Opinion: Atmospheric Multiphase Chemistry: Past, Present, and Future

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Abstract

Multiphase chemistry occurs between chemicals in different atmospheric phases, typically involving gas-solid and gas-liquid interactions. The importance of atmospheric multiphase chemistry has long been recognized. Its central role extends from acid precipitation and stratospheric ozone depletion, to its impact on the oxidizing capacity of the troposphere, and to the roles that aerosol particles play in driving chemistry-climate interactions and affecting human health. This opinion article briefly introduces the subject of multiphase chemistry and tracks its development before and after the start of *Atmospheric Chemistry and Physics*. Most of the article focuses on research opportunities and challenges in the field. Central themes are that a fundamental understanding of the chemistry at the molecular level underpins the ability of atmospheric chemistry to accurately predict environmental change, and that the discipline of multiphase chemistry is strongest when tightly connected to atmospheric modeling and field observations.

1 Introduction

When *Atmospheric Chemistry and Physics* was launched over two decades ago, Abbatt was invited to submit an article to the inaugural issue, which addressed the kinetics of the multiphase reaction between SO_2 and H_2O_2 on ice surfaces (Clegg and Abbatt, 2001):

$SO_2 + H_2O_2 \rightarrow H_2SO_4$ R1

This contribution built upon concepts described in a review article published a few years earlier by Ravishankara, which presented a conceptual view of tropospheric heterogeneous and multiphase chemistry (Ravishankara, 1997). As part of the Special Issue entitled *20 Years of Atmospheric Chemistry and Physics*, both authors value the current opportunity to contribute to the overall goal of the special issue "to reflect on the developments of the field of atmospheric chemistry and physics in the last 20 years and point to exciting directions for the future" by addressing the evolution of the field of atmospheric multiphase chemistry. Specifically, this paper will briefly describe multiphase chemistry, its origins, and the progress made in the past twenty years since the inauguration of *Atmospheric Chemistry and Physics*. It then focuses in depth on future research opportunities and associated challenges. For the sake of brevity, the citations in this paper are illustrative and not comprehensive. And so, the reader is directed to in-depth reviews on specific aspects of multiphase chemistry (Ravishankara, 1997; Jacob, 2000; Rudich, 2003; Usher et al., 2003; Finlayson-Pitts, 2003; Rudich et al., 2007; Kolb et al., 2010; George and Abbatt, 2010; Abbatt et al., 2012; McNeill et al., 2012; Poschl and Shiraiwa, 2015; Herrmann et al., 2015; McNeill, 2015; Burkholder et al., 2017; Pye et al., 2020; Tilgner et al., 2021).

In the 1997 paper, Ravishankara distinguished between heterogeneous and multiphase chemistry based on the extent of diffusion into the bulk. At that time, the term "heterogeneous chemistry" was in vogue to describe ozone hole chemistry. Over the years, it has become clear that diffusion depths vary continuously from solid-like substrates to dilute water solutions. Therefore, in this article, we use the term "multiphase chemistry" to refer to all chemistry that involves more than one phase. Interfacial chemistry falls under this umbrella, with interfaces invariably present when more than one phase is present. We note that "heterogeneous chemistry" is a useful term to describe exclusively interfacial processes (Svehla, 1993), such as for the reactions of gas phase molecules and atoms on solid material such as metallic or mineral catalysts. Similarly, "bulk chemistry" refers to chemistry that occurs mainly in only one phase. In this article, our focus is primarily on processes involving the gas phase interacting with atmospheric condensed phases, so we do not describe in-depth advances in the associated chemistry that takes place in the bulk phase.

One underlying theme in the paper is that understanding multiphase processes at the molecular level improves our ability to accurately predict atmospheric change, which in turn aids in developing sustainable environmental policy and practices. Positive impacts arise across multiple fields, from climate and air quality to human health and ecology. Another theme is that multiphase chemistry studies are most impactful when closely connected to the entire atmospheric science field, noting the interrelated nature of fundamental chemistry, field measurements, and atmospheric modeling that together constitutes the "three-legged stool" model of our field (see Figure 1) (Abbatt et al., 2014). Multiphase chemistry studies should be conducted to guide, interpret, and encourage field observations, and to quantitatively inform atmospheric models.

