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Atmospheric Radioactive Aerosols and Climate Change Charging Mechanisms, Transport Dynamics, and Modelling Advances for Sustainable Assessment

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ABSTRACT

Accurate prediction of the transport and deposition of atmospheric radioactive aerosols necessitates the integration of multiple tightly coupled multiphysical processes. This study systematically evaluates the governing principles underlying transport behaviour, deposition fluxes, charge development, radioactive particles with particular emphasis on hot, arid environments and dust storm conditions. In addition, it presents a critical synthesis of recent progress in the modelling radioactive aerosol transport in the open atmosphere, highlighting the coupled interaction between electrical charging and transport phenomena. This work is intended both as a structured introduction for newcomers and as an updated critical resource for experts aiming to identify decent technological progress, key knowledge gaps, and future research priorities in the field.

Keywords: Radioactive aerosols, Transport dynamics, Charge evolution, Deposition mechanisms, Dispersion Modelling, Atmospheric simulation

1. Introduction

Radioactive aerosols consist of solid particles and/or liquid droplets suspended within a gaseous medium, typically air, that contain radionuclides of either anthropogenic (human-related) or natural origin. An individual entity

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within this system is referred to as a radioactive aerosol particle. These particles exhibit a wide size spectrum, ranging from few nanometres to several hundred micrometres, and their atmospheric residence time vary from less than a day in the troposphere to over a year in the stratosphere (von Schoenberg et al., 2021). Anthropogenic radionuclides are released into the atmosphere from source such as research facilities, medical institutions, industrial operations, and nuclear fuel cycle activities. Moreover, large-scale emissions may occur during nuclear weapons testing or nuclear power plant accidents, predominantly in particular form. Radioactive aerosols may form indirectly through gas-to-particle conversion processes (secondary aerosols), in addition to direct emissions (primary aerosols). Once emitted, atmospheric circulation-particularly wind-can transport these aerosols over considerable distances. The removal of airborne radionuclides occurs exclusively through wet deposition, dry deposition, or radioactive decay (Rehfeld and Heimann, 1995). The resulting fallout can contaminate soil, vegetation, water systems, and built environments through direct contact, ingestion, or inhalation, thereby posing long-term environmental and health risks (Poinssot and Geckeis, 2012).

Radionuclides released during the Fukushima nuclear disaster led to radioactive contamination hundreds of kilometres from the source (Yamauchi, 2012). A comparable scenario followed the Chernobyl accident, where radioactive particles (with activities exceeding 100 kBq, sufficient to pose immediate health hazards) were transported over long distances and deposited across several European countries (Pollanen et al., 1997a). The dispersion and deposition behaviour of radioactive aerosols is strongly influenced by particle size distribution, physicochemical properties, and prevailing meteorological conditions. Among particle characteristics, electrical charge plays a potentially significant yet often neglected role in influencing aerosol transport (Jang et al., 2022). Additional charging mechanism may arise under strong electric fields (field charging) or through frequent contaminant plumes. These processes can alter the surface charge distribution of particles in ambient air (Kim et al., 2015). The coupling of charge evolution with transport, coagulation, and deposition dynamics is therefore critical, as electrostatic interactions between particles and surface can substantially modify coagulation rates and deposition kinetics. Consequently, there is a need for predictive tools capable of accurately simulating the spatial and temporal evolution of radioactive aerosol, incorporating variations in charge state, aggregation behaviour, deposition pathways, and radioactivity (Abdelmonem, et al., 2015, Adams and Seinfeild, 2002). Over several decades, extensive academic and industrial efforts have been devoted to developing such models to support decision-making in both routine and accidental release scenarios (von Schoenberg et al., 2021; Thiessen et al., 1999). It is widely acknowledged that these models differ significantly in scope and complexity. Some and designed to simulate the global-scale, long-term impacts of radionuclide dispersion (Bytwerk et al., 2011), whereas others focus on detailed processes within localized systems, such as turbulent transport and deposition in confined environments (Gu et al., 2019). The objective of this study is to provide a comprehensive examination of the principles governing charge and transport dynamics of atmospheric radioactive aerosols, along with selected recent advancements in the field (Adisa, O et al., 2020; Agbo, F et al., 2021). Special emphasis is placed on the interconnected effects of particle transport, electrical charge, radioactivity, deposition, and coagulation, particularly under dust storm conditions in hot, arid, and semi-arid regions. Such environments-characteristic of norther Africa, western Asia, and inland Australia-are marked by elevated temperatures,

