

The Chemistry of Atmospheric Aerosols: At the Nexus Between Climate, Energy, and Air Quality

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Abstract: Atmospheric aerosols can be emitted directly as particles or formed in the atmosphere from phase transitions of gaseous compounds with low enough vapor pressure. During their lifecycle in the atmosphere, aerosols undergo multiphase changes, altering chemical composition, reactivity, physical and optical properties, ultimately influencing how they impact climate, human health and ecosystems. The understanding of the chemical processes in the atmosphere is crucial to assess these effects. Here we provide a brief overview on relevant aerosol chemical processes and measurement techniques with no claim to completeness and describe the Swiss contribution to the European infrastructure ACTRIS for long-term monitoring and its relevance for the research field.

Keywords: Aerosols · Climate · Energy · Human health



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1. Introduction: Anthropogenic and Natural Chemical Components in the Atmosphere

The chemistry of the atmosphere plays a fundamental role in many of the big societal challenges humankind is facing today. The use of fossil fuels as an energy source since the beginning of industrialization, but prominently since the 1950s, has led to increasing CO₂ emissions into the atmosphere^[1] and concentrations higher than in the last 800,000 years.^[2] A prominent consequence is the rise in global average temperature, currently 1.17 °C ±

0.05 °C above the average level from 1951 – 1980,^[3,4] with implications also for the chemistry of the atmosphere.

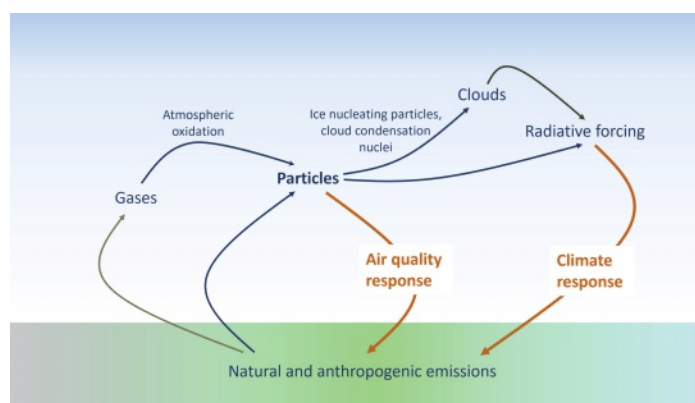


Fig.1. Natural and anthropogenic emissions of gases and particles to the atmosphere, their reactions and effects on air quality and human health.

Combustion of fossil and other fuels (*e.g.* solid fuels such as wood), more precisely incomplete combustion, also emits a large number of trace gases and aerosol particles, which further influence the composition, chemistry and reactivity of the atmosphere, and can be harmful to humans and ecosystems,^[5] see also the review by Kalberer *et al.* in this issue.^[6] Compounds include gases such as CO, NO_x, and volatile organic compounds (VOCs, *e.g.* C₇H₈ (toluene), C₆H₆ (benzene)), or particulate matter consisting of *e.g.* black carbon (BC, soot), organics or inorganic salts. Further anthropogenic emissions to the atmosphere include NH₃ (ammonia) from agricultural processes (see also review by Ammann *et al.* in this issue),^[7] and SO₂ from industrial emissions (and sulfur-containing fuels), among others.

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Further complicated is the picture by natural emissions, which are responsible for a background concentration of trace gases and aerosol particles already present in preindustrial times – the quantification of which is highly challenging and highly important, as it serves as a baseline for the estimation of anthropogenic climate change and pollution.^[8] Natural emission sources include: i) the biosphere with emissions of *e.g.* primary particles such as pollen, and VOCs such as isoprene (C₅H₈), monoterpenes (C₁₀H₁₆), and sesquiterpenes (C₁₅H₂₄); ii) deserts and soils of the Earth's surface emitting dust particles; iii) the oceans emitting sea spray particles consisting of sea salt (NaCl and other alkali metals and alkaline earth metals)^[9] and organics, as well as gaseous halogen and organic sulfur species. Even though we term them 'natural emissions', they are influenced by anthropogenic climate change, as the rise in atmospheric and ocean temperature leads to changes in the cryosphere and the marine and terrestrial biospheres,^[10] together with changes in land use altering emissions and reactivity.