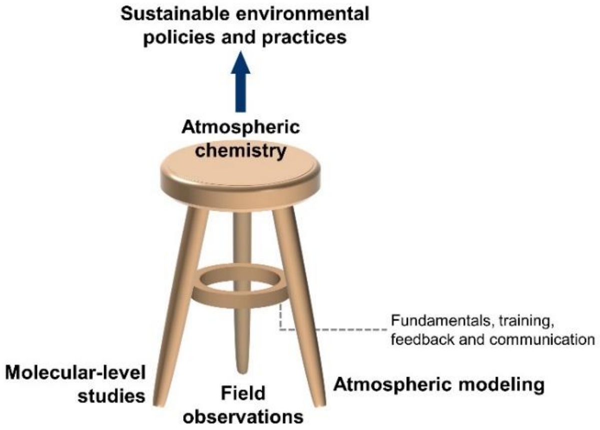

Figure 1: The atmospheric chemistry three-legged stool, where the legs are: i) atmospheric modeling, ii) field observations, and iii) molecular-level studies via experimental and theoretical investigations of gas- and multiphase chemistry. The support ring represents the value arising from collaboration, training, and feedback that occurs across the field, along with the need to focus on fundamental science throughout. (Figure credit: Zilin Zhou)

2 What is atmospheric multiphase chemistry?

Multiphase chemistry involves interactions of chemical species present in two or more atmospheric phases, including gas-solid, gas-liquid, and liquid-solid processes. These interactions typically require both chemical reactions and mass transfer, i.e., the movement of a molecule within a phase or from one phase to another. Also important are processes in the interfacial regions, which are the thin transition zones from one phase to another.

To illustrate a multiphase process, consider R1, a key reaction leading to acid precipitation. Gas-phase reactants must first partition to the condensed phase, such as an aerosol particle, cloud water droplet, or ice crystal. Once molecules collide with the interface, intermolecular forces promote adsorption for a short period, during which they can diffuse and react via an interfacial process (see Figure 2). If diffusion into the bulk is sufficiently fast, they can also react in the bulk. In the case of R1 in liquid water, dissolved SO_2 forms HSO_3^-, which can be oxidized by dissolved H_2O_2 and O_3 to form sulfate (Hoffmann and Edwards, 1975; Penkett et al., 1979). When the substrate is ice instead of liquid water, the reaction proceeds either at the gas-solid ice interface or within a thin, liquid-like layer prevalent on the ice's surface below its melting point (Girardet and Toubin, 2001; Abbatt, 2003).

The interface is involved in all multiphase processes, participating in mass transfer and reactivity. For a low-volatility material, such as a bare mineral or metal, there is a step function drop in the density moving from the condensed phase to the gas phase. A solid's interface, roughly one molecule (or atom) deep, can promote heterogeneous chemistry that occurs in a two-dimensional space (Tabazadeh and Turco, 1993). However, solid particles in the atmosphere, such as soot or mineral dust, frequently have mobile, semi-volatile molecules, such as water, sulfuric acid, and organics, deposited as multilayer-thick films or islands. In these cases, the multiphase chemistry is likely occurring in a three-dimensional space involving chemical interactions with not only the solid substrate but also the liquid coatings (Worsnop et al., 2002; Poschl et al., 2007). For cloud droplets and liquid aerosol particles, which are very dynamic with large fluxes of substrate molecules being rapidly exchanged between the phases, the interfacial region is also thicker than it is for a pure solid (Gopalakrishnan et al., 2005). When the underlying substrate is a liquid, the depth to which a molecule diffuses from the interface into the bulk before reacting is called the reacto-diffusive length, which can be very short for species reacting close to the interface and extending to the size of an aerosol particle for reactions that are aerosol-volume limited (see Figure 2) (Hanson et al., 1994). It is not easy to determine whether a reaction is occurring exclusively in a two-dimensional manner at an interface, where the concept of reacto-diffusive depth breaks down because of different energetics and solvation than in the bulk phase. For particle chemistry, the reaction kinetics dependence on particle size and reactant concentration can provide some information. Nevertheless, even in reactions which exhibit Langmuir-Hinshelwood kinetics (Poschl et al., 2001), the depth of the reactive region at the surface of a particle is not clear for liquids or coated solids.