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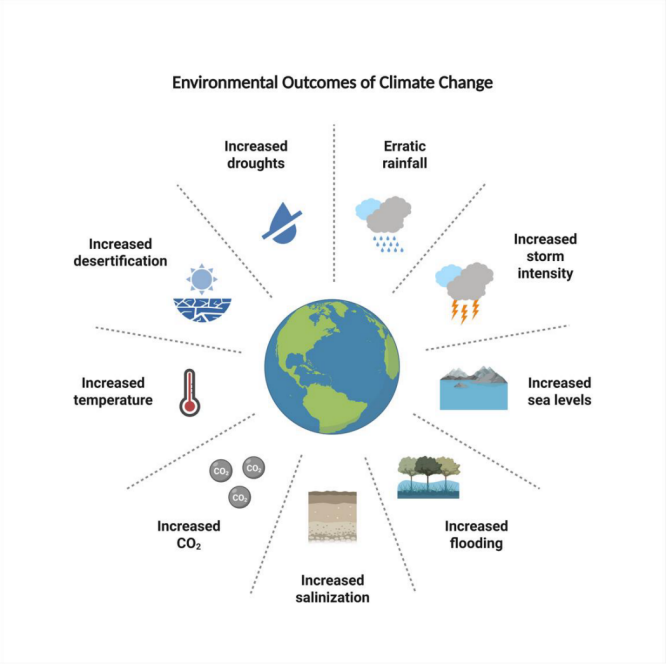


Figure 1. Environmental outcomes associated with climate change. Schematic illustration of major environmental consequences of climate change, including increased atmospheric CO_2 concentrations and temperatures, drought and desertification, erratic rainfall and intensified storms, sea-level rise, flooding, and salinization.

minimal precipitation, low cloud cover, and dry atmospheric conditions.

Over the past two decades, numerous studies have investigation the emission, dispersion, and deposition behaviour of radioactive aerosol. Increasing anthropogenic activity has contribute to a global rise in Aerosol Optical Depth (AOD) (Ajoku et al., 2020; Albayrak et al., 2013; Forkel et al., 2012). Observations from instruments such as MODIS and OMI indicate a consistent increase in AOD over a 17-year period during pre-monsoon moths (March-May) across major regions of India, driven by enhanced aerosol loading and temperature anomalies (Forster, D et al., 2020; Gayatri K., 2017). This increase in aerosol concentration is closely linked to population growth and associated industrial expansion. India's population rose by approximately 27% between 2002 and 2021, increasing from 1.02 billion to 1.39 billion (The Worl Bank WBD, 2022). This growth has intensified demands for industrial output, food supply, energy generation, and consumer goods, thereby exacerbating environmental pressure. As a result, air pollution has increased, posing risk to human health and contributing to climate change (Geertsema and Schreur, 2004; Givati and Rosenfeld, 2004).

2. Aerosol Spatiotemporal Distribution and Component Retrievals from Remote Sensing Data

Satellite-based remote sensing has fundamentally advanced the understanding of aerosol spatiotemporal variability by providing high-resolution, large-scale, and long-term observational dataset. Key aerosol parameters-including AOD, single scattering albedo (SSA), Angstrom exponent, and vertical distribution profiles-are retrieved from instruments such as MODIS, OMI, CALIPSO, and VIIRS. These datasets are essential for analysing aerosol behaviour in regions such as the Taklimakan Desert, Sichuan Basin, Yangtze River Delta, and eastern China. The integration of multi-source

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satellite observations with advanced retrieval algorithms, including Deep Blue and Dark Target, enables accurate tracking of aerosol trends over extended periods (2003-2020). Furthermore, the GRASP/Component algorithm, which assimilates radiance measurements from AERONET, POLDER/PARASOL, and GF-5 platforms, facilitates the retrieval of aerosol constituents such as mineral dust, brown carbon, and black carbon. This enhances the quantification of radioactive forcing differences between terrestrial and marine environments and improves understanding of aerosol composition during regional haze events. Vertical profiles derived from CALIPSO provide insights into aerosol transport pathways, as demonstrated by three-dimensional analyses of dust storm dynamics in East Asia. Additionally, China's satellite missions, including Fengyun-4 and Gaofen-1/5, complement global datasets and progressively improve the spatiotemporal characteristics of aerosol properties (Che et al., 2024).

2.1. Long-Term Aerosol Optical Properties Based on Modelling, Assimilation, and Reanalysis

Long-term assessment of aerosol optical characteristics relies heavily on numerical modelling, data assimilation, and reanalysis systems such as MERRA-2, CAMS, and JRAero. These frame-works integrate satellite and ground-based observations to generate consistent, high-resolution, and spatially continuous datasets. Reanalysis products are particularly valuable for overcoming observational limitations, especially over bright surfaces or cloud-covered regions.

