4 ± 1.3		8.7 ± 3.9	132.9 ± 31.3 Liu et al. ⁷²
				Jan–Feb 2019	UHPLC-MS (PM _{2.5})	8.5 ± 2.7	58.7 ± 25.8	10.9 ± 8.0	2.7 ± 1.1	8.4 ± 4.4		130.7 ± 51.9 Liu et al. ⁷²
				Dec 2019	AIM-IC (PM _{2.5})		65 ± 68	280 ± 180				Chen et al. ⁷³
Wangdu, China	Rural	38.71	115.16	Dec 2018–Jan 2019	IC (PM _{2.5})		<4 to 100	<2 to 21		206 ± 292 ⁴⁴		Feng et al. ²²⁰
Kobe, Japan	Roadside	34.69	135.19	Oct 1998	GC-MS (PM _{1.0})	1.34	3.82	0.11				Suzuki et al. ²²¹
Qingdao, China	Urban	36.18	120.58	May–Jun 2013	IC (PM _{1.0})		4.95	5.20				Hu et al. ²¹²
Nanjing, China	Urban	32.06	118.77	Feb 2001	HPLC-MS (PM _{2.5})	19.0			8.7		33.5	Yang et al. ²²²
Guangzhou, China	Urban	23.12	113.25	Sep–Oct 2014	GC-MS (PM _{1.0})	51.8 ± 13.9	19.0 ± 3.80		4.19 ± 0.56	5.41 ± 1.59	108.7 ± 21.6	Liu et al. ²²³
Xi’an, China	Urban	34.25	108.97	2008–2009	HPLC (PM _{2.5})	20.28			2.23			Ho et al. ²²⁴
				Jan–Feb 2013	HPLC (PM _{2.5})		24.7 ± 9.7	3.8 ± 1.6		12.6 ± 5.6	2.0 ± 0.8	105.3 ± 41 Ho et al. ²²⁵
Tampa, FL, USA	Urban	27.78	–82.54	Jul–Sep 2005	IC (PM _{2.5})		31.11 ± 27.9					Calderón et al. ²²⁶

Table 2 (continued) | Summary of measurement location, time period, technique and concentrations of particle-phase amines in diverse locations

Location	Type	Lat. (deg.)	Lon. (deg.)	Time Period	Technique (particle range)	Concentrations (ng m ⁻³)					Reference		
						MMA	DMA	TMA	EA	DEA	TEA	ΣAmines	
Beijing, China	Urban	39.90	116.41	Jan–Feb 2013	HPLC-MS (PM _{2.5})	31.0 ± 10.8	4.3 ± 1.7		14.8 ± 6.5	2.1 ± 0.9		133.2 ± 52.8	Ho et al. ²²⁵
Xiamen, China	Urban	24.47	118.08	Jan–Feb 2013	HPLC-MS (PM _{2.5})	10.2 ± 2.0	1.7 ± 0.4		5.3 ± 1.4	0.9 ± 0.2		53.7 ± 13.2	Ho et al. ²²⁵
Hong Kong, China	Urban	22.31	114.16	Jan–Feb 2013	HPLC-MS (PM _{2.5})	12.1 ± 2.4	1.5 ± 0.5		4.5 ± 1.1	0.9 ± 0.2		45.7 ± 9.8	Ho et al. ²²⁵
Shanghai, China	Urban	31.30	121.5	Jul–Aug 2013	IC (PM _{1.0})	7.7 ± 5.0	13.4 ± 6.5		9.7 ± 12.7				Tao et al. ⁵³
				2013–2014	IC (PM _{2.5})	15.56					16.0		Zhou et al. ^{227*}
Zonguldak, Turkey	Urban	41.31	31.85	Oct 2006–Apr 2007	GC-MS (PM _{2.5})	4.48 ± 1.75	4.58 ± 2.28		4.37 ± 2.27	4.11 ± 1.75			Akyüz ²⁰⁵
				Mar–Sep 2007	GC-MS (PM _{2.5})	2.33 ± 1.30	2.79 ± 1.55		0.19 ± 1.13	2.82 ± 1.76			Akyüz ²⁰⁵
Yangzhou, China	Urban	32.38	119.4	Apr–Nov 2016	HPLC-MS (PM _{2.5})	1.42 ± 1.34	3.62 ± 3.94		12.6 ± 10.5				Cheng et al. ²²⁸
Toronto, ON, Canada	Urban	43.66	−79.39	Jun–Jul 2009	AIM-IC (PM _{1.8})			<LOD-16 ^a					VandenBoer et al. ⁵⁴
Tucson, AZ, USA	Urban	32.22	−110.9	Jul 2012–Jun 2013	IC (PM _{1.0})		9.3–20.5						Youn et al. ²⁸
Newfoundland, Canada	Wildfire	53.13	−57.66	Jul 2013	IC (PM _{2.5})			9 ± 7		1.6 ± 0.8	0.2 ± 0.1		Place et al. ²²⁹