Emitted gases can undergo oxidation reactions in the atmosphere, leading to products that are less volatile and thus have the tendency to contribute to particulate matter *via* condensation.^[11,12] The feedbacks between emissions and a changing climate influencing emissions and particle formation processes are a very important aspect in the quantification of climate change and air pollution (Fig. 1).^[13]

The rise in global average temperature is to some extent (~25%, model estimates range from almost no effect to ~twice that percentage)^[14] masked by atmospheric aerosol particles, which reflect a portion of the incoming short-wave solar radiation back to space, and in addition influence cloud radiative properties. These effects depend on aerosol physicochemical properties.^[15–17]

2. The Chemistry of Atmospheric Aerosols

Formally, the definition of atmospheric aerosols is a suspension of particles in air, which includes the gas and condensed phase. Consequently, the chemistry of atmospheric aerosols involves reactions in the gas and particle phase, multiphase reactions, as well as composition-dependent gas-particle partitioning of semi-volatile species. The multitude of natural and anthropogenic sources, species emitted into the atmosphere as described in the previous section, and reactions in different phases make atmospheric aerosols a highly complex chemical system.

2.1 Atmospheric Reaction Types, Aerosol Formation and Aging

As visualized in Fig. 1, oxidation reactions play a central role in atmospheric aerosol chemistry. Directly emitted gases are often rather volatile, their emissions thermodynamically favoured. Upon emission, some of these gases get oxidized, resulting in more functionalized products with lower volatilities and higher tendencies to condense, hence, to form new particles or contribute to pre-existing particle mass. Particles (whether directly emitted or formed in the atmosphere) undergo further chemical transformation in the atmosphere (termed 'aging') through multiphase- or condensed-phase reactions. Note that here we largely exclude the discussion of aqueous-phase oxidation of dissolved compounds in cloud droplets (see *e.g.* ref. [18]) and focus on the in comparison highly concentrated aerosol particles. The properties of aerosols resulting from this multitude of reactions determine their reactivity, solubility, volatility, and phase state, and ultimately their fate and effects on air quality and climate.

Photochemical reactions,^[19,20] driven by energetic input from solar radiation, are critical for the chemistry of the atmosphere and aerosols as they lead to the formation of radicals *via* photolysis. Radical-initiated atmospheric oxidation pathways are one of the key foci in organic aerosol research, as they play a crucial role in organic aerosol formation. A particular challenge for source identification lies in the fact that intermediate oxidation products of

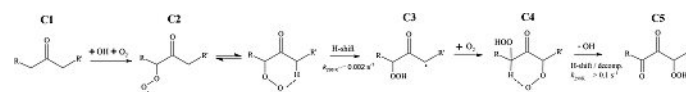
different precursors can react with each other (*e.g.* the formation of peroxide accretion products from peroxyradicals)^[21] and thus source fingerprints are smeared.

The multiphase oxidation pathways of inorganic species such as NO_x and SO₂ forming nitric acid (HNO₃) and sulfuric acid (H₂SO₄) are fairly well understood. Acid-base reactions are then *e.g.* relevant for the formation of inorganic aerosol species ammonium sulfate, -nitrate, and -chloride *via* reactions of NH₃ with H₂SO₄, HNO₃, or hydrochloric acid (HCl).