Model performance varies regionally; for instance, CAMS demonstrates high accuracy in eastern China, whereas MERRA-2 performs better in northern and western regions. These datasets support the identification of key drivers influencing AOD variability, including SO2 emission, black carbon concentrations, wind patterns, and humidity. Despite their utility, uncertainties persist due to limitations in emission inventories, model parameterization and incomplete representation of chemical processes (e.g., absence of nitrate components). Nevertheless, reanalysis datasets have been instrumental in identifying decadal trends in aerosol loading and evaluating aerosol-climate interactions (Figure 1). Statistical models linking AOD to meteorological and emission variables further illustrate the combined influence of atmospheric conditions and policy intervention on air quality trends in China. Overall, these tools form a critical foundation for climate modelling, pollution forecasting, and environmental policy development (Che et al., 2024).

2.2. Effective Radiative Forcing (ERFaci)

Effective radiative forcing from aerosol-cloud interactions (ERFaci) quantifies the alteration in Earth's energy balance resulting from aerosol-induced modifications to cloud microphysics and dynamics, aerosols function as cloud condensation nuclei (CCN) or ice nuclei (IN), leading to the formation of a greater number of smaller cloud droplets. This increases cloud reflectivity (albedo) and typically suppresses precipitation formation. These processes correspond to the Twomey effect (enhanced reflectivity) and the Albrecht effect (extended cloud lifetime).

However, ERFaci strong variability depending on aerosol concentration, composition (absorbing versus scattering), atmospheric moisture, and cloud development stage. In humid and convectively active environments, aerosol can enhance cloud growth and precipitate through increased latent heat release. Conversely, under dry or highly polluted conditions, aerosols may inhibit precipitation by promoting evaporation and competition for cloud water.

ERFaci also influences precipitation characteristics, including intensity, duration, and

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timing, and can affect large-scale atmospheric systems such as tropical cyclones and storm tracks. Its effects are often temporally delayed due to the time required for aerosol incorporation into cloud systems and subsequent microphysical adjustments. Consequently, ERFaci represents a complex yet essential component of the climate system and remains a major source of uncertainty in climate projections (Zhao et al., 2024).

3. The Influence of Meteorological Conditions on ACI and ARI

Meteorological factors play a critical role in determining the magnitude and nature of Aerosol-Cloud Interactions (ACI) and Aerosol Radiation Interactions (ARI), thereby influencing cloud formation and precipitation processes. Key parameters-including humidity, low-level tropospheric stability (LTS), convective available potential energy (CAPE), wind shear, and synoptic dynamics-interact intricately with aerosol processes (Zhang et al., 2024). In atmospheres characterised by low stability (Low LTS) and high CAOE, convective activity is enhanced, allowing aerosols to intensify cloud development and precipitation. In humid conditions, particularly during warmer periods, aerosols can facilitate droplet growth and rainfall through latent heat release. In contrast, under dry or cold conditions, aerosols may suppress precipitation by enhancing evaporation or stabilizing the atmosphere. Wind shear also significantly influences aerosol-cloud interactions; string shear can disrupt vertical transport and weaken interactions, whereas weak shear conditions favour stronger aerosol effects. Synoptic conditions further modulate these processes: under weak large-scale forcing, aerosol impacts are more evident, while strong weather systems such as cyclones or frontal systems may dominate and mask aerosol influences. Additionally, aerosols can modify meteorological conditions by altering stability, redistributing moisture, and affecting energy balance, thereby creating feedback mechanisms that further complicate their role. Overall, the impacts of ACI and ARI are highly sensitive to prevailing atmospheric conditions, underscoring the importance of integrating meteorological variability into climate assessments (Zhao et al., 2024).

3.1. Environmental Chambers

Environmental chambers have become indispensable tools in atmospheric chemistry for investigating the formation of secondary organic aerosol (SOAs), particularly from emission sources such as agricultural residue burning (ARB). These systems replicate atmospheric conditions under controlled environments, allotting for the isolation of variables such as humidity, radiation, and oxidant concentrations. By introducing volatile organic compounds (VOCs) into these chambers and exposing them to oxidant such as hydroxyl radicals (OH), ozone (O3), and nitrate radicals (NO3), researchers can examine the transformation of gaseous precursors into particulate matter. Environmental chambers are broadly categorized into static systems (smog chambers) and dynamic systems (oxidation flow reactors), each offering distinct advantages in terms of temporal resolution and simulation scale. They are widely applied in emission characterization, source attribution, reaction pathway analysis, and the study of photochemical ageing processes of ARB emissions (Joshi et al., 2024).