^a annual average; ^{*}DEA + TEA; [†]TMA + DEA; ΣAmines: total mass of aliphatic amines.

ATOFMS aerosol time-of-flight mass spectrometer, UHPLC Ultra-high performance liquid chromatography, FLD fluorescence detection.

Coastal and marine atmospheres

Amines in marine areas are released through various mechanisms, such as direct emissions from phytoplankton, excretion or decomposition of marine organisms^{50,112,113}, bubble bursting at the air-sea interface¹¹⁴, or biological degradation of quaternary nitrogen osmolytes (R_4N^+ , where R is an alkyl substituent on the N atom)¹¹⁵. Direct observations of both gas- and particle-phase amines in remote marine and polar atmospheres are extremely scarce (Fig. 1), and mostly reported from coastal environments which are also affected by anthropogenic emissions of various gaseous precursors from coastal continental areas, including amines. The sea ice microbiota and plankton in the marginal ice zones and adjacent open ocean of the Weddell Sea in the Antarctic have been reported to be important sources of volatile sulfur and alkylamines¹¹⁶, along with NH_3 from penguin and seabird colonies in coastal Antarctica^{117,118}. Measurements at the Spanish research station on the south coast of Livingston Island in the South Shetland Islands revealed abundant SA-amine peaks during NPF events⁶³. Observations in the eastern Pacific Ocean indicated that an increase in gas-phase amines and organosulfur compounds has a profound impact on both particulate chemical composition and cloud properties¹¹⁹. Previous studies have shown that MSA, approximately 10% to 100% of that of SA over marine regions, contributes to particle formation and growth in coastal and oceanic regions^{120,121}. While MSA, SA, and NH_3 /amines coexist in the marine atmosphere, how these compounds interact to form particles is not well understood. Given the exceedingly low binary nucleation efficiency of MSA- H_2O under typical atmospheric conditions^{122,123}, MSA's contribution to NPF is predominately determined by the enhancing effect of other species, particularly atmospheric bases such as NH_3 and amines^{124,125}. Laboratory experiments have shown that the reactions of MSA and DMA can produce new particles, even in the absence of SA^{21,126}, and MSA can contribute to the particle growth^{121,127}. As a result, methanesulfonate clusters grow more rapidly in marine air with the addition of a DMA molecule compared to an NH_3 molecule. Previous studies showed that amines, such as DMA, displace NH_3 with near-unit reaction efficiency on the surfaces of ammonium methanesulfonate clusters¹²⁸. This indicates that dimethylammonium methanesulfonate salts are preferred over ammonium methanesulfonate salts in small clusters. Such an exchange of ammonium in sub-micron aerosols by aminium salt is expected to occur within a few hours¹²⁹. Laboratory experiments and ab initio calculations further demonstrated that the MSA-amine intermediate contributes to new particles for TMA due to its highly hygroscopic nature, whereas for MMA and DMA, water is required¹²⁴. Nucleation experiments, however, provide evidence that MSA suppresses the SA-TMA pathway due to the steric hindrance of the MSA and TMA, while it enhances the SA-MMA pathway via the formation of the SA-MSA heterodimer¹³⁰. Quantum chemical calculations and cluster kinetic modelling further highlight that the MSA-DEA system has a relatively stronger nucleation potential in marine environments than MSA-DMA and SA-MMA systems¹³¹. The hydrogen-bonding capacity of substituted MSA-amine species may contribute to increased water uptake, with an amine-specific effect on particle hygroscopicity and growth.

