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Molecular Mechanisms of Aerosol Nucleation: from CLOUD Chamber Experiments to Field Observations

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Abstract: Atmospheric aerosol particles contribute to over four million premature deaths annually and play a critical role in modulating Earth's climate. Most atmospheric particles and more than 50% of the cloud condensation nuclei are formed through a secondary process named new particle formation involving unique precursor vapors. This article summarizes current knowledge of how new atmospheric particles form, based on experiments at the CERN CLOUD chamber. While the role of sulfuric acid has long been known, other vapors like highly oxygenated organic molecules and iodine oxoacids are also important, along with stabilizers like ammonia, amines, and ions from cosmic rays. We explain how findings from CLOUD experiments help us understand particle formation in various atmospheric conditions and improve air quality and climate models.

Keywords: Chamber experiments · Ions · Molecular clusters · Particle formation rates · Vapors

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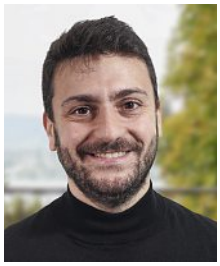

Dr. Imad El-Haddad focuses on understanding the role of anthropogenic emissions on atmospheric composition, climate, and human health. He has experience in analytical chemistry, online mass spectrometry, atmospheric reactivity, aerosol generation, atmospheric simulation chambers and flow reactors, emission characterization and chemical transport modelling. El Haddad leads the Atmospheric Molecular Processes group at the Laboratory of Atmospheric Chemistry. He is the current CLOUD team leader at the Paul Scherrer Institute, and part of the CLOUD steering committee, who is responsible for making the executive decisions on the experiment and steering it scientifically, technically, and administratively.

1. Introduction

Atmospheric nucleation is the process by which low volatility gas-phase molecules condense into small molecular clusters that can grow into new aerosol particles. This transition must overcome a Gibbs free energy barrier, which slows the nucleation rate far below the rate of molecular collision.[1] The formation of a stable critical cluster is essential, as its subsequent growth leads to new particle formation (NPF). NPF is the prime source of ultrafine atmospheric particles, accounting for more than half of the global cloud condensation nuclei (CCN).[2] NPF significantly influences cloud properties and Earth's radiative balance, providing a buffering effect that makes the climate less sensitive to variations in emissions or extreme events like wildfires.[3] NPF has been observed worldwide across diverse environments, from remote regions with limited precursors to heavily polluted megacities where high concentrations of existing particles might be expected to scavenge precursor vapors and nucleating clusters.[4] Understanding the mechanisms and precursor vapors involved in NPF is essential for accurately estimating the sensitivity of the climate to anthropogenic greenhouse gases and primary aerosol emissions.

Until a decade ago, there were enormous uncertainties surrounding NPF mechanisms. While it was clear that sulfuric acid is involved, experimental NPF rates from pure sulfuric acid differed by orders of magnitude, with some studies claiming that it could account for atmospheric observations,[5] but others not.[6] In 2009,

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the *Cosmics Leaving OUtdoor Droplets* (CLOUD) experiment at the European Center for Nuclear Research (CERN), in Switzerland, was created to address these uncertainties,[7] aiming to provide a molecular understanding of NPF mechanisms. Given its high precision and low contaminant concentrations, CLOUD is established as a world-leading experiment for studying atmospheric NPF. The experiment also extends our understanding more broadly into atmospheric chemistry and oxidation mechanisms. It currently involves a consortium of 23 institutes from 15 European countries and the U.S., with > 60 researchers working on the experiment following the model of particle physics experiments at CERN. Switzerland has a high profile at CLOUD, with both the Paul Scherrer Institute (PSI) and CERN teams as steering members.