3.2. Smog chambers

Smog chambers are particularly effective for examining the gradual transformation of chemical precursors and associated changes in aerosol properties due to their long residence times and controlled experimental conditions. These large, static batch reactors-typically ranging from 5 to 300 m3- are constructed from materials such as quartz, stainless steel, or FEPTeflon to minimize wall interactions while maintaining optical transparency. Within these chambers, VOCs and oxidants are introduced

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and exposed to radiation (either artificial UV sources or natural sunlight) to initiate photochemical reaction. These processes, which may extend from several hours to multiple days, replicate atmospheric ageing mechanisms experienced by VOCs. Smog chambers have been extensively used to study emission from agricultural residues such as rice, wheat, and maize straw, revealing complex chemical transformations and variability in SOA formation depending on fuel type and combustion conditions (Joshi et al., 2024).

3.3. Oxidation flow Reactors

Oxidation flow reactors (OFRs) are compacts, dynamic systems designed to stimulate atmospheric ageing over significantly shorter timescales compared to smog chambers. Among these, the Potential Aerosol Mass (PAM) reactors have enabled rapid evaluation of SOA formulation under elevated oxidative conditions. Typically, cylindrical with volumes of approximately 10-13 litres, OFRs generate hydroxyl radicals via UV-induced processes, either through ozone photolysis (OFR254 mode) or through photolysis of water vapour and oxygen (OFR185 mode). Their continuous-flow operation makes them suitable for field deployment and real-time measurements. OFRs can stimulate days of atmospheric ageing within minutes, enabling high-throughput experimentation. However, the elevated oxidant concentrations and short residence time may limit their ability to capture slower or more complex atmospheric reactions. Despite these limitations, OFRs remain valuable tools for assessing pollutant oxidation potential and investigation rapid SOA formation from ARB emissions (Joshi et a., 2024)

4. Analytical Instrumentations

The characterization of SOA formation and evolution within environmental chambers require advanced analytical instrumentation capable of resolving complex gas-particle mixtures. Gas-phase analysis commonly employs instruments such as Proton-Transfer-Reaction Time-of-Flight Mass Spectrometry (PTR-ToF-MS) and gas Chromatography-Mass Spectrometry (GC-MS). PTR-ToF-MS enables high-sensitivity, real-time monitoring of VOCs and their oxidation products, whereas GC-MS provides detailed offline chemical specification from collected samples. Particle-phase characterization is achieved using instruments such as the High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS) and the Aerosol Chemical Speciation Monitoring (ACSM), which measure aerosol mass, composition, and size distributions. Particle number concentration and size are determined using the Condensation Particle Counter (CPC) and Scanning Mobility Particle Sizer (SMPS). Advanced dual-phase systems, including Chemical Ionization Mass Spectrometry (CIMS) with FIGAERO inlets and Extractive Electrospray Ionization Time-of-Flight Mass Spectrometry (EESI-ToF-MS), provide molecular-level insights into both gas and particle phase without fragmentation. Collectively, these instruments enable comprehensive assessment of SOA formation composition, and transformation under controlled experimental conditions (Joshi et al., 2024).

4.1. Forest Fire and Fuel Characteristics

Forest fires are complex, uncontrolled combustion phenomena occurring in natural vegetative environments, involving processes such as flaming and smouldering combustion, moisture evaporation, inert heating, and pyrolysis. The classification and properties of forest fuels-including live and dead vegetation such as lichens, mosses, shrubs, herbs, litter peat, woody debris, and canopy foliage- are fundamental to understanding fire behavior. These fuels primarily consist of cellulose, hemicellulose, and lignin, but exhibit significant variability in composition, structure, moisture content, and flammability. Their thermophysical

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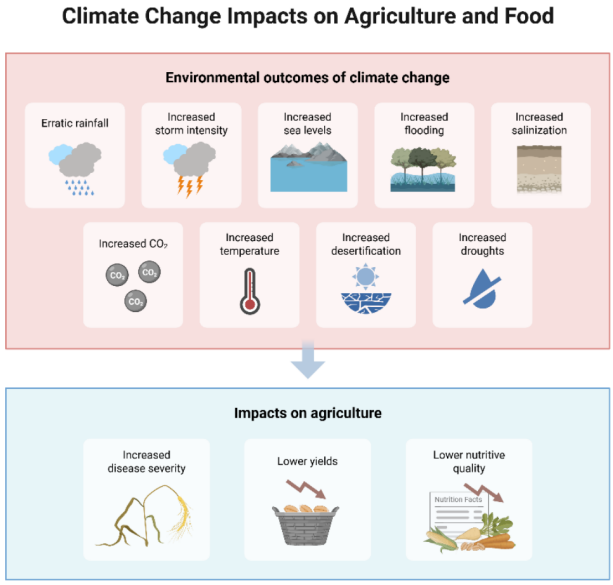


Figure 2. Impacts of climate change on agriculture and food systems. Schematic representation of major climate-related environmental changes, including erratic rainfall, increased storm intensity, sea-level rise, flooding, salinization, atmospheric CO_2 concentrations, temperature, desertification, and drought. These environmental stressors can adversely affect agricultural systems by increasing crop disease severity, reducing crop yields, and lowering the nutritional quality of food.

properties and spatial distribution strongly influence fire dynamics and the emission of pollutants such as soot and aerosols. Globally, several fuel classification systems exist, including the Fuel Characteristic Classification System (FCCS) in the United States, the Canadian Forest Fire Danager Rating System (CFFDRS), and Arc Fuel in Europe. Accurately representing these heterogeneous fuel systems remain a major challenge in fire modelling and simulation (Baranovskiy et al., 2024).