The hydroxyl radical (OH) and nitrate radical (NO₃), together with ozone (O₃), are the main atmospheric oxidants for organic and inorganic compounds (in the cloud aqueous phase also hydrogen peroxide, H₂O₂, is of relevance), with complex multi-generation reaction mechanisms leading to a distribution of intermediate and termination products with varying properties. The concentrations of these oxidants vary with time of day, season, cloudiness and location, consequently dominating oxidation pathways can differ. For example, the reactions of α -pinene, a monoterpene emitted by vegetation, with O₃ or NO₃ (adding to the double bond) lead to night-time aerosol formation. Photochemical reactions with OH-initiated oxidation dominate during the day.^[22]

2.2 Autoxidation and New Particle Formation

A recent milestone in atmospheric chemistry has been the seminal discovery of the role of autoxidation in the atmosphere, whereby intramolecular H-shifts and the subsequent rapid addition of O₂ lead to the formation of highly oxygenated molecules (HOMs, Scheme 1).^[23]



Scheme 1. Autoxidation mechanism, in oxidation of ketones initiated by OH forming a peroxy radical (C₂) undergoing H-shifts and addition of O₂ (C₄). Figure from Bianchi *et al.* ref. [23], Creative Commons CC BY 4.0.

The detection of HOMs has been pivotal for the elucidation of atmospheric nucleation mechanisms.^[24,25] This clustering of molecules forming a new particle (termed new particle formation, NPF) is a phase transition of atmospheric gases to the condensed phase *via* overcoming an energy barrier associated with creating a new interfacial area between two phases. It is an important particle source not just in today's atmosphere, but also during pre-industrial times.^[26] NPF is relevant for particle populations' size distributions. While particle mass concentration directly follows from phase equilibrium, the number and size distribution of particles strongly depends on the kinetics of the phase transition (nucleation in competition with condensation on existing particles) and on particle coagulation frequency. This is of direct relevance to environmental impacts of aerosols, as most processes depend on number and size of the particles in addition to the particulate mass concentration.

After initial clustering, particle growth proceeds *via* condensation of oxidation products in the atmosphere (Fig. 2).^[27–29]

The review by Dada *et al.* in this issue^[30] gives further details on the mechanisms of NPF, and specifically, their investigation in a laboratory setting.

2.3 Condensed-Phase Reactions

Whereas we have briefly touched upon phase transitions for aerosol formation in the previous chapter, we will not dwell further on the kinetics of gas – aerosol mass transfer and the thermodynamics of gas – particle partitioning. Multiphase chemical processes in the atmosphere, in the context of which particle-

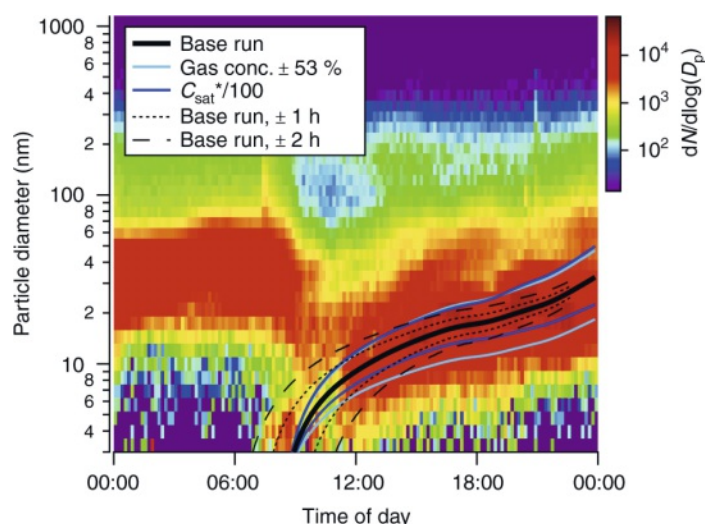


Fig. 2. Particle size distribution measurements show particle growth after new particle formation (NPF) in Hyytiälä, Finland, on April 16th, 2014. The lines are modelled particle growth rates using measured concentrations of precursor gases as model input. Figure from Mohr *et al.* [28], Creative Commons CC BY 4.0.

phase reactions are often discussed, are reviewed in detail by Markus Ammann *et al.* in this issue.^[31] Here we will focus on a few aspects of particle-phase reactions relevant for the assessment of their impact.