The CLOUD experiment was originally conceived to test Svensmark's thesis about the influence of ionization from galactic cosmic rays (GCRs) on aerosols and climate.[8] In its first experimental run, CLOUD provided the first evidence that NPF rates from pure sulfuric acid are a million times slower than the boundary-layer NPF observations,[9] highlighting the role of bases and ions in stabilizing the nucleating clusters.[9,10] Later, CLOUD identified the role of additional vapors in NPF, including Oxygenated Organic Molecules (OOMs), particularly ultra- and extremely-low volatility organic compounds (ULVOCs and ELVOCs),[11] and iodine oxoacids,[12] which were observed to participate in nucleation inside a boreal forest[13] and over a peatland[14] in southern Finland. Subsequent particle growth is generally richer, often involving higher volatility organic molecules[11a,b,15] and nitric acid.[16] Usually, growth is the limiting step for the survival of freshly nucleated clusters to larger size particles that can scatter light and seed clouds.[17] This is especially important for sub-10 nm particles with low survival probability, which can be rapidly lost by coagulation scavenging to pre-existing particles, unless they grow rapidly.[18] As nucleation and growth rates are extremely dependent on the concentrations of condensing species, it is vitally important to simulate these processes at ambient, contamination-free conditions, as done at CLOUD.

Numerous comprehensive reviews can be found on atmospheric NPF, focusing on rural, mountain, and urban sites.[4,19a,b,c] However, several questions about the molecular mechanisms of ambient aerosol nucleation remain open, including which key molecular species are involved, how they interact, and how environmental factors like temperature, humidity, and ions influence nucleation rates and pathways. Additional uncertainties concern the role of organic molecules, water, and trace gases, as well as how to accurately model and experimentally validate these processes to better understand their impact on climate and air quality. Over the past years, the CLOUD experiments focused on answering these questions. For example, Kirkby *et al*.[20] has presented the state-of-the-art knowledge on NPF from laboratory experiments at the CERN CLOUD chamber and their relevance to the ambient atmosphere. Zhao *et al*.[21] implemented CLOUD experimental NPF rates into a global model to determine the vapors driving NPF in different regions of the atmosphere. Similar to Kirkby *et al*.,[20] this paper capitalizes on CLOUD results, but with a main focus on the chemical mechanisms involved in the production of condensable species and the molecular composition of nucleating clusters.

2. Methods

2.1 *Facilities*

2.1.1 *The CLOUD Chamber*

The CLOUD chamber is the world's first and, so far, unique laboratory facility to reach the demanding technical performance and ultra-low contaminant levels required to measure NPF under controlled atmospheric conditions. At the heart of the experiment is a continuous flow, 26.1 m^3 chamber, surrounded by state-of-the-art instrumentation (Fig. 1). In addition, the chamber is supplied continuously with synthetic air from the evaporation of liquid nitrogen and oxygen. An ultra-clean gas system supplies the chamber with ~25 trace gases at concentrations as low as 1 parts per trillion by volume (pptv) from concentrated sources. The oxidant concentration, and hence the production rates of oxidation products, can be varied by modulating the intensity of different lights covering different wavelengths. Moreover, CERN's Proton Synchrotron is used to simulate the influence of GCRs on NPF through the modification of atmospheric ion levels. All experimental conditions, including temperature, organic and inorganic precursor concentrations, chamber light settings and GCR levels are precisely controlled and measured in real time.[9] Around ten mass spectrometers, equipped with different ionization techniques, monitor the changes in the concentrations of thousands of precursor vapors and oxidation products with a time resolution down to 1 Hz. The evolution of the number and size distributions of the particles generated from these oxidation products is measured using several particle sizers covering the entire size range between few several hundred nanometers. From these measurements, nucleation and growth rates are derived and compared against the concentrations of oxidation products.[22] NPF rates are measured as the flux of particles that reach a size of 1.7 nm – a proxy that separates particles from molecular clusters. Results are compared to ambient observations and parameterized for integration in global models.[23]

