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Multiphase Chemistry in the Atmosphere

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Abstract: Earth's atmosphere comprises a complex mix of gas and condensed phases, where condensed phases facilitate multiphase chemical reactions that would not occur in the gas phase alone. These reactions drive dynamic physical and chemical processes across various spatial and temporal scales, playing a crucial role in the cycling of atmospheric trace constituents. Multiphase chemistry significantly influences geochemical cycles, human health, and climate. This review focuses on the chemical steps governing the cycling of important species, such as halogens, reactive nitrogen, and organics, within aerosol particles, a key type of atmospheric condensed phases, and at condensed phase–air interfaces. These interfaces include mineral oxides, ice, and aqueous solutions found in particulate matter, clouds, snow, and on oceanic and terrestrial surfaces. This review also discusses the important role of redox chemical cycling, the hydrogen bonding network and water activity in these processes.

Keywords: Anthropocene · Atmosphere · Chemistry · Multiphase

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CECs in natural waters. In her PhD she focused on the multiphase chemistry initiated by iron(III) carboxylate photolysis.

Kevin Kilchhofer obtained his MSc in Atmospheric and Climate Science at ETH with a focus in atmospheric composition, in which he conducted cloud-processing experiments of ice nucleating particles. During his PhD at PSI, he is investigating photochemical aging of metal-containing organic aerosol.

Andrés Laso obtained a degree as a technician in mechanical engineering in 2015. He worked in the Institute of Physical Chemistry at ETH for over eight years, before joining PSI in 2017. He is vital for the conceptual development, design and construction of our *in situ* spectroscopy tools.

Luca Longetti obtained his MSc in Physics in 2014 at Sapienza University of Roma. In 2020 he earned a PhD in Photonics from EPFL and worked there as a postdoctoral fellow on photoinduced phenomena until he joined PSI in 2022 to investigate atmospheric relevant systems by means of X-ray spectroscopy.

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1. Introduction

1.1 *The Atmosphere as a Multiphase System*

Earth's atmosphere interacts with various condensed phase materials, both in the air as suspended particles and on the ground as soil, snow, vegetation, and urban surfaces (Fig. 1). Particles and ground surfaces are crucial for the gas-phase composition, geochemical cycling and atmosphere–climate interactions.[1] These complex condensed materials have diverse chemical and physical properties, enabling chemical processes not possible in the gas phase alone. For example, photolysis of nitrate in aerosol particles is a significant source of gas-phase nitrogen oxides.[2] Such effects are important even though the particle number concentration is low, typically ranging from a few hundred cm^{-3} in remote marine air to 10^6 cm^{-3} in urban regions.[3] As another example, snow is porous with an internal surface area on the order of 10^1–10^3 cm^2 in a 1 cm^3 snow volume,[4] and strongly impacts atmospheric halogen chemistry.[5]

Photochemically-generated oxidants like ozone (O_3), the hydroxy radical (OH), and the hydroperoxy radical (HO_2) drive essential steps in atmospheric trace gas cycling. Their reaction products with other trace gases are generally less volatile, more soluble, and acidic than their precursors, driving partitioning to aerosols, clouds, or ground surfaces through dry and wet deposition. This limits their accumulation in the atmosphere and provides nutrients to ecosystems but also leads to acidification, eutrophication, and entry of toxins (*e.g.* mercury) into the marine and terrestrial food chains.

1.2 *The Role of Water*

Water is ubiquitous in the atmosphere. It is present as a liquid in aqueous particles and cloud droplets,[6] as crystalline ice in snow and clouds,[7] or as a molecular layer covering solids or less hydrophilic organics.[8] Cloud droplets are highly dilute with a water activity close to that of pure water, 1.0. The aqueous fraction of aerosol particles comprises a complex mix of organic solutes from oxidation of volatile organic compounds (VOCs) and inorganic electrolytes from the oxidation of inorganic precursor gases, partially neutralized by ammonia (NH_3). As a consequence, aerosol particles span a wide range of water activities, linked to the relative humidity (RH), which strongly influences kinetics, solubility, and viscosity with significant implications for transformative exchange dynamics with the gas phase and for the reactivity within the condensed phase.[9] The high solute strength of aqueous aerosol particles often prevents them from forming a single homogeneous phase.[10] Complex multiphase transformations within