Oxalic acid, formed via oxidation of glyoxal, a highly prevalent dicarboxylic acid originating from both natural marine and anthropogenic continental sources, has been identified in tropospheric aerosols^{132,133}, which was observed to have a stronger binding affinity with MMA than with NH_3 ^{126,134,135} and TMA¹³⁶. The addition of oxalic acid to the MSA-MMA system reasonably enhances NPF, likely due to increased hydrogen bonding capacity or promoting proton transfer. However, it does not affect the MSA-MMA- H_2O system because water at atmospherically relevant concentrations overwhelms the contribution of the much smaller concentrations of organics¹²⁶. It is important to note that both the basicity and hydrogen-bonding capacity are key factors in determining the enhancing effect of a base molecule on MSA-induced NPF^{125,137–139}. Nevertheless, field measurements have reported a strong correlation between the methane sulfonate ion and particle number concentrations^{7,140}, as well as between particle growth and MSA concentrations¹⁴¹. Several studies have observed the presence of amine, NH_3 , and MSA in sub-micron particles in coastal

environments^{51,119,142,143}, but observations of these compounds in ultrafine particles (diameter less than 100 nm) or nanoparticles (diameter less than 25 nm) are practically non-existent.

While SA, MSA, oxalic acid, and nitric acid all require an additional vapor (NH_3 or amines) to form particles, organic compounds such as HOMs⁶ and iodine^{144–146} can form particles on their own under specific atmospheric conditions. The potential of iodine compounds to form new particles has been shown in laboratory experiments^{144,147,148} and field studies^{149–153}. Among iodine compounds, iodic acid (IA, HIO_3) has been identified as the key driver of NPF in the coastal, open ocean, and ice-covered polar regions^{146,151,154,155}. However, atmospheric observations of IA cannot be correlated with predicted particle formation rates¹⁵³. Huang et al.¹⁵⁶ demonstrated that heterogeneous reactions between higher iodine oxides and condensing alcohols or carbonyls from the oxidation of marine VOCs lead to the formation of low-volatility oxidized organics for newly formed ultrafine particles. These organic acids can then react with basic molecules, such as amines, accelerating early particle growth to form highly hygroscopic salts. Semicontrolled seawater-air enclosure measurements in the Bay of Calvi (Corsica, France) by Sellegri et al.¹⁵⁷ showed that iodine-containing species are the major precursors for new cluster formation and subsequent growth to larger diameters, which was mainly driven by Chlorophyll *a* (Chl *a*) related organic precursors. The high correlation between particle growth rate (1–10 nm) with TMA and the presence of amines in particles larger than 70 nm diameter and Chl *a*, plausibly suggests iodine-amines clustering. Using a quantum chemical approach and Atmospheric Cluster Dynamics Code (ACDC), Ning et al.¹⁵⁵ showed that DMA can structurally stabilize IA through hydrogen and halogen bonds, and the clustering process is energy barrier-less. Their study further showed that DMA can enhance the formation rate of IA clusters by five orders of magnitude, demonstrating its much greater efficiency in promoting IA cluster formation compared to NH_3 . The IA-DMA clustering pathway exhibits greater stability and energy favourability compared to both the pure-IA and IA- NH_3 systems. Consequently, these stable IA-DMA clusters could serve as abundant seeds for the marine NPF. Several precursors for IA have been proposed, including hydrated iodine atoms^{153,158}, hydrated IO radicals¹⁵⁸, iodine dioxide (OIO) radicals¹⁵⁹, and larger iodine oxides (I_2O_3 , I_2O_4 and I_2O_5)^{158,160,161}. However, these mechanisms remain speculative and lack experimental confirmation, leaving atmospheric IA observations unexplained. Nevertheless, global iodine emissions have tripled over the past 70 years and are projected to rise further as sea ice thins¹⁶². The potential increase of iodic-induced CCN concentrations in the Arctic region could increase longwave radiative forcing from clouds and provide a positive feedback mechanism that would further accelerate the loss of sea ice. However, the potential impact of other nucleation precursors on IA-DMA and their molecular mechanism remains unclear and non-verified with field observations.