Atmospheric aerosol particles, of diameters from the nm to the μm scale, are very small reactors whose properties and chemical reactions are mutually influenced. The phase state of aerosol particles, *i.e.* liquid, solid, viscous, or phase separated, *etc.* greatly influences diffusion limitations and thus particle-phase reactions.^[32] At moderate to high ambient relative humidity (RH), depending on particle composition between around 40 to 80% RH, the soluble components of aerosol particles transform to aqueous solution droplets. The aqueous solution can serve as a matrix for absorption of trace gases followed by heterogeneous chemical reactions. A discussion on aqueous-phase chemistry in cloud and fog droplets or hydrometeors, and their influence on aerosol chemistry is beyond the scope of this article, and we refer the reader to selected reviews.^[33,34]

Acidity or pH of particles present as aqueous solution droplets catalyses accretion reactions, determines acid-base reactions, reaction rates, and the phase partitioning of semi-volatile gases.^[35] Phase state and acidity have implications for the atmospheric lifetime of aerosols and thus their impacts on human health, climate and ecosystems.

Further relevant are particle-phase accretion or oligomerization and decomposition reactions, as they can change molecules' volatility,^[36,37] with examples for accretion products being ester dimers,^[38] and photochemical processes, *e.g.* photolysis of nitrates^[39] or organics.^[40]

3. The Study of Aerosol Chemical Composition and Reactions

The study of aerosol chemical composition and reactions encompasses laboratory experiments simulating processes of the real atmosphere, as well as short- and long-term field observations in different environments. Experimental and observational results are interpreted in the context of theoretical and numerical work using models from molecular scale to process-level and the global scale. In the following we give a brief overview on analytical instrumentation used for aerosol chemical composition analysis and present a small chapter on a European infrastructure for long-term observations.

3.1 Analytical Instrumentation

The rapid advancement in analytical techniques and instrumentation in the last couple of decades has greatly accelerated the rate of discovery and quantification of aerosol chemical processes. For this development to be relevant for atmospheric chemical processes, a number of aspects are of particular importance.

First of all, the sample sizes are small. An atmospheric aerosol particle consists of $\sim 10^{12} - 10^{15}$ molecules, and the mass of a particle ranges from $\sim 10^{-20} - 10^{-6}$ g. Trace gas concentrations in the atmosphere are at the parts per billion (ppb) to parts per trillion (ppt) levels. Such low concentrations require instrumentation that is sensitive and covers the full range from gases and clusters through nanoparticles, ultrafine particles, accumulation mode particles to coarse mode particles.

Second, individual aerosol particles represent a highly complex mixture, as detailed in previous sections. Taking the example of organic compounds, particles can consist of tens to thousands of compounds with varying functionalities (*e.g.* carboxylic acids, peroxides, alcohols^[41]) to name a few) and properties. This complexity often requires a choice of analytical techniques between selective and non-selective, with the 'ideal' instrument detecting the entirety of chemical complexity in the atmospheric aerosol mix.^[42]

Third, the atmosphere is a highly dynamic and reactive system, and the chemical composition of gases and particles changes accordingly. Hence, ideally, processes can be observed in real time, and at Hz rates, or even faster when trace gas fluxes are measured. Consequently, measurement techniques with high time resolution are paramount for further elucidating atmospheric chemical processes.

A technique that has become widely deployed in the field of atmospheric chemistry is aerosol mass spectrometry. It allows for highly time and chemically resolved measurements of the atomic composition of gases and particles (after vaporization, desorption or dissolution), from the bulk to the individual particle level.^[43–45] The use of mass spectrometric techniques has been pivotal for many of the findings presented in Sections 2 and 4. Several review articles have appeared in recent years on this topic.^[46–48]

Downsides of the high-performing analytical techniques are their complexity and price. The latter can make their purchase or deployment impossible for institutions with limited financial resources, and consequently measurements providing novel insights are over-proportionally from institutions and regions in the Global North. The complexity of these instruments effectively disables long-term deployments, as the person power and resources to run these instruments continuously are considerable. Hence, long-term observations are always a trade-off between complexity and feasibility.