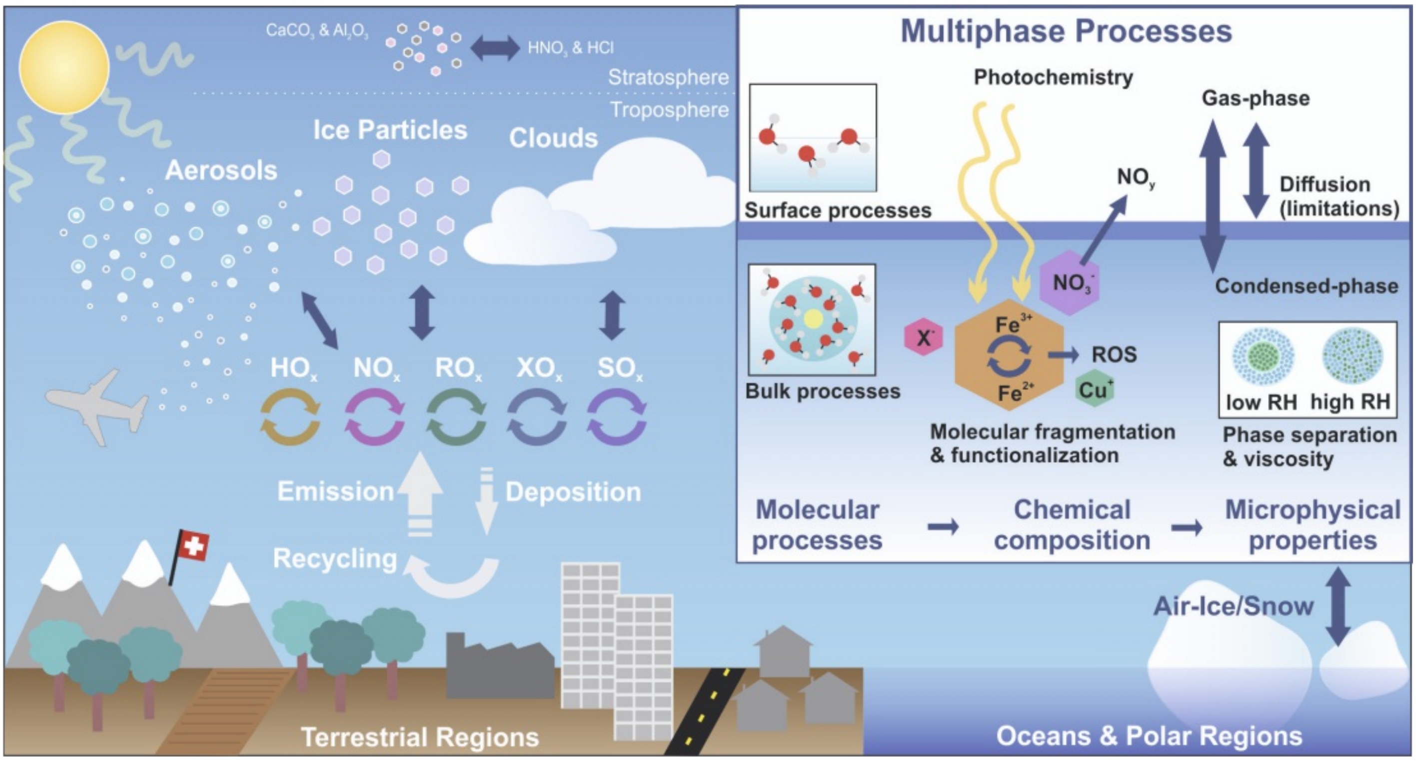

Fig. 1. The atmosphere as a multiphase chemical reaction system. HO_x, NO_x, RO_x, XO_x and SO_x refer the family of hydrogen peroxy radicals, nitrogen oxides, oxygenated organics with their peroxy and alkoxy radicals, halogen oxides and sulfur oxides, respectively.

the particles modify their health effects upon inhalation, driven by the toxicity, mutagenicity, or oxidative stress potential of particle components.[11]

Hydrogen bonds hold aqueous condensed phases together defining their physicochemical properties, solute hydration and chemical reactivity.[12] The hydrogen bonding network at the liquid-gas interface is highly asymmetric,[13] which uniquely links chemical composition with surface tension,[14] and is fundamentally important for the partitioning of trace gases,[15] and for many surface-specific properties and processes.[16] On ice, when temperatures approach the melting point, the increased flexibility of the hydrogen bonding network, often referred to as pre-melting or *quasi*-liquid layer (QLL), has been a topic of considerable debate.[7] Understanding the role of hydrogen bonding on aqueous and ice phases provides a foundation for exploring how multiphase chemistry influences chemical processes associated with aerosol particles, cloud droplets, ice, and snow cover.