Amines in models: Efforts and knowledge gaps in governing processes

The development of process-based numerical models, involving many different acid and base molecules, organics, water, and ions, is crucial for integrating intricate and non-linear airborne aerosol production processes in regional and global climate models to simulate and predict their impact on climate¹⁶³. However, modelling global amines remains a challenge due to the scarcity of ambient measurements (Fig. 1). Quantum chemical calculations suggest that alkylamines efficiently stabilize SA clusters¹⁶⁴, and these clusters are further grown to larger diameters by abundant concentrations of VOCs⁷⁷. A process model based on cluster kinetics and quantum chemistry has demonstrated that the collision of SA and amine (e.g., DMA) clusters is the dominant mechanism to trigger NPF events in Beijing¹⁶⁵. Although amines can enhance aerosol formation rates by up to a thousand-fold compared to NH_3 ^{4,44}, model simulations suggest that nearly all present-day atmospheric SA nucleation involves NH_3 or biogenic volatile organic compounds^{47,166}. Primary emissions and airborne secondary production are difficult to disentangle as these are intricately coupled, and therefore, current

global climate models fail to produce realistic particle formation and growth rates.

Myriokefalitakis et al.³⁵ used a three-dimensional chemical transport model to estimate SOA formation, focusing exclusively on the potential contribution of the oceanic amine sources, assuming that amines account for one-tenth of the ocean-derived NH_3 . Yu and Luo³⁴, on the other hand, modelled the global distributions of gas-phase amines by adjusting emission ratios for particular amines (such as MMA, DMA and TMA) to investigate the primary mechanism regulating amine concentrations (including emission, transport, oxidation, deposition, and aerosol uptake), but validation of simulated concentrations of amines with observations was hindered by the limited availability of ambient measurement data^{33,34}. The global aerosol-climate model ECHAM-HAMMOZ, which incorporates amine-enhanced NPF parameterization and a kinetic nucleation parameterization, showed that amine-enhanced NPF is limited to areas near the source regions of amines due to their short gas-phase residence time in the atmosphere. However, the simulated gas-phase concentrations of amines were mostly underestimated compared to ambient observations³³. Their study further showed that kinetic nucleation, which depends solely on SA concentration, produces particles more uniformly globally due to the long-range transport of SO_2 . On the contrary, the baseline scenario simulation, using a coupled zero-dimensional cluster-aerosol dynamic and technology-based emission projection model, showed that a 10–20% increase in amine concentrations and a doubling of SO_2 results in more than a 100% increase in particle formation rate across NPF events, outcompeting increase in condensation sink¹⁶⁷. SO_2 concentrations are declining in Europe, the USA, and China, due to an economic slowdown and government efforts to restrain anthropogenic emissions^{168,169}, which could also alter the SA-amines pathways near and away from the SO_2 sources. Julin et al.¹⁷⁰ used the chemical transport model PMCAMx-UF, including the NH_3 -DMA pathway as well as the condensation of organic compounds onto particles to simulate particle number concentrations, and demonstrated that a decrease in NH_3 and amines concentrations reduce total particle number concentrations by 10–50% across Europe, however, the amine species were treated as a surrogate compound representing MMA, DMA, and TMA, with the assumption that the amine emissions are equal to the NH_3 emissions scaled by a factor of 0.0057. Subsequently, Mao et al.¹⁷¹ and Li et al.¹⁷² derived amine-to- NH_3 mass emission ratios specific to the source to distinguish between different amines, including sources such as agriculture, residential, transportation, chemical industry and other industries, while they exclude other prominent sources such as livestock operations and marine in their simulation. Machine learning model constructed to study enhancing potential of amines showed that the nucleation rate of DEA, mainly emitted from ethanol gasoline vehicles, with SA is 3–7 times higher than that of DMA, which was thought to be key SA-driver nucleation¹⁷³. A quantitative structure-activity relationship (QSAR) model constructed for 63 precursors of IA-containing dimer clusters demonstrated that DEA exhibits the highest potential to enhance IA-induced nucleation at a mere sub-pptV level (0.1 pptV), with nucleation rates comparable to the IA-iodous system¹⁷⁵.