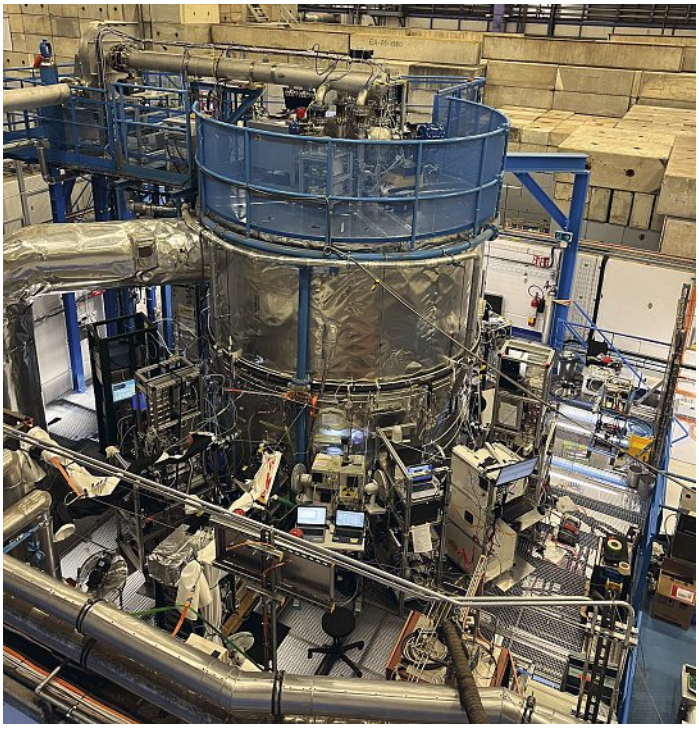

Fig. 1. Image showing the CLOUD chamber at CERN and the surrounding instruments which measure the chemical and physical properties of gases and particles along with meteorological conditions inside the chamber.

2.1.2 *Field Observations*

Field observations for measuring NPF monitor atmospheric conditions across diverse environments, such as remote forests, coastal areas, and cities, to measure when and how new particles form. These measurements use instruments to track particle size distributions, temperature, humidity, solar radiation, and precursor gas concentrations, helping identify the factors driving or in-

hibiting NPF events. Short-term campaigns provide snapshots of NPF mechanisms, while long-term measurements offer a more comprehensive understanding and track trends over long periods of time. In Switzerland, long-term monitoring stations at Payerne and Jungfraujoch, part of the GAW (Global Atmosphere Watch Programme) and ACTRIS (Aerosols, Clouds, and Trace gases Research Infrastructure Network) networks, are key sites for studying NPF precursors and events, with ongoing studies building on previous findings.[24]

2.2 *Instrumentation*

2.2.1 *Particle Number Size Distributions*

Thanks to significant advancements in particle number size distribution measurements, NPF events can now be tracked down to 1 nm sizes. Newer instruments, like the nano-cluster nuclei counter (nCNC) developed by Vanhanen *et al.*,[25] include a particle size magnifier (PSM) that grows particles up to 90 nm in size, allowing them to be optically measured using a condensation particle counter (CPC), placed in series. This setup enables the detection and counting of particles as small as 1 nm. Another method for measuring particle number size distribution is the neutral cluster and air ion spectrometer (NAIS). The NAIS can operate in either ion mode, measuring naturally charged ions from 0.8 to 40 nm, or in total mode, where neutral particles are charged, allowing for the measurement of aerosol particles between 2.5 and 42 nm.[26] To achieve a complete size distribution, an nCNC, NAIS, and traditional (nano) scanning or differential mobility particle sizer (SMPS/DMPS), which detects particles from a few up to hundreds of nanometers, are typically employed in parallel. Other developments include, but are not limited to, differential mobility analyzer train (DMA-train),[27] nano-scanning electrical mobility spectrometer (nSEMS),[28] cluster ion counter (CIC),[29] Caltech nano-radial DMA (nRDMA).[30]

Combining the measurements from different instruments especially in the sub-10 nm range remains tricky[31] and relies on the traditional comparison between the number size distributions within the overlapping size range, or an overall combined inversion as introduced by Stolzenburg *et al*.[32]

2.2.2 *Precursor Vapors*

Online mass spectrometry is key for studying particle formation mechanisms at a molecular level.[33] As different mass spectrometers have varying sensitivities towards different molecules, a suite of online mass spectrometers is needed to cover the full volatility range of vapor precursors.[34] Volatile organic compounds (VOCs), dimethylsulphide (DMS) and their moderately-oxygenated oxidation products can be detected with a proton-transfer-reaction time-of-flight mass spectrometer, PTR-ToF.[35] A chemical ionization atmospheric pressure interface time-of-flight mass spectrometer (APi-ToF), CI-APi-ToF[36] using NO_3^- as the reagent ion is often deployed to measure the concentrations – down to pptv levels – of ambient neutral clusters including sulfuric acid, iodic acid, and extremely low volatility clusters. The molecular composition of ambient positively/negatively charged clusters can be analyzed with an APi-ToF, which has been instrumental for detecting the nucleating ions involved.[37] Finally, multiple mass spectrometers have been proposed to measure the composition of growing particles. A notable example is the filter inlet for gases and aerosols (FIGAERO) coupled with a time-of-flight chemical ionization mass spectrometer (ToF-CIMS), which thermally desorbs semi-continuously sampled aerosols before detection.[33,38] Another example is the extractive electrospray ionization time-of-flight mass spectrometer (EESI-ToF-MS), which continuously measures aerosols down to 15 nm with time resolutions of a few seconds.[39] The Thermal Desorption Differential Mobility Analyzer (TD-DMA) coupled to a ToF-CIMS is also one additional example of an instrument which provides size-resolved chemical analysis of nucleation mode aerosol particles in the size range from ~10 to ~30 nm.[40]