2. Atmospheric Multiphase Chemistry

2.1 *Multiphase Chemistry of Nitrogen Oxides*

Nitrogen oxides are pivotal for O_3 formation in the gas phase by coupling with OH and HO_2 radical reaction cycles. A termination product of these cycles is nitric acid (HNO_3), which is either formed directly from the reaction of nitrogen dioxide (NO_2) with OH, or *via* the hydrolysis of dinitrogen pentoxide (N_2O_5). HNO_3 efficiently interacts with ambient surfaces, such as mineral dust,[17] aqueous particles[18] or ice surfaces.[19] The adsorption of HNO_3 to ice has attracted considerable attention because it can represent a significant loss of nitrogen oxides (and thus of O_3) from the upper troposphere. Using ambient pressure X-ray photoelectron spectroscopy (APXPS),[20] we tracked HNO_3 at the ice surface at low, atmospherically relevant surface concentrations (Fig. 2).[19] Coupling APXPS with electron yield near edge X-ray absorption fine structure (NEXAFS) spectroscopy we found that nitrate was fully hydrated at the ice surface, while at the same time a slight perturbation of the hydrogen bonding structure occurred when compared to pure ice. The small perturbation induced by HNO_3 can be rationalized by water molecules forming a hydration shell for each HNO_3 without affecting the structure of the remaining ice.[19] We obtained similar results for HCl,[21] formic acid,[22] and acetic acid.[23] We are currently working on establishing the dissociation degree of HNO_3 on pure ice and in the presence of neutralizing NH_3 or amine[24] using APXPS. We determined earlier that HCl is fully protonated at the very top of the interface, while dissociated chloride populates the interfacial region underneath by several nanometer.[21] Interfacial acid-base chemistry has remained a highly debated topic, which is not surprising, given the importance of acid-base chemistry in atmospheric chemistry.[25]

The presence of HNO_3 and HCl on cold surfaces is also relevant to climate intervention ideas, such as the injection of sulfuric acid particles into the stratosphere to increase scattering of sunlight back to space, thereby reducing the radiation reaching Earth's surface.[26] However, substantial concerns regarding adverse effects on stratospheric dynamics, precipitation formation, and on the extent of the stratospheric O_3 layer, have been raised. Increased particle surface area in the stratosphere in mid-latitudes or addition to polar stratospheric clouds (PSCs) over the poles would allow more efficient heterogeneous reaction of chlorine and nitrogen containing reservoir gases, which yield molecular chlorine, the precursor to O_3 depleting chlorine atoms. HCl and HNO_3 play critical roles in these processes. As an alternative to sulfuric acid, injection of solid particles, such as alumina or calcite, is currently being discussed.[27] To support assessment of these climate interventions, we have recently started to employ APXPS to study HNO_3 and HCl uptake on calcite particles under stratospheric conditions. Additionally, we aim to quantify the thickness of the resulting calcium nitrate layer, which is especially relevant for the impact on the O_3 layer.[28]

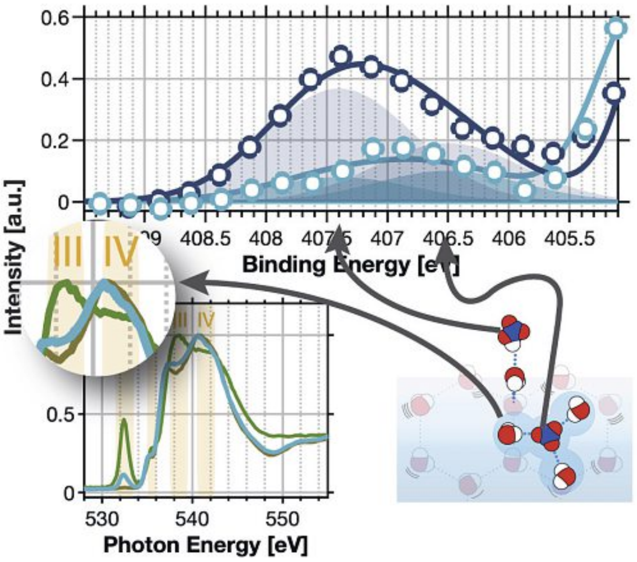

Fig. 2. HNO_3 adsorption and dissociation at the air-ice interface. Upper plot: Photoemission spectra of NO_3^- at 406.5 eV and HNO_3 at 407.4 eV at –30 °C (light blue) and –50 °C (dark blue). Lower left: The NEXAFS spectrum of ice with adsorbed HNO_3 (light blue line) is very similar to pure ice (brown line). The small changes in region III shows that some of the water molecules at the interface are arranged similarly to those in a nitrate solution (green line). Reproduced from ref. 19 with permission from the Royal Society of Chemistry. Illustration: Emerging picture of trace gas species adsorbing to ice with dissociation and formation of solvation shells deeper in the interfacial region.