3.2 Long-term Observations: The European Infrastructure ACTRIS

Observations of atmospheric trace contaminants led to the understanding that anthropogenic emissions can have far-reaching consequences for the environment (compare also review by Baltensperger in this issue).^[49] Once in the air, they are transported across national borders on continental up to global scales. Accordingly, the need for internationally coordinated monitoring and research came onto the agenda of the United Nations as early as the mid-twentieth century. Here we present as an example the Aerosol, Clouds and Trace Gases Research Infrastructure (ACTRIS), which provides coordinated observations of short-lived atmospheric constituents and clouds. The importance of these long-term observations applies to Earth observations in general, with the Integrated Carbon Observation System Research Infrastructure (ICOS) as an example for tracking the carbon cycle across biosphere, hydrosphere and atmosphere. The full potential of Earth Observations can only be completely exploited by pro-

viding data openly through repositories following the FAIR guiding principles,^[50] where the environmental domain is in a leading role.^[51]

The Swiss contribution to ACTRIS is illustrated in Fig. 3. Central facilities are a first building block playing a pivotal role in ensuring FAIR data. The unit of the topical Centre for Aerosol Remote Sensing located at Davos works towards the calibration and traceability of all ACTRIS sun photometers linking the ACTRIS scale with the reference of the World Meteorological Organization.^[52] The unit of the topical Centre for Reactive Trace Gases *in situ* Measurements located in Dübendorf works towards quality assurance and control of trace gas observations, and supports development of new data products from ground-based remote sensing of Fourier transform infrared solar absorption spectra such as these acquired at the Jungfraujoch observatory.^[53]



Fig. 3. The Swiss contribution to ACTRIS includes central facility units (blue), an atmospheric chemistry simulation chamber (orange) and continuous atmospheric observatories (red).

Observatories are a second building block of ACTRIS. Each of the three observatories in Switzerland has its own speciality depending on environment and co-location with other networks. The Payerne observatory has one major focus in vertical profiling of the atmosphere in addition to a wide range of ground *in situ* measurements. Extension of aerosol and cloud remote sensing capabilities currently is at its final implementation stages, to contribute data in near real-time for coordinated calibration and validation of data products of the atmospheric lidar on the EarthCARE satellite, which has been launched in May 2024. The Jungfraujoch observatory at the high-altitude research station is ideally suited to investigate aerosol and trace gases in the lower free troposphere and their interactions with clouds. Particle chemical composition plays a key role for their ability to trigger ice formation in supercooled clouds. Saharan dust was shown to make the dominant contribution to ice nucleating particles at around -30 °C .^[54] Time-resolved and continuous measurements of the composition of particulate organic matter are essential for source apportionment (see Section 4). Coordinated analyses of data from across the ACTRIS network showed for example that solid fuel combustion makes a substantial contribution to atmospheric particulate matter in Europe.^[55] The Beromünster observatory is ideally located for trace gas measurements representative of Switzerland, which serve, together with data from other European sites, for validation of respective model simulations.^[56]

Facilities and observations available through the ACTRIS network also serve as a basis for additional research. Coordinated investigation of transport and degradation of pesticides in the atmosphere was such a planned activity,^[57] and it identified the need to revise assessment of their risks. By contrast, the COVID-19

lockdown caused unforeseen emission reductions for black carbon up to 20%–40% in several European countries,^[58] where numerous ACTRIS observatories serving as receptor sites provided data as input for the top-down emission estimate approach based on Bayesian inverse modelling.