2.2.3 *Cross Calibration*

It is worth mentioning that the CLOUD chamber also serves as an additional asset providing the possibility of cross calibration of several instruments. This is why several spin-off companies bring along their newly developed instrumentation where they are tested for their detection limits and calibrated against standard measuring instruments. The cross calibration is not limited to particle measuring instruments but also mass spectrometers for trace gas and precursor vapor measurements.

3. Particle Formation and Growth Mechanisms

3.1 *Sulfuric Acid – Base*

Sulfuric acid (H_2SO_4) due its low vapor pressure is expected to be critically involved in aerosol nucleation and growth. H_2SO_4 is primarily formed from the oxidation of sulfur dioxide (SO_2). Once emitted into the atmosphere, SO_2 undergoes oxidation through reactions with hydroxyl radicals (OH) or carbonyl oxides known as stabilized Criegee intermediates, leading to the formation of sulfur trioxide (SO_3), which rapidly reacts with water to produce sulfuric acid.[22] The CLOUD experiment revealed that binary sulfuric acid, water (H_2SO_4-H_2O) nucleation at ambient H_2SO_4 concentrations of 0.1 pptv is too slow to account for NPF in the warm boundary layer. However, the presence of trace amounts of ammonia (NH_3) at just 100 pptv can increase nucleation rates by over 100-fold. Additionally, GCRs can further boost these rates by 10-fold.[9] The nucleation process is fundamentally driven by the formation of acid–base pairs, with each NH_3 molecule pairing stoichiometrically with an H_2SO_4 molecule. This mechanism confirmed by quantum chemical calculations, suggests that while binary nucleation is insufficient in warmer conditions, the addition of NH_3 significantly enhances the formation of stable clusters, particularly in colder environments.[41]

Ternary nucleation involving H_2SO_4, NH_3, and H_2O has been observed in various environments, including the free troposphere at high-altitude sites like Jungfraujoch[24a] and Chacaltaya,[37b] as well as in cooler boundary layer locations such as the boreal forest[10a] and polar regions.[42]

NH_3 in the atmosphere originates from both natural and anthropogenic sources. Natural sources include emissions from soils, oceans, and biomass burning, while anthropogenic sources primarily involve agricultural activities, such as the application of fertilizers and livestock waste management.[43] SO_2, the precursor vapor for sulfuric acid, has both natural and anthropogenic origins as well. While anthropogenic SO_2 is largely emitted from fossil fuel combustion for energy production and shipping, accounting for approximately 75% of global sulfur emissions, the dominant natural source of SO_2 is the oxidation of DMS, a compound emitted by phytoplankton in the oceans. The oxidation of DMS not only produces sulfuric acid but also methanesulfonic acid (MSA), particularly in cooler conditions.[44] Like H_2SO_4, MSA may cluster with NH_3 to form particles, especially in marine environments, although its role in NPF remains unclear, calling for further CLOUD experiments and atmospheric observations.

Acid-base nucleation also drives NPF in the polluted boundary layer, although rates vary widely spanning over four orders of magnitude.[45] CLOUD has shown that this variability results from a complex interplay between temperature, acid and base concentrations, and the nature of the base. Amines, particularly dimetyl-amine (DMA), dominated by anthropogenic activities,[46] can form extremely stable clusters with H_2SO_4. The addition of just a few pptv DMA to an H_2SO_4-NH_3 mixture can enhance particle formation rates by several orders of magnitude compared to