For tropospheric chemistry, OH radicals are key drivers of oxidation. Apart from O_3 photolysis, another source of OH is the photolysis of gas phase nitrous acid (HNO_2, usually denoted as HONO) at wavelengths between 300 and 400 nm. HONO can be formed by the reaction of OH with NO (thus the reverse of photolysis) in the gas phase. It can also be formed *via* multiphase processes of NO_2, such as the reduction of NO_2 by electron-rich phenols and methoxy phenols.[29] These species are emitted by biomass burning (from pyrolysis of lignin), or occur in soil as part of humic material. The electron transfer from these species to NO_2 can further be accelerated by the excited triplet states of aromatic carbonyls acting as photosensitizers.[30] This may represent one pathway to explain the high HONO levels observed during the day. Another potentially important source of HONO is the decomposition of peroxy nitrate that forms from the reaction of HO_2 and NO_2 in the gas or condensed phase.[31] We are currently investigating whether transition-metal induced photochemistry, which delivers HO_2 radicals (discussed in the next section), could be an indirect route to promote HONO formation *via* the peroxy nitrate pathway (PhD thesis L. Iezzi, in preparation).

The photolysis of nitrate presents an additional pathway for HONO and NO_X production, and at the same time represents a recycling process from nitrate-containing aerosols back to the gas phase (termed 'renoxification'). Renoxification is of particular interest in remote regions, where NO_X sources, other than long-range transported aerosol, are missing.[2a] However, since the nitrate absorption cross section drops off sharply above 310 nm, where the solar actinic flux increases, many studies have suggested enhanced photolysis rates of HNO_3 or nitrate at surfaces[32] in order to explain the observed renoxification rates. These enhanced photolysis rates are thought to result from incomplete hydration shells which reduce the recombination of initial photodissociation products, and thus enhance the quantum yield. Though, as mentioned with our work on ice,[19] we found nitrate at the surface exhibited a rather full hydration shell. We are currently working on a more detailed spectroscopic characterization of nitrate on ice[33] and in aqueous solution, to obtain more insight into the local hydrogen-bonding environment of nitrate and the role of ion-pairing (L. Longetti, in preparation). An additional avenue to initiating nitrate reduction (as an alternative explanation for efficient renoxification and HONO formation), is *via* indirect photochemical processes as discussed above for NO_2. We are considering Fe(III) carboxylates (Fig. 3, L. Iezzi, PhD thesis, in preparation) and biomass-burning derived brown carbon (F. Bao, in preparation) as photosensitizers to drive nitrate reduction under conditions where direct nitrate photolysis is not efficient.

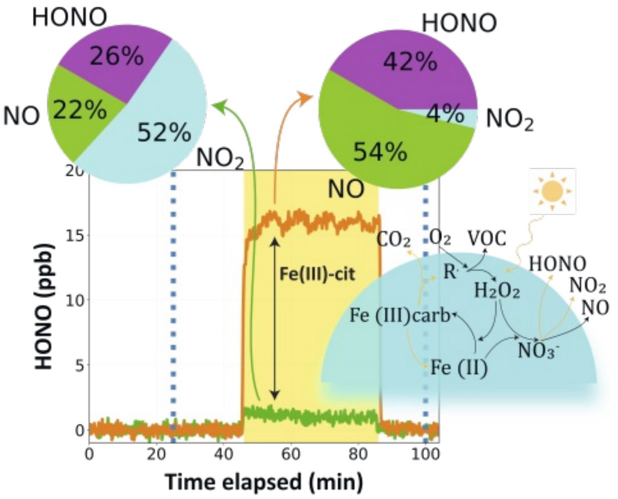

Fig. 3. The production of HONO upon irradiation (yellow shaded period) of a mixed aqueous film of Fe(III) citrate, citric acid and sodium nitrate (orange line) is about an order of magnitude above that in absence of Fe(III) citrate (green line). The dotted lines indicate the exposure of the film to the humidified carrier gas glow (N_2 + O_2 + RH (~80%)) in the dark. The pie charts show the speciation of gas-phase products.