4.2. Aerosol Emission

Aerosol emissions from forest fires represent a significant environmental concern due to their impact on climate, air quality, and human health. These emissions comprise a complex mixture of pollutants, including organic carbon, black carbon, Sulphur oxides, nitrogen oxides, carbon monoxide (CO), carbon dioxide (CO2), and fine particulate matter (PM2.5). Such aerosols influence Earth's radiative balance, contributing to both short-term atmospheric effects-such as reduced visibility, fog formation, and cloud suppression-and long-term climate change. Soot particles are particularly impactful, as they absorb solar radiation, reduce surface albedo, and accelerate ice and glacier melting up on deposition. Emission characteristics depend on fuel type, combustion conditions, and fire intensity (Figure 2). For instance, peat fires produce prolonged emissions with high toxicity potential. These pollutants also pose serious health risks, contributing to increased mortality and exacerbating respiratory and cardiovascular disease during smoking events. Forest fires account for up to 34% of global atmospheric soot, with regional contributions exceeding 60% in areas such as Southeast Asia and Siberia (Baranovskiy et al., 2024).

4.3. Forest Fuel Pyrolysis

Pyrolysis is a key thermochemical process in forest fires, occurring under oxygen-limited conditions and resulting in the production of char, tar, and volatile gases that sustain combustion and generate soot. Different biomass components undergo pyrolysis at distinct temperature ranges: cellulose (300-390°C), hemicellulose (290-350°C), and lignin (450-

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800°C), producing diverse chemical products. This process plays a direct role in the initial formation of aerosol nanoparticles. Pyrolysis is typically represented using mechanistic, network-based, or kinetic modelling approaches, with kinetic models-such as single-step Arrhenius formulations or distributed activation energy models-favoured for their simplicity and empirical capability. However, variability in biomass composition, moisture content, and environmental conditions introduces significant modelling challenges. Pyrolysis remains a critical stage in the aerosol lifecycle, as emissions-particularly black carbon-are strongly influenced by dispersion characteristics and environmental exposure (Baranovskiy et al., 2024).

4.4. Forest fire Modelling and simulation

Forest fire modelling is essential for predicting fire behaviour, assessing environmental impacts, and estimating pollutant emissions. Existing modelling approaches include empirical, graph-based, machine learning, physical, and quasi-physical frameworks. Physical models, such as FIRETEC and WRF-SFIRE, simulate detailed combustion processes-including pyrolysis and emissions-but their high computational and data requirements limit their application to local scales. Quasi-physical models simplify certain processes while retaining essential physical realism. Empirical models, such as the Rothermel model, rely on statistical relationships derived from experimental data and are suitable for large-scale applications, although they lack detailed physical representation. Cellular automata and machine learning approaches-including LSTM and SVM-based models-simulate fire spread using rule-based or data-driven methods and have shown promise for real-time prediction. Advanced integrated systems, such as Meso-NH, couple atmospheric and fire dynamics, while modern simulation frameworks increasingly incorporate remote sensing data. These models support fire spread prediction, emission estimation, and mitigation planning. While physical models provide detailed emission estimates, empirical and hybrid approaches are generally more practical for operational and regional-scale fire management (Baranovskiy et al., 2024).

5. GC- and LC-MS for molecular analysis

Gas chromatography-mass spectrometry (GC-MS) and liquid chromatography-mass spectrometry (LC-MS) are two widely employed analytical techniques for the molecular-level characterization of atmospheric organic aerosols (Oas). Since the 1970s, GC-MS has been extensively applied to identify volatile and semi-volatile organic compounds in aerosols, particularly in source apportionment studies. This technique relies on volatility-driven separation and is especially effective for thermal stable compounds with low to moderate polarity. It has successfully identified source-specific tracers such as polycyclic aromatic hydrocarbons (PHAs) originating from fossil fuel combustion and levoglucosan derived from biomass burning. However, GC-MS exhibits limitations in analysing highly polar or thermally unstable compounds, which often require chemical derivatization for detection. Advances such as comprehensive two-dimensional gas-chromatography (GCxGC) have significantly enhanced its resolving capability, enabling the identification of hundreds of compounds in a single analysis.