ammonia alone. In the absence of DMA, nucleation rates become highly sensitive to NH_3 concentrations, increasing by 100-fold, when NH_3 levels rise from 1 part per billion by volume (ppbv) to 10 ppbv,[47] a concentration frequently found in polluted cities.[48] Temperature plays another key role in stabilizing the formed clusters. The formation rates for both H_2SO_4–NH_3 and H_2SO_4–DMA particles are 100 times faster at winter temperatures (5 °C) compared to summer (20 °C), with H_2SO_4–DMA nucleation rates reaching kinetic limits in winter at ambient relevant DMA concentrations. In urban environments, where high condensation sinks are common, haze episodes initiated by NPF require a rapid nucleation mechanism.[49] DMA, when present at a few pptv, is sufficient to trigger kinetically limited nucleation of H_2SO_4 particles at lower temperatures. This mechanism has been observed in urban environments such as Shanghai and Beijing.[45,50]

3.2 *Iodine Oxoacids*

Coastal seaweeds release iodide during low tide, which reacts with ozone to produce molecular iodine.[51] Observations at Mace Head, on the west coast of Ireland, have shown a strong correlation between iodine emissions and high NPF rates, later identified iodic acid (HIO_3) as a key nucleating agent. Iodine-driven nucleation has also been observed in other coastal regions, as well as over the high Arctic pack ice, with little contribution from sulfuric acid.[52]

CLOUD experiments have confirmed that both ion-induced and neutral nucleation of HIO_3 proceed efficiently under atmospheric conditions. The charged nucleation pathway involves the formation of iodine oxides (I_nO_m) from the additions of iodine oxoacids. Neutral nucleation invokes the stepwise condensation of HIO_3 followed by iodous acid (HIO_2), which act as a base stabilizing the neutral clusters, as confirmed by quantum chemical calculations.[12]

More recent experiments suggest that in marine and polar regions, where NH_3 levels are typically low, iodine oxoacids can significantly interact with H_2SO_4, enhancing the nucleation rates by up to 10,000 times. HIO_3 binds strongly with sulfuric acid in charged clusters, driving nucleation synergistically, while HIO_2 can substitute for ammonia, forming robust acid-base pairs in molecular clusters.[53] This enhancement of HIO_3 nucleation by iodine oxoacids could be particularly impactful in low-level marine stratocumulus clouds, which reflect a significant portion of solar radiation back into space, thus influencing the climate.

Furthermore, CLOUD precisely isolated the gas-phase formation mechanism of HIO_3.[54] It proceeds with the photolysis of molecular iodine by green light, producing iodide radicals ($I^\bullet$) that react with ozone (O_3) to form $IO^\bullet$. The self-reaction of $IO^\bullet$ produces iodooxy hypoiodite, IOIO, which then reacts with O_3 to form $IOIO_4$. The subsequent reaction of $IOIO_4$ with water generates HIO_3 and HOI. This newly identified mechanism is highly efficient at producing gas-phase HIO_3 in overcast daylight, explaining its observed levels in the remote lower troposphere. The mechanism has been confirmed through kinetic modeling, mass spectrometry measurements at CLOUD, and quantum chemical calculations.

The growth of charged or neutral iodine oxoacid particles to CCN sizes is driven almost entirely by HIO_3, which condenses at the kinetic limit. Particulate iodate can be reduced within particles, releasing iodine back into the gas phase, thus recycling iodine and sustaining the nucleation process. However, it remains unclear whether this reduction occurs on the aerosol surface or in the bulk, whether it requires light, or what the reducing agent might be. The rising iodine emissions, driven by increased sea surface ozone and retreating sea ice, suggest that iodine oxoacids will play an increasingly important role in NPF in marine and polar regions.

3.3 *Nitric Acid*

CLOUD has shown that nitric acid (HNO_3) plays a critical role in both the rapid growth of atmospheric particles in the boundary layer and their formation in the free troposphere.[16,23b] Gas phase HNO_3 is produced from the oxidation of NO_X emissions, from anthropogenic sources, like traffic, and natural sources, like lightning and wildfires.