2.2 *Halogen Chemistry*

The halogens (X): chlorine (Cl), bromine (Br), and iodine (I), are important catalytic sinks for O_3 both in the troposphere and the stratosphere.[34] In the cold and shallow boundary layer of polar areas during spring, halogen chemistry can deplete ozone entirely over large regions.[35] Halogen chemistry is, therefore, essential for the regional and global oxidation capacity, and impacts the role of O_3 as a greenhouse gas.[36] Halogens predominantly derive from the oxidation of halide ions in the oceans, in sea salt aerosol, or in snow.[35]

Consistent with earlier work, our flow tube experiments on O_3 oxidation of aqueous mixtures containing bromide have established that a surface process is the rate-limiting step.[37] This was previously attributed to bromide occurring in enhanced concentration at the surface of salt solutions, thanks to its large polarizability, and was based on classical molecular dynamics (MD) simulations.[38] Using a combination of XPS on a liquid jet of these solutions[39] with *ab initio* MD simulations, we demonstrated that bromide lacks surface propensity, as is expected for an ion at the boundary of a dielectric medium. The origin of the surface specific reaction kinetics with O_3 results from a bromide ozonide intermediate (also surface active), which we detected *in situ* by XPS for the first time while dosing O_3 around the bromide containing liquid jet.[37] Since bromide almost exclusively occurs together with surface active organic compounds (either from marine biota or partitioning of VOC oxidation products from the gas phase), we have started to investigate their impact on the abundance of bromide and its oxidation. Specifically, we are interested in surface active compounds with either neutral or charged surfactants.[40]

Building on our earlier discussion of efficient O_3 destruction in polar regions, we studied the temperature dependence of bromide oxidation by O_3 in solutions containing bromide and citric acid. Citric acid was used as a proxy for secondary organic aerosol (SOA), *i.e.* to represent the complex composition of aged secondary particle phase organics present in the coastal polar air, and as deposits in surface snow.[41] We showed that the multiphase reaction with gas-phase O_3 shows an overall decrease in reaction rate rather than an increase as expected with decreasing temperature. This was attributed to a decrease in O_3 solubility and diffusivity with increasing brine concentration.[41] This established an example of how the impact of high solute strength on viscosity and activities starts to dominate the kinetic behavior (Fig. 4).

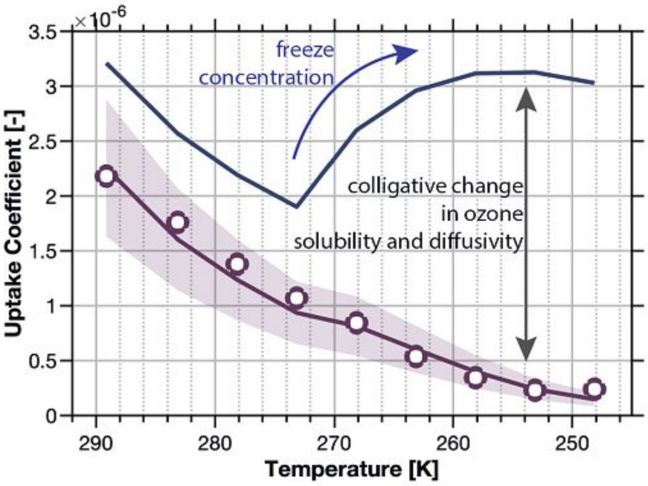

Fig. 4. Kinetics of the oxidation of bromide by gas-phase ozone. The uptake coefficient data (red dots) describe the heterogeneous reaction at varying temperatures and are related to a first-order rate coefficient. The blue line denotes model results based on the increasing concentration of bromide as temperature and humidity decrease. The red line accounts for changes in ozone solubility and diffusivity. The shaded area gives the systematic uncertainty for the parameterization of the colligative properties. Reproduced from ref. [41] with permission from the Royal Society of Chemistry.

Surface snow continuously changes its structure and properties through a process called metamorphism.[42] We showed that metamorphism can change the reactivity of bromide brine within days. It is thought that gas-phase water fluxes driving the metamorphism, burying the brine deeper in the snow crystals, making it inaccessible to gas-phase O_3 and thus inert to this oxidation process.[43] This result may help explain the varying reactivity of snow of different ages.