Simulation chambers such as the PSI Atmospheric Chemistry Simulation Chamber are a third building block of ACTRIS to investigate sources and chemical transformation of pollutants under constrained conditions. They can *e.g.* provide tracer species and molecular footprints of a wide range of combustion sources to aid interpretation of ambient observations,^[59] or point to the importance of considering condensed-phase chemical reactions to describe oxidation of organic particulate matter during its lifecycle.^[36]

4. Nexus Air Quality – Climate – Net Zero

The pressing societal issues of our time, climate change, net zero, and air pollution, are tightly connected through atmospheric chemistry. The frequent co-emission of climate and air pollutants can result in co-benefits between air pollution and climate mitigation strategy (*e.g.* net zero) effects, *i.e.* a reduction in CO_2 emissions also leads to a reduction in emissions of air pollutants.^[60] However, there are also trade-offs, *e.g.* the potential decrease of marine cloud brightness and hence reduced albedo due to reduced sulfur emissions from ships.^[61]

4.1 Source Apportionment

Relevant for effective mitigation strategies of climate and air pollutants is a quantitative apportionment of atmospheric constituents to their sources. The large number of different compounds emitted from many different sources and being modified by numerous atmospheric chemical processes on short timescales makes source apportionment challenging. Emissions lose their initial source fingerprint, and their unique markers for ‘natural’ or ‘anthropogenic’ origin, requiring complex data and analytical tools to disentangle source contributions.

Source apportionment *via* receptor modelling has been proven successful in elucidating and quantifying the sources of atmospheric aerosols, in particular positive matrix factorization (PMF) of the data acquired by mass spectrometers.^[62] This method has the advantage over the more traditional chemical mass balance models that no *a priori* information is required. By means of factorization data reduction, PMF yields a linear combination of a number of stable factors representative of the emissions of a certain source, and their time series, respectively. This technique has allowed to assign not only broad chemical particulate species, such as inorganic nitrate, sulfate, ammonium, sea salt, dust, and organics to their sources, but even to separate organics into primarily emitted organic particles from *e.g.* biomass burning, cooking and traffic, and secondarily formed organic compounds. The result is an aerosol phenomenology for large parts of Europe and other places in the world *e.g.* Chen *et al.*^[55], which is of high relevance for: i) the development of air pollution mitigation strategies, has quantified *e.g.* the significant contribution of solid fuel combustion to aerosol mass; and ii) the assessment of composition change of particulate matter through net zero strategies.

5. Conclusions and Outlook

We conclude this review article on aerosol chemistry, which has no claim to completeness, with a number of points we deem relevant for future developments, scientific foci, and investigations in the field of aerosol chemistry.

How much complexity is possible/required in models? This review has largely ignored the discussion on methods, and particularly on theoretical tools. A fundamental understanding of observations, and projections of future climate, pollution, health, and socioeconomic scenarios requires the development and use of

model frameworks along the entire scale chain. The discussion on consistent and relevant simplifications of the complexity of atmospheric chemical processes for representation in models requires enhanced collaboration between experimentalists and modellers.

The advances in analytical instrumentation and their integration in monitoring networks resulting in data at unprecedented temporal and chemical resolution provides new opportunities for air quality research and exposure studies. The investigation of health effects of atmospheric constituents, and especially particulate matter, may thus require a shift in paradigm from regulating mass concentrations to including information on number, size, and especially chemical con.^[6]

Changes in ecosystems through climate change, as well as net zero strategies have profound implications for atmospheric processes and chemistry. Adamant long-term monitoring efforts are required for the observation of effects and trends.

We hope the reader may feel inspired to join our quest to pursue a fundamental understanding of atmospheric chemical processes in order to contribute knowledge relevant for making effective climate change and air pollution policies. In this regard, the intended modification of weather and climate through the release of particles to either influence cloud formation and precipitation, or to reflect solar radiation back to space (solar radiation management), are topics we have not discussed in this review, as their complexity requires research efforts that span the entire field of atmospheric sciences. The chemical composition of aerosol particles, their formation and transformation processes, will continue to evolve with the perturbations, inadvertent or intended, humans cause in the atmosphere. Continuous efforts to monitor, quantify and understand these changes will remain relevant to assess the impacts on human health and climate.

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