At warm boundary layer temperatures, HNO_3 and NH_3 combine to form semi-volatile ammonium nitrate (NH_4NO_3) particles. However, CLOUD has revealed that in urban areas, localized super-saturated concentrations of NH_3 and HNO_3, caused by inhomogeneities in temperature or emissions, can trigger a sudden, ultra-rapid growth of few-nanometer-sized particles. This rapid growth, driven by NH_3 and HNO_3 concentrations at ppbv levels –compared to the sub-pptv levels of H_2SO_4 – is short-lived and difficult to detect as equilibrium quickly reestablishes. These bursts of growth may explain the unexpected NPF observed during smog episodes in Asian megacities, even in environments subject to high loads of pre-existing particles acting as condensation sink, that would normally inhibit NPF.[16,55]

CLOUD has also shown that at upper tropospheric low temperature conditions, HNO_3 forms particles synergistically with H_2SO_4 and NH_3 at rates far exceeding those from any two of these vapors alone.[23b,56] This mechanism is particularly important above regions with high ammonia emissions, particularly over the Asian monsoon region, explaining the observed high concentrations of NH_4NO_3. NH_3 released from convective cloud droplets upon freezing, mixes with H_2SO_4 and HNO_3, resulting in rapid NPF.

The rapid activation of nanometer-sized clusters by nitric acid strongly depends on the cluster composition, temperature, and relative humidity, highlighting the need for further characterization of the contribution of NH_4NO_3 formation to CCN in different environments. This is especially important in the context of atmospheric denitrification, with Netzero emission strategies.

3.4 *Oxygenated Organic Molecules*

Most of the VOCs originate from biogenic emissions, serving functions like attracting pollinators, and deterring pests. The major biogenic VOCs include isoprene (C_5H_8), monoterpenes ($C_{10}H_{16}$), and sesquiterpenes ($C_{15}H_{24}$). Anthropogenic sources, mainly from combustion or evaporative processes, contribute to aromatic and aliphatic VOCs.

In the atmosphere, VOCs are highly reactive, with lifetimes of just a few hours. They are rapidly oxidized by O_3, OH, or nitrate radicals (NO_3) to form peroxy radicals ($RO_2^\bullet$). $RO_2^\bullet$ radicals can undergo autoxidation, which involves sequential intramolecular hydrogen transfers and molecular oxygen additions, resulting in highly oxygenated radicals. The process ends by radical–radical reactions that generate OOMs, including monomers or covalently bound dimers (ROOR′) with ultra-low volatility. $RO_2^\bullet$ radicals terminate through reactions with $HO_2^\bullet$ and $RO_2^\bullet$ radicals in pristine environments, or with NO in polluted urban atmospheres.

CLOUD has demonstrated that OOMs can nucleate homogeneously in the absence of inorganic acids[11a] and are crucial in the early stages of particle growth into CCN.[15b,c] In the warm boundary layer, low-volatility dimers are key for pure organic nucleation, while at colder upper tropospheric conditions, even oxygenated monomers have low enough volatility to nucleate. Although lower temperatures slow autoxidation, leading to less oxygenated products, they substantially reduce the volatility of these products, increasing NPF rates by 2 orders of magnitude at –25 °C compared to +25°C.

The significance of pure biogenic nucleation lies in its ability to provide a copious source of new particles in pristine environments, such as the preindustrial climate or Earth's future climate with lower anthropogenic SO_2 emissions. Biogenic nucleation raises the baseline preindustrial aerosol levels, reducing estimates

of anthropogenic aerosol radiative forcing and, consequently, Earth's climate sensitivity. Although few truly pristine regions remain today, biogenic organic nucleation has been observed in the boreal forest and peatland of Finland[13,14] and at the Pyramid Station in the Himalayas (5,050 m).[57]

Unlike SO_2 or NO_X, whose oxidation produces a single compound, VOC precursors generate thousands of oxidation products. Out of these products, only a minor fraction is of sufficiently low volatility to contribute to particle growth (<5%),[15b,15c] with an even smaller fraction forming particles (<0.1%).[11b,c] This makes NPF and growth rates extremely sensitive to the precursor structure, $RO_2^\bullet$ formation and termination processes, and the gas phase composition.[58] Conditions used during simulation experiments are generally simplified, promoting autoxidation, low-volatility dimer formation and pure organic NPF.[59] While there is a need for additional experiments that focus on precisely reproducing the sinks of $RO_2^\bullet$ radicals in the atmosphere, examples from CLOUD on the sensitivity of NPF to the fate of $RO_2^\bullet$ radicals are presented below.