Bromide oxidation occurs not only by O_3 but also by other oxidants. These include the triplet excited states of aromatic carbonyls, to which halide ions act as electron donors.[30b] We demonstrated substantial oxidation of iodide and bromide from mixtures of citric acid, bromide or iodide, and imidazole carboxaldehyde (IC).[44] IC is a product of the multiphase reaction of ammonium with glyoxal, an important oxygenated VOC.[45] In collaboration with the group of Rainer Volkamer at the University of Colorado in Boulder we have explored the photochemically induced redox chemistry of iodide and iodate by Fe(III) carboxylate complexes and other chromophores (PhD thesis L. Iezzi, in preparation; Reza *et al.*, in preparation). We found an abundance of more reduced iodine compounds in the gas-phase, which must be attributed to efficient iodate recycling. Furthermore, we hypothesize this efficient recycling can be extended to the occurrence of particulate iodide in both the troposphere and the stratosphere, despite its short lifetime due to oxidation by O_3.[46] Such recycling of iodate is also relevant as a path to catalytic new particle formation *via* iodine oxide species,[47] which may sustain the formation of Arctic marine clouds essential to the radiative budget in a region sensitive to climate change.

2.3 *Aging of Secondary Organic Aerosol*

The chemical complexity of organic aerosols – which contribute between 20% and 70% of the submicron mass of atmospheric particulate matter[48] – makes it difficult to disentangle the individual reactions occurring during their lifetime in the atmosphere. As such, the ozonolysis of unsaturated organics has provided great opportunity to study the mechanisms, kinetics, and products of organic aerosol aging in detail. For this, we have used a combination of aerosol flow tube (AFT), coated wall flow tube (CWFT), and synchrotron based *in situ* scanning transmission X-ray spectromicroscopy (STXM) experiments,[49] which were coupled with an explicit multilayer model to resolve chemistry and intra-particle transport.[50] The STXM work was conducted using a unique *in situ* cell[51] that allowed for the study of particles under relevant humidity and O_3 concentrations directly at the synchrotron. For example, this has allowed us to elucidate the kinetics during particle-phase state transitions, *i.e.* from a highly viscous (semi-solid or glassy) state at low RH to a liquid-like phase state at high RH.[52]

In follow-up studies, we explored the oxidation of citric acid that was initiated by Fe(III) citrate photochemistry and represents a key mechanism for carboxylic acid removal in the atmosphere. Fe(III) carboxylate complex photolysis initiates peroxy radical chemistry, which is then sustained by catalytic Fenton chemistry through HO_2, OH, H_2O_2, and organic peroxides. This leads to molecular fragmentation, evident by significant mass loss due to the release of higher volatility VOCs and CO_2.[53] Such repartitioning of SOA mass from the particle phase to the gas phase impacts the burden and lifetime of SOA in the atmosphere, with important implications for air pollution and climate. We expect similar processes to occur in the atmosphere when SOA particles become internally mixed with Fe from urban emissions or mineral dust. An important caveat to peroxy radical chemistry is that it requires O_2. The combination of the low O_2 solubility with diffusion limitations leads to anoxic conditions within particles under a wide range of ambient conditions. This was demonstrated by spatially resolved maps of the Fe(III) fraction within particles using STXM indicating the persistent presence of Fe(II) in particle cores.[54]

These Fe oxidation state gradients are also illustrated in Fig. 5 (K. Kilchhofer, PhD thesis, in preparation) as part of follow-up work addressing the role of other transition metals, *e.g.* copper (Cu). Cu is known to strongly interfere with radical cycling and is suggested to be a significant peroxy radical sink. Initial results show that Cu leads to a decrease in the Fe(II) quantum yield during initial photolysis, while at the same time it accelerates reoxidation of Fe.[72]

Recently, we have expanded our kinetics and spectroscopy work to include use of an atmospheric simulation chamber to generate monoterpene-derived SOA. This work demonstrated that Fe(II) in seed particles impacts the formation and aging of SOA. Under dark conditions, *i.e.* in absence of photochemistry, Fe enhanced the organic SOA mass and decreased the SOA volatility, *i.e.* increased the relative dimer abundance through Fenton-driven oligomerization reactions of condensed-phase organic species (Fig. 6). Furthermore, comparing the compositional similarities of SOA for experiments using Fe(II) *vs.* Fe(III), demonstrated that reduction of Fe occurs in the dark when Fe is mixed with SOA.[55] These experiments provide novel insights into the relevance of transition metal induced processing in ambient SOA. They also demonstrate the impact of condensed-phase dark and photochemical SOA aging on the chemical and microphysical properties of the aerosol, and the way it varies under the wide range of atmospheric humidity conditions. Furthermore, we are also exploring the photochemistry of these monoterpene systems (N. Garner, in preparation), as an extension to our work with Fe(III)citrate.