Furthermore, Zhang et al.⁷⁶ developed an emission inventory for amines using multi-source data sets from marine biological emissions that account for air-sea exchange fluxes. Their findings revealed that marine regions can serve as either a sink or a source for amines, depending upon ambient conditions such as the atmospheric oxidation capacity^{50,52}. The transfer of NH_3 and MMA together with DMS, across the air-sea interface to the atmosphere is suggested to play an important role in the regulation of aerosol pH, cloud water and rainfall^{174,176}. None of the above processes is currently incorporated into global climate models, and the gridded emission inventories predominately rely on a fixed amine-to- NH_3 ratio method^{33–35} and lack highly spatially and temporally resolved amine emissions, except for the source-dependent amine-to- NH_3 ratio established by Mao et al.¹⁷¹ and subsequently used in Energy Exascale Earth System Model (E3SM) version-1 incorporated with different NPF mechanisms¹⁷⁷ and a three-dimensional Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) regional model¹⁷⁸. More recently, quantum

chemical calculations, atmospheric cluster dynamic simulations, and WRF-Chem simulations showed that IA has great potential for participation in the SA-DMA nucleation process not only in marine NPF but also in continental NPF¹⁷⁹, but this has not yet been validated with ambient measurements.

Recent computational and theoretical studies indicate that amines undergo autooxidation in ambient conditions, resulting in previously unidentified reaction products^{23,180}. For instance, the autooxidation of TMA generates a completely novel type of nitrogen-containing compound, hydroperoxyl amides. Furthermore, atmospheric autooxidation pathways for the production of highly oxidised organic molecules involving peroxy radicals have been identified to oxidise amines at a rate that competes with bimolecular reactions with oxidants, such as oxides of nitrogen²³. However, unimolecular autooxidation reactions of VOCs, and especially amines that compete with bimolecular reactions in atmospheric conditions are underestimated²³. Our understanding of the oxidation of various classes of amines and the chemical pathways leading to the formation of oxidized compounds from bases in the atmosphere is still extremely limited. While strong amines, even at low concentrations and when undetected in the smallest clusters, can be crucial for particle formation, the contribution of different amines and whether they compete among themselves or with other bases have yet to be quantified. Until now, climate models have not yet factored in amines, their chemical properties, or their effects on particle formation and growth, which is the major missing link in the Earth's climate.

Atmospheric amines: Implications to climate and public health

Amines play a key role in aerosol formation and growth through acid-base neutralization^{4,5,22,24,28}, potentially leading to sizes capable of acting as CCN, thus contributing to aerosol cooling effect via cloud microphysical processes. However, they can also act as precursors to climate-warming agents like methane and brown carbon, which may offset the cooling effect. Furthermore, CCS facilities utilize amine scrubbing technology to remove carbon from the air to combat climate change. However, CCS facilities release carcinogenic amines into the atmosphere, raising critical risk to public health.

Amines, such as TMA, serve as precursors for methane production over marine regions. Under oxygen-deprived conditions, microbial conversion of TMA originating from the degradation of quaternary amine precursors can account for up to 90% of methane emissions from salt marsh sediment or slurries^{181,182}. Hence, amines found in marine ecosystems¹⁸³ from surface seawaters to deep sediments⁵⁰ have critical importance to global warming over oceans via the release of methane through methanogenesis in marine/coastal sediments¹⁸⁴. Several studies report the formation of light-absorbing BrC¹⁸⁵ via aqueous-phase reactions involving glyoxal, methylglyoxal, and formaldehyde^{26,186}, as well as the oxidation of ethylamine (EA) mediated by nitrate (NO_3^-) photolysis¹⁸⁷. Although BrC produced from the particle-phase reaction of methylglyoxal with MMA can contribute to atmospheric warming¹⁸⁸, the BrC formation via aldehyde-amine-ammonium sulfate browning reactions is about ten times lower than that generated from wood burning, which accounts for <10% of light absorption in the atmosphere²⁶. These findings suggest that specific reaction pathways involving particular amines may have differential effects on the climate.