In contrast, LC-MS is better suited for analysing polar, non-volatile, and high molecular weight species, including dimers and oligomers of secondary organic aerosols (SOAs). This method employs soft ionization techniques-such as electrospray ionization (ESI), atmospheric pressure chemical ionization (APCI), and photoionization (APPI)-which minimize fragmentation and facilitate more detailed molecular identification (IPCC, 2021; IREA, 2023; Kumler et al., 2019; Craig et al., 2019).

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When coupled with high-resolution mass spectrometry (e.g., Orbitrap or FT-ICR-MS), LC-MS enables near-complete molecular characterization through precise molecular formula assignment and isomer differentiation. Although GC-MS remains advantageous due to its robustness and separation efficiency, LC-MS has become indispensable for investigating complex aerosol chemistry, particularly where soft ionization and detection of high molecular weight compounds are required (Hildmann et al., 2024).

5.1. Analytical approaches and resulting analytical conditions

Three principal analytical strategies are employed in the study of organic aerosols (OAs): targeted analysis (TA), non-targeted analysis (NTA), and semi-targeted analysis (STA), each characterized by distinct capabilities and requirements. Targeted analysis is applied when specific, well-characterized compounds-often serving as markers for particular sources or processes-are of interest. This approach provides high sensitivity, accuracy, and precision, and depends on the availability of authentic reference standards. Techniques such as tandem mass spectrometry (e.g., triple quadrupole MS) and reversed-phase liquid chromatography (RPLC) are commonly utilized in TA. In contrast, non-targeted analysis aims to detect and characterize all measurable organic compounds, including unknown species. This approach facilitates the investigation of complex and poorly understood atmospheric processes, such as cloud formation, aerosol ageing, and the discovery of novel chemical markers. Semi-targeted analysis serves as an intermediate approach, enabling the quantification of compound classes for which only limited representative standards exist. This method is particularly valuable when accurate standard generation is difficult, as is often the case for complex SOA constituents or organosulfates. Although STA may not yield absolute concentration values, it provides insight into relative trends and guides further investigation. The selection of an appropriate analytical strategy must align with the research objectives, and in many cases, a combination of approaches is employed to achieve a comprehensive understanding of OA composition and transformation (Hildmann et al., 2024).

5.2. Instrumental approaches

Advancements in instrumentation have played a pivotal role in improving the chemical characterization of organic aerosols. Traditional one-dimensional liquid chromatography (LC) and mass spectrometry (MS) systems have evolved into more advanced configurations, including multidimensional LC and tandem MS techniques. Liquid chromatography coupled with ultraviolet/visible detection (LC-UV/Vis) has long been used to detect compounds with strong chromaphoric properties, such as PAHs and derivatized carbonyl compounds. While UV/Vis detection lacks specificity, its integration with MS (LC-UV/Vis-MS) enhances selectivity and compound identification, particularly for optically active species like brown carbon. More advanced approaches, such as LC-MSn (tandem mass spectrometry), provide structural information by inducing fragmentation of selected ions. Techniques including product ion scanning and multiple reaction monitoring (MRM) are widely applied in studies of SOAs and biomass burning aerosols, offering high specificity for target compound identification and quantification. Two-dimensional liquid chromatography (2D-LC) techniques have emerged as powerful tools for resolving complex aerosl mixtures. In heart-cut (LC-LC) and multiple heart-cut (mLC-LC) configurations, selected fractions from the first chromatographic dimension undergo further separation in a second dimension, thereby enhancing resolution and reducing co-elution. Comprehensive LC (LCxLC) extends this approach by transferring the entire effluent from the first dimension into

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the second, providing maximum analytical coverage and making it particularly for non-targeted analyses. However, these techniques face practical challenges, including reduced sensitivity at trace concentrations, complex system configurations, and issues related to solvent compatibility (Hildmann et al., 2024).

6. Overview of Aerosol Acidity

Aerosol acidity, typically expressed in terms of pH, describes the acid-base characteristics of airborne particulate matter, particularly fine particles such as PM2-5. It plays a critical role in determining particle toxicity, atmospheric chemical processes, and ecological impacts. The acidity of aerosols primarily arises from the oxidation of precursor gases such as sulphur dioxide (SO2) and nitrogen oxides (NOx), leading to the formation of sulphuric acid (H2SO4) (Carreno et al., 2020; Tebaldi et al., 2013; Knuston et al., 2021; Onha et al., 2023). These acidic species interact with alkaline constituents, including ammonia (NH3) and non-volatile cations (NVCs) such as calcium and magnesium. The resulting compounds, notably ammonium sulphate and ammonium nitrate, constitute a major fraction of fine particulate matter, particularly during haze events. Despite partial neutralization by these species, PM2-5 in emission-intensive regions remain predominantly acidic (Doss-Gollin et al., 2023; Konapala et al., 2020; Olesen et al., 2011; Kanoglu et al., 2020). This persistent acidity influences cloud formation, radiative forcing, the gas-particle partitioning, while also enhancing the solubility of toxic metals. Although regulatory measures have significantly reduced emissions of acidic precursors, PM2-5 continues to exhibit substantial acidity in many regions, underscoring its ongoing relevance to environmental and health concerns (Song et al., 2024).