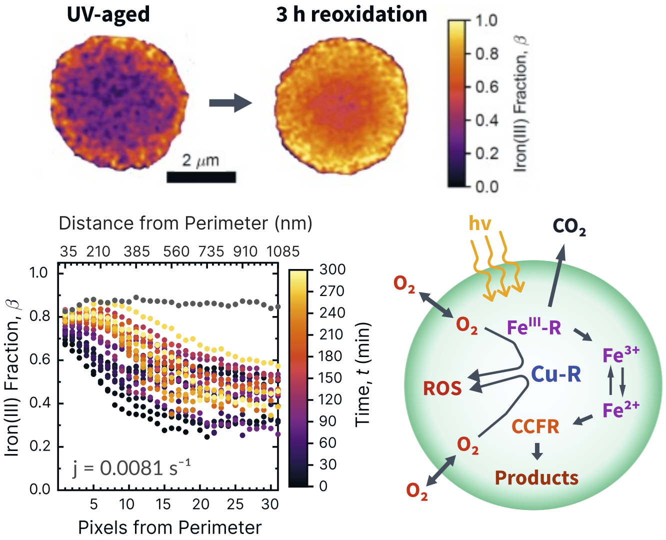

Fig. 5. Reoxidation of Fe(II) in the dark after photolysis of Cu-containing Fe(III)-citrate/citric acid particles. The STXM images on the top were taken directly after UV irradiation (left) and after 3 h of dark reoxidation at 45% RH. The color scale represents the Fe(III) fraction (β). Lower left: β as a function of the number of pixels or distance from the perimeter of particles, now color coded as reoxidation time. The grey dots represent β before UV-aging. Lower right: Scheme showing Fe-Cu-ROS interactions in a highly viscous organic aerosol. The green gradient mimics oxygen availability and its loss due to diffusion limitations in viscous particles. ROS = reactive oxygen species, CCFR = carbon-centered free radicals.

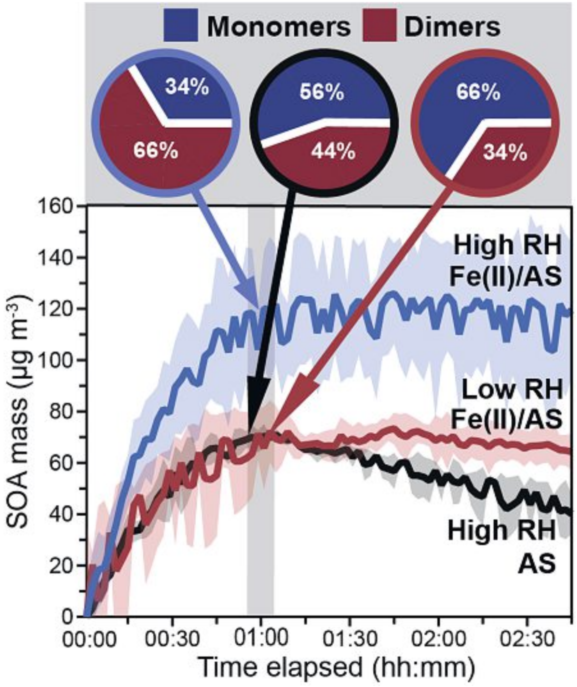

Fig. 6. Evolution of SOA mass formed in the dark and under different RH and seed conditions. The pie charts show the relative composition of SOA ~1 hr after α-pinene injection. Fe(II)/AS= $FeSO_4$ & $(NH_4)_2SO_4$ seed mixture, AS= pure $(NH_4)_2SO_4$ seed. Adapted from ref. 56, distributed under the CC BY 4.0 License.

3. Summary, Outlook, and Perspectives

The aforementioned work combines the microphysical, mechanistic, kinetic, and macroscopic chemical details of multiphase chemistry in atmospheric condensed phases.

Interfacial processes have been a recurring topic, highlighting the role of asymmetric hydrogen bonding driving properties and reactivity. Recently spontaneous radical production and interfacial oxidation has been reported,[56] possibly associated with this asymmetry.[57] Our *in situ* tools based on electron spectroscopy (XPS and NEXAFS) are perfectly suited to simultaneously explore the local structure and chemistry at interfaces.[39a]

Concerning the ice–air interface, our work has focused on chemical characterization and surface specific dissociation of acidic adsorbates that needs considering due to the role of bases such as NH_3 or amines. Moreover, in surface snow, better characterization of multiphase chemistry in distinct compartments, *e.g.* grain boundaries and brine enclosures, is important. Our recent work showing how grain boundaries enhance the uptake of H_2O_2[58] paves the way for future studies. An important aspect to consider is the impact of physical snow dynamics. This not only applies to the halogens, organics, and nitrogen oxides discussed in this review, but also expands to other species such as sulfur compounds. For example, there is significant interest in the mechanism of cloud formation from biogenic emissions in the Southern Ocean, where the role of snow in modifying sea-air fluxes is essentially unknown. We have started to work with the 1-dimensional process model PACT-1D, combined with laboratory experiments, to tackle transport dynamics of trace gases within snow. This work considers snow metamorphism, wind dynamics of the overlaying air, and chemical processes.