Limiting the rise in global temperatures to less than 2 °C relative to pre-industrial times requires reductions in human-made greenhouse gas emissions, mainly carbon gases². Several technologies for capturing and separating carbon dioxide (CO_2) have been applied¹⁸⁹, such as chemical absorption via amine scrubbing, separation by adsorption^{190,191}, membrane separation¹⁹², and calcium chemical looping¹⁹³. However, these technologies have several drawbacks, notably the degradation of the amines occurring at relatively low temperatures, particularly in the presence of oxygen in the inlet flue gas stream¹⁹⁴. In the case of CCS facilities, 2-aminoethanol (MEA) is used as an absorption solvent in post-combustion capture^{195,196}, leading to the release of 80 tons of MEA into the atmosphere for every 1 million tons of CO_2 removed annually^{39,197}. A modelling framework demonstrated that

realistic emission of amines from a typical post-combustion CO₂ capture results in the sum of carcinogenic amines (nitrosamines and nitramines) of 0.6–10 pg m⁻³ and 0.04–0.25 ngV L⁻¹ in the ground-level air and drinking water, respectively, which are below the current safety guideline by National Institute of Public Health (NIPH) for human health³⁶, while the deposition of MEA into small lakes in the Norwegian west coast could exceed toxicity limits for aquatic organisms³⁹. This suggests that if the technology were widely deployed, a substantial amount of MEA would be released into the atmosphere from post-combustion CO₂ capture units which could have implications to both climate and public health.

Summary

Despite vigorous research over the last more than three decades, a holistic understanding of the formation mechanisms of airborne nanometre-sized aerosols and their growth to CCN, toxicity and overall effect on the climate remain elusive. This is because the understanding of aerosol precursors is built on measurements of strictly selected groups of acidic or highly oxygenated organic compounds due to technological limitations. In laboratory conditions, amines are known to enhance aerosol formation rates by over a thousand-fold compared to ammonia, yet their roles in ambient aerosol production and interactions with other bases remain poorly quantified. Only a few pptV mixing ratios of DMA are needed to saturate the formation rate in atmospherically relevant SA concentrations. Mixing ratios below 1 pptV of DMA can enhance particle formation rates by stabilizing the clusters and minimizing evaporation compared to ammonia. This process seems to be valid for polluted areas, where neutral SA-DMA (-water) nucleation is found to produce sub-3 nm particles efficiently. Observations from the boreal forest or other continental areas do not seem to support a major role for DMA in SA stabilizing base compound but ammonia is observed to take the leading role. The exact contributions of DMA, ammonia and HOMs to the growth of sub-3 nm clusters are yet to be elucidated in most environments. The challenge to achieve this information is a major one; the current DMA mixing ratios measured from the same field site in Finland vary from ppqV to sub-ppbV levels, pointing toward colossal measurement errors in some or all used methods. Due to the technological challenges and lacking methodologies, only a minority of atmospheric bases have been quantified, and their properties and reactions are not understood well.

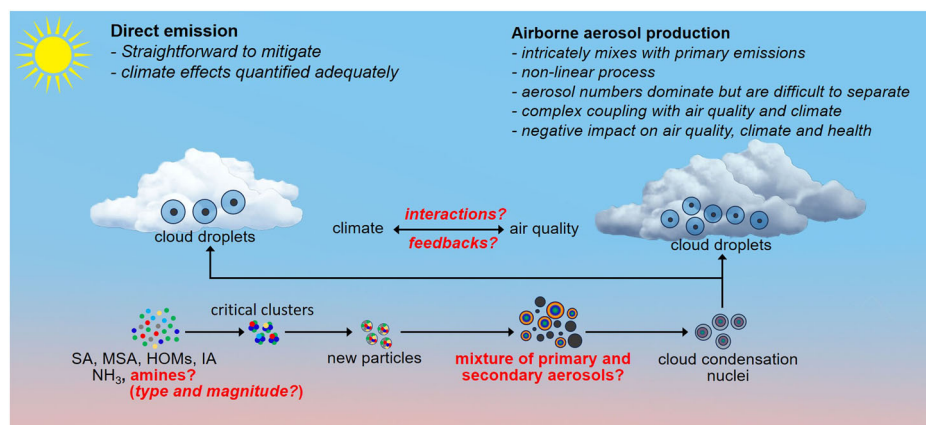
Further, ambient measurements of atmospheric amines are almost solely conducted in the Northern Hemisphere as shown in Fig. 1. However, there are limited data from the rest of the world, and no measurements exist from the continents of Africa, South America, Australia, or from the Arctic regions. Current studies are predominantly concentrated in urban and coastal areas, with minimal data from rural, agricultural, and polar environments, as well as regions with intensive livestock activities, which are believed to be a substantial source of amine emissions. Additionally, CCS technologies increasingly utilize specific amines to reduce carbon emissions from fossil-fuel-based energy and industrial sectors. However, detailed

quantification of amine emissions from CCS facilities is still lacking, which limits our understanding of the broader environmental and climatic implications of amines, particularly within the context of the climate-air-health nexus.