In both the boreal forest and the Amazon rainforest, high monoterpene concentrations (~1 ppbv) are common. However, while NPF is frequently observed in the boreal forest, it is rarely seen in the Amazon boundary layer. This difference is likely due to the suppression of NPF by isoprene, which reaches concentrations of 3–5 ppbv in the Amazon, compared to below 20 pptv in the boreal forest. Isoprene produces small C_5-$RO_2^\bullet$ radicals, leading to more volatile C_{15} dimers compared to C_{20} dimers from monoterpenes alone.[60] By contrast, sesquiterpenes increase the yield of less volatile $C_{20\text{-}30}$ covalently bound dimers, doubling the NPF rate with just a 2% molar addition to a 10:1 isoprene-to-monoterpene mixture.[11c]

Anthropogenic emissions of SO_2 or NO_X have significantly altered the atmospheric reactions of biogenic VOCs and the ability of their oxidation products to produce particles. NO_X effects are multifaceted and involve (1) altering the fate of biogenic $RO_2^\bullet$ radicals, and (2) increasing the atmospheric oxidant concentrations.[61] CLOUD experiments show that high NO concentrations terminate $RO_2^\bullet$ autoxidation and dimer formation, favoring the production of volatile organonitrate monomers and reducing nucleation rate.[62] NO_X is also a source of NO_3 radicals during night-time, which, even with just few pptv levels, produce highly reactive $RO_2^\bullet$ radicals that suppress NPF, as shown at CLOUD and in the field.[33] By contrast, NO_X drives radical recycling and oxidant production, enhancing $RO_2^\bullet$ formation rates and possibly NPF. While recent modelling analyses,[61c] along with *in situ*[61a] and airborne measurements[61b,63] consistently suggest that NO_X leads to a net enhancement in biogenic organic aerosol concentrations, its net effect on NPF remains unknown. Unlike NO_X, SO_2 does not directly interfere with VOC oxidation, but through the production of H_2SO_4 it enhances biogenic NPF.[10a] The overall effect of anthropogenic emissions on biogenic NPF should be studied in more detail, through experimental and modelling efforts, particularly for future projections in a hotter VOC-rich climate with lower anthropogenic emissions.

While OOMs from autoxidation are crucial for NPF and growth, over 95% of the first-generation oxidation products remain in the gas phase. These products can enhance NPF when transported to colder regions, be further oxidized to form less-volatile species dominating organic aerosol production,[64] or be scavenged by hydrometeors. Although largely absent in the Amazon rainforest itself, NPF occurs abundantly in convective cloud outflows. Intense NPF has also been observed in the tropical upper free troposphere over the Pacific and Atlantic oceans, spanning about 40% of Earth's surface.[65] Biogenic organic vapors may explain this broad scale nucleation, potentially serving as a major source of CCN for shallow liquid clouds over oceans, which dominate the cloud radiative effect on climate. Transport plays even a larger role for anthropogenic vapors, as products become increasingly more reactive with each oxidation step. Investigating the effects of transport, multi-oxidation, and wet scavenging on the fate of OOMs and their role in particle formation remains a critical area of research and is particularly important for understanding the future NPF mechanisms in a changing world subject to climate change and Netzero emissions.[66]

4. Conclusions and Outlooks

Advancing our understanding of NPF is crucial for mitigating urban air pollution, refining climate sensitivity estimates, and anticipating shifts in global radiative forcing as anthropogenic aerosols decrease due to air quality measures. The CERN CLOUD experiments have played a key role in uncovering the molecular processes behind NPF, enabling more realistic simulations across various atmospheric environments. These findings are beginning to improve the representation of aerosol nucleation in climate models, offering a clearer picture of future climate impacts. CLOUD ongoing research will focus on capturing the complex interactions between anthropogenic-biogenic organic-inorganic vapors, radicals, and stabilizers such as ammonia and cosmic-ray ions. It will also examine the effects of transport, multi-generation oxidation, and wet scavenging on the fate of precursor vapors and their role in NPF, particularly in the free troposphere. Additionally, CLOUD will leverage its extensive experimental data to derive the thermodynamic principles driving nucleation and develop simplified parameterizations for integration into climate models. As aerosol particles continue to pose significant but poorly understood health risks, particularly in densely populated areas, CLOUD future work will be crucial in informing both climate projections and public health strategies.

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