We have emphasized that microphysical conditions in particles can cause high ionic strength, salting effects, and diffusion limitation, impacting aerosol and snow chemistry. Another important aspect is liquid-liquid phase separation. SOA type particles can form multiple condensed phases when mixed internally with each other or with primary organic aerosols, which are to a large degree is controlled by the oxidation state of the different components of a mixture.[10,59] The feedbacks between formation of multiple condensed phases and particle aging remain largely unexplored, but can be addressed with our cutting-edge STXM tools. Also here the role of sulfur species – tremendously important in Asian haze episodes[60] and of huge relevance on human health in these densely populated regions – should receive more attention.

Significant research has focused on the physical (optical and radiative) properties of particles, driven by their relevance to Earth's energy budget and climate. As discussed in this review, absorption of light triggers particle-phase radical chemistry. Aerosol particles also enable nano-focusing effects, leading to enhanced photochemical processing.[61] Again, the feedbacks between such processing and the phase state and phase separation has only recently become an emerging topic.[62] The global impacts of these aspects on multiphase processes in the atmosphere remain unresolved.

With the decarbonization of mankind's energy demand, the multiphase chemistry of nitrogen oxides will remain a top priority. Species not discussed in this review include N_2O_5 and NH_3. The future increase of NH_3 may shift the composition of atmospheric reactive nitrogen towards the greater prevalence of reduced nitrogen compounds. This will be addressed in the ETH Domain Joint Initiative ReCLEAN (Reactive nitrogen at the CLimate, Energy, Agriculture, water, and health Nexus, https://reclean.epfl.ch/).

Our *in situ* electron spectroscopy[20] and STXM[52b] tools developed at the Swiss Light Source synchrotron have become mature methods that allow us to tackle cutting-edge research questions related to atmospheric multiphase chemistry. Coupling these powerful tools to atmospheric simulation chamber experiments, where SOA formation and processing can be mimicked under realistic and controlled conditions, will allow us to gain novel, mechanistic insight. Our research has demonstrated the importance of transition metals on the chemical and physical properties of SOA. We are currently using this two-experimental approach to study the redox chemistry of Fe-containing SOA on a single particle level (N. Garner, in preparation), further expanding our understanding of Fenton chemistry on more complex atmospheric systems. For STXM, we successfully pushed experimental temperature limits, and observed and imaged heterogeneous ice nucleation processes for the first time.[63] Similarly, XPS/NEXAFS on liquids and solids can be used to explore the hydrogen bonding structure at interfaces of solids and liquids beyond those of ice,[64] which is relevant for, apart from the chemical reactivity reviewed here, heterogeneous ice nucleation. Thus, molecular-level details of ice nucleation, which is of utmost importance to reduce uncertainties associated with current climate predictions,[65] become a feasible research topic.

We are now applying the chemical and mechanistic knowledge gained from our fundamental laboratory studies to engage in explicit aqueous phase chemical modelling. In collaboration with TROPOS in Leipzig,[66] our aim is to assess the atmospheric implications of our laboratory studies in detail. This also builds on our previous work with process models, which couple transport and chemistry in the condensed phase[50,67] to provide optimized thermodynamic and kinetic parameters (K. Kilchhofer, PhD thesis, in preparation).

A vital prerequisite for this and related modeling efforts, is a sound foundation of experimental data. Freely accessible databases, such as those for gas phase, heterogeneous and aqueous phase reactions[68] provided by the IUPAC Task Group of Atmospheric Chemical Kinetic Data Evaluation, where M. Ammann is a member, are critical to achieve this goal. NASA JPL operates a similar project.[69] To this end, ensuring that data is findable, accessible, interoperable, and reusable is critical. We are developing[70] an open research data management framework based on open-source technologies like HDF5 and Git. This framework is amenable to current data management practices in atmospheric science and interoperates with the requirements of the Aerosols, Clouds, and Trace gases Research Infrastructure ACTRIS.[71]

Overall, continued advancement of our fundamental understanding of atmospheric multiphase chemistry will benefit our knowledge of large-scale processes, such as air pollution and climate change, which are of high societal relevance. A concerted effort encompassing measurements from the laboratory and field, along with modelling will be required to achieve this goal.

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