6.1. The Estimation and Driving Factors of Aerosol Acidity

Aerosol acidity is commonly estimated using thermodynamic equilibrium models such as ISORROPIA II and E-AIM, which simulate aerosol pH based on inputs including temperature, relative humidity, and chemical composition (Karnauskas et al., 2018; Gernaat et al., 2021; Soden et al., 2002; Canty et al., 2013). Direct measurement of aerosol pH remains challenging due to the small size and complexity of atmospheric particles; however, emerging techniques such as Raman spectroscopy, aerosol optical tweezers, and fluorescence-based probes are improving measurement accuracy. Key factors influencing aerosol acidity include concentration of acidic precursor (SO2 NOx), ammonia (NH3) levels, aerosol liquid water content (ALWC), and meteorological conditions such as temperature and relative humidity (MacMartin et al., 2018; Parker et al., 2018; Irvine et al., 2016; Baur et al., 2024). Elevated humidity increases ALWC, thereby diluting acidity. Whereas higher temperatures promote volatilization of neutralizing species, potentially enhancing acidity. Organic components generally have a limited direct effect on ALWC but can influence pH through interactions with inorganic constituents. Temporal variations also play a role, with seasonal and diurnal fluctuations affecting aerosol acidity. Lower pH values are typically observed during warmer seasons and daytime conditions due to enhanced secondary aerosol formation and reduced relative humidity (Dagin and Schrag, 2019; Cheng et al., 2019; UNEP, 2003). Spatial variability further reflects differences in emissions, atmospheric conditions, and the buffering capacity of regional air masses (Song et al., 2024).

6.2. Synergistic Effects Between Acidity and Gaseous Oxidants

Aerosol acidity interacts synergistically with gaseous oxidants such as nitrogen dioxide (NO2) and ozone (O3), significantly enhancing particulate toxicity. Acidic particles can reduce

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local pH within the respiratory tract, prolonging the activity of reactive oxygen species generated during oxidant exposure and thereby intensifying oxidative stress in lung tissues. Experimental evidence indicated that simultaneous exposure to acidic aerosols and O3 results in greater reductions in lung function and increased collagen deposition compared to exposure to O3 alone (Gokmen et al., 2016; Al-Rasheedi et al., 2020; Conceicao et al., 2018). This is attributed to the formation of reactive intermediates such as ozonide's and peroxides under acidic conditions. Furthermore, acidic environments diminish the effectiveness of endogenous antioxidants, such as uric acid, thereby amplifying inflammatory responses. When inhaled alongside particulate matter, NO2 can generate reactive nitrogen species (RNS), which may enhance allergic sensitivity and disrupt cellular signaling pathways (Zamuda et al., 2023; Wild et al., 2015; Sawadogo et al., 2020, IPCC, 2021). These combined effects contribute to increased respiratory and cardiovascular morbidity, particularly during periods of elevated oxidant concentrations, such as summer months. The findings highlight the importance of considering combined exposure effects in public health assessments and air quality management strategies (Song et al., 2024).

6.3. Possible Impacts of Emission Control on Aerosol Acidity

Despite substantial reductions in SO2 and NOx emissions achieved through global air quality regulations, aerosol acidity has remained relatively stable in many regions, particularly those with significant agricultural NH3 emissions. This persistence is largely due to the buffering effect of ammonia, which maintains aerosol pH within an acidic range (typically pH 1-3). In the eastern United States, PM2-5 acidity has remained low density declining sulphate levels, due to sustained NH3 concentrations and limited neutralization by non-volatile cations. A similar trend has been observed in China, where reductions in SO2 and NOx emissions during the 2010s did not result in substantial increase in aerosol pH. Consequently, while emission control strategies have successfully reduced particulate concentrations and improved air quality, they have not significantly altered aerosol acidity. Moreover, without substantial reductions in NH3 emissions-which are technically challenging to achieve-the health risks associated with persistent aerosol acidity may offset the benefits of reduced PM2-5 levels. Modelling studies suggest that only drastic reductions in NH3 emissions (≥90%) would lead to a meaningful increase in aerosol pH. Therefore, future air quality management strategies must explicitly address aerosol acidity alongside pollutant concentration reductions (Song et al., 2024).