Ambient base concentrations are only reported in polluted environments due to extremely low concentrations elsewhere in the atmosphere. Molecular clusters consisting of a few aerosol precursor molecules exist at mixing ratios approximately 1000 times lower (e.g., the maximum atmospheric concentrations of SA have been measured at $<3 \times 10^7$ molecules cm⁻³, approximately 1.2 pptV), which makes their analysis extremely challenging. Current base molecule measurements suffer from contamination and memory effects that increase detection limits in field conditions, insufficient resolution mass spectrometers creating fatal uncertainty in peak identification and the lack of analytical standards and zero samples. This perspective article underscores the need for novel measurement technologies to measure sub-pptV concentrations of amines in the gas-phase as well as in nanoparticles. Furthermore, no single detection method is adequate to capture the full spectrum of amine emissions, highlighting the need for hybrid approaches that combine multiple techniques (chromatographic, spectroscopic, etc.) for comprehensive measurements.

Direct emissions from anthropogenic activities are often straightforward to mitigate through targeted interventions and regulatory measures, although airborne aerosol production involves the intricately mixed interactions between primary emission and secondary atmospheric processes, and usually are non-linear, thereby challenging to separate from primary emissions (Fig. 2). Airborne aerosol formation is the largest source of aerosol numbers in the global atmosphere, and therefore it is critical to understand the physiochemical mechanisms driving it to correctly represent this process in climate models, and thereby future climate predictions². Very little information is available on the atmospheric bases involved in airborne aerosol production. This represents an important knowledge gap, as various bases, such as NH₃ or amines, are known to stabilize negative ion clusters^{20,25,198–200}, and results from ambient measurements are not sufficient to confirm or refute this information. The relevance of amines-driven NPF to the global CCN budget remains to be assessed due to the lack of measurements and accurate amines emission inventories. It is important to focus on reducing direct emissions of both aerosols and their precursor to effectively address their immediate impact on climate and air quality. This is because the climate and air quality represent two distinct, yet interrelated, entities. Any policy action aimed at mitigating one of these entities must consider the feedback from the other, as advancements in one entity may worsen conditions in the other. For example, the declining trend in SO₂ emissions at the Station for Measuring Ecosystem-Atmosphere Relations (SMEAR-II, Hyytiälä, Finland), resulted in reduced SA concentrations, particle formation rates and NPF frequency³⁸, and thereby lower aerosol numbers and improved air quality, which may dampen the cooling effect of aerosols. Therefore, ambient measurements integrated with laboratory experiments and theoretical studies aided with the development of the

Fig. 2 | Amine emissions and their impact on air quality and Earth's climate via airborne aerosol production. Atmospheric amines, emitted from a wide range of natural and anthropogenic sources, facilitate the fastest base-neutralization mechanism, thereby reducing the evaporation of critical clusters and promoting the formation of new particles. These newly formed particles not only intricately mix with primary particles leading to haze formation but also have the potential to enhance aerosol hygroscopicity and the activation of cloud condensation nuclei. Therefore, the complex interplay and feedback mechanisms between air quality and climate are inadequately quantified, posing significant challenges to accurately constraining their combined impacts on air quality, climate, and human health.



process-based models are required to improve the representation of amines (and other bases) in Earth system models to advance our understanding of the complex interactions of amines in atmospheric chemistry, ultimately addressing their climatic and environmental impacts more effectively.

Data availability

No new data were generated for this analysis.

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Author contributions

T.J. conceived the idea. V.P.K. wrote the first draft and revised it with critical inputs from T.J.

Competing interests

The authors declare no competing interests.

Additional information

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