6.4 Methods to collect and analyses individual fog/cloud residual particles

To investigate individual fog or cloud residual particles, researchers commonly employ Counter-Flow Virtual Impactors (CVI) or ground-based CVI (GCVI) systems to separate activated cloud droplets from non-activated interstitial particles. These instruments selectively admit droplets from non-activated interstitial particles. These instruments selectively admit droplets exceeding a defined size threshold (typically 6-10 μm), which are subsequently evaporated at elevated temperatures (~40°C), leaving residual particles for analysis. These residues provide critical insight into particle transformation processes occurring during cloud interactions. For real-time (online)analysis, techniques such as Single Particle Mass Spectrometry (SPMS), including ATOFMS and SOAMS, are used to determine particle size and chemical composition at the individual particle level. The Single Particle Soot Potometer (SP2) is specifically employed to analyses black carbon and its associated coating.

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Offline analytical methods include Scanning Electron Microscopy (SEM) and Transmission Electron Microscopy (TEM), often combined with Energy-Dispersive X-ray Spectroscopy (EDS), to characterize particle morphology and elemental composition. Additional advanced techniques-such as micro-PIXE, STXM/NEXAFS, NanoSIMS, and ToF-SIMS-provide detailed chemical and spatial information on particle structure and surface chemistry. These methods collectively enable comprehensive analysis of particle size, morphology, internal mixing states, and the formation of secondary species resulting from cloud processing. However, potential artefacts-such as the loss of volatile components and complex transformations during droplet activation and evaporation-must be carefully considered when interpreting results (Zhang et al., 2025).

6.5. Compositions of individual fog/cloud droplets residual particles in different environments

The chemical composition of individual fog or cloud droplet residual particles varies significantly across different environments due to their differences in emission sources and atmospheric processing mechanisms (Sun et al., 2018). In urban and industrial regions, these particles are typically dominated by soot; ammonium sulphate, fly ash, and metal-rich constituents (e.g., Pb, Fe, Zn), often exhibiting complex internal mixing with secondary species such as nitrate and sulphate, indicative of strong internal mixing with secondary species such as nitrate and sulphate, indicative of strong anthropogenic influence (Sunder et al., 1996; SuYing et al., 2010). In costal environments, residual particles are primarily composed of sea salt and aged sea salt, frequently mixed with sulphates, organic matter, and nitrogen-containing organic compounds formed through interactions between marine emissions and anthropogenic pollutants (Takemura et al., 2004). Dust-dominated regions are characterized by mineral particles such as quartz, aluminosilicates, calcite, and gypsum. Their effectiveness as cloud condensation nuclei (CCN) is enhanced when coated with soluble substances, including sulphates and organic compounds formed through atmospheric reactions (Tao et al., 2007). In remote background regions, particles are typically highly aged and internally mixed with sulphate, nitrate, and organic components due to long-range transport and extensive atmospheric processing. These particles often exhibit complex morphologies and modified microphysical properties resulting form in-cloud chemical transformations, such as the formation of sulphate and oxalate. Such changes influence their cloud-forming ability and optical properties relevant to climate processes (Zhang et al., 208, 2020, 2024).

Author's contribution

All authors have significantly contributed to the conception, development, and completion of this work.

7. Conclusion

The development of advanced predictive framework for radioactive aerosol dispersion is critical due to the significant environmental and public health risks associated with both natural and anthropogenic release. Accurate dispersion modelling is essential for reliably forecasting the transport and deposition behaviour of radioactive particles in the complex interactions among key physical processes, including transport dynamics, decomposition mechanism, charge evolution, radioactive decay, and aerosol coagulation. Electrical charge represents a crucial yet often overlooked factor in current modelling approaches, despite its substantial influence on aerosol transport behaviour. Accurate prediction of steady-state surface charge distributions under both typical and extreme atmospheric conditions necessitates a

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detailed understanding of particle charging mechanisms. In radioactive aerosols, intrinsic processes such as self-charging and diffusion charging coexist with external mechanisms, including field and triboelectric charging. The interplay and competition among these processes ultimately determine the accuracy of model predictions. This highlights the need to move beyond conventional modelling approaches that neglect electrostatic interactions and treat radioactive aerosols as analogous to neutral, non-radioactive particles. Accurate trajectory predictions also depend on understanding particle size distributions, which may evolve during atmospheric transport due to microphysical processes such as aggregation and fragmentation. Incorporating population balance models into simulations is therefore essential to capture these dynamic changes. An additional, largely unexplored aspect is the resuspension of deposited radioactive particles induced by saltation processes and electric fields generated during dust storms. Incorporating such mechanism into dispersion and deposition models would improve predictive accuracy by accounting for the redistribution of radioactive material to new locations. Overall, this study provides a critical evaluation of recent advancements, identifies key technological gaps, and outlines future research, emphasizing the importance of integrating charge dynamics with transport processes in atmospheric radioactive aerosol modelling.

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