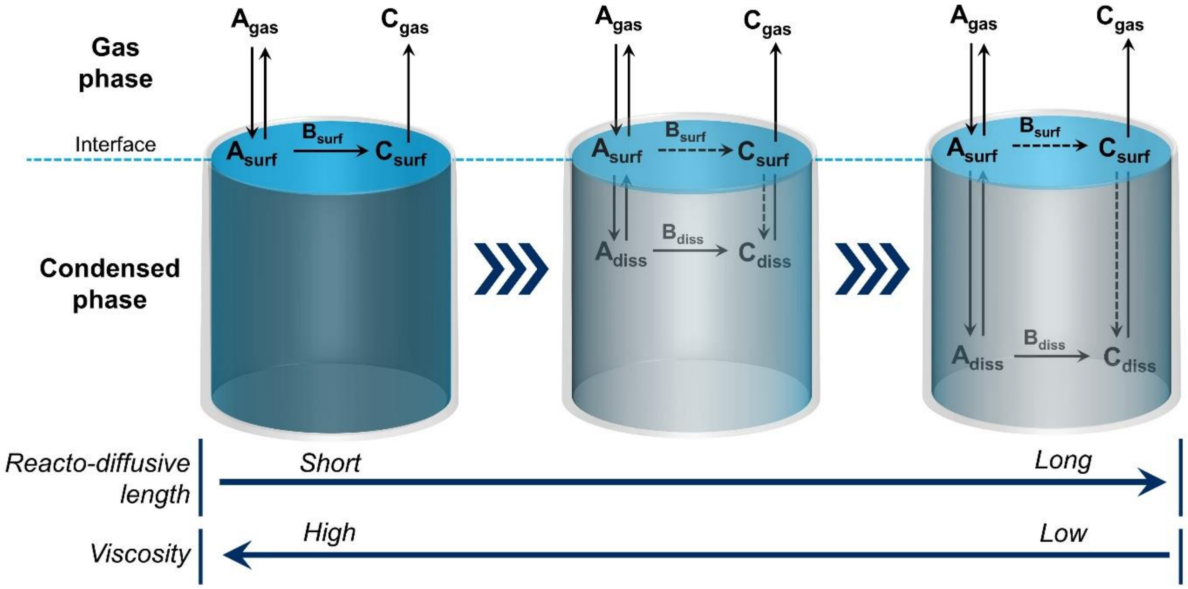

Figure 2: Multiphase reactions occur at the interface and within the bulk of condensed phase materials. Bare solids undergo heterogeneous chemical reactions at the interface. High-viscosity liquids may have sufficiently short reacto-diffusive lengths to restrict reactivity to the surface of the particle. As the viscosity lessens and the reacto-diffusive length deepens, multiphase chemistry can occur deeper into the bulk phase. For reactions that proceed in the bulk, some component of the reaction may also simultaneously occur at the interface (as indicated by the dashed reaction arrow). As noted in the text, the concept of the reacto-diffusive length, which is formally calculated from the bulk diffusion and rate constants, may break down in the interface region. (Figure credit: Zilin Zhou)

3 How does multiphase chemistry differ from gas-phase chemistry?

Although thermodynamically allowed, reactions between neutral closed-shell molecules are slow in the gas phase because of large reaction barriers. However, the rates of multiphase reactions involving the same reactants (or suitably altered versions in the condensed phase) can be larger than in the gas phase, either because the free energy barrier to reaction is lowered or because the concentrations of reactants are enhanced in the condensed phase. To illustrate, SO_2 and H_2O_2 do not react efficiently in the gas phase, but oxidation with dissolved H_2O_2 can proceed once SO_2 dissolves in water and forms HSO_3^-. Moreover, the gas phase H_2O_2 is efficiently scavenged in clouds, enhancing its concentration for reaction.

Another example is the set of reactions that drive polar stratospheric ozone depletion (Solomon, 1999). Chlorine reservoir compounds such as $ClONO_2$ and HCl do not react rapidly in the gas phase. However, HCl partitions strongly to polar stratospheric clouds by either adsorbing to their surfaces or dissolving within them. For example, it is likely that both adsorbed and dissolved HCl dissociates on/in ice to form chloride ions, which are reactive with $ClONO_2$:

$$ClONO_2 + HCl \rightarrow Cl_2 + HNO_3 \qquad R2$$

leading to the formation of Cl_2, which is then released to the gas phase and is readily photolyzed, forming radicals that catalyze gas phase ozone destruction in the Antarctic ozone hole. Also, $ClONO_2$ can be protonated in strongly acidic cloud droplets, or it may dissociate to form Cl^+ and NO_3^- (although there is no direct experimental evidence yet for forming Cl^+ as an independently solvated species). Other possible mechanisms include a concerted reaction between Cl^- and $ClONO_2$ to produce Cl_2 (Bianco and Hynes, 1999). It is unclear if such a reaction is truly an SN2-type process or if it can occur in a cage in the condensed phase.

Another distinguishing feature of multiphase chemistry is that it can lead to the formation of products that do not arise in the gas phase. Consider the acid-catalyzed nucleophilic reactions between sulfate and isoprene-derived epoxydiols that form organo-sulfate molecules and secondary organic aerosol (SOA) (Riva et al., 2019). Water molecules lower the transition state energies of such reactions. The solvent shell, which confines reactant partners via the so-called cage effect, can also promote novel products. For example, the formation of biologically-active secondary ozonides is facile in the condensed-phase ozonolysis of unsaturated fatty esters and triglycerides, arising from reactions of Criegee and carbonyl intermediates that form in the same solvent shell after dissociation of a primary ozonide (Zhou et al., 2019b, 2022). In the gas phase, the solvent shell is essentially absent (except for some specific cases, such as cluster formation with H_2O), so the carbonyl and Criegee intermediates fly apart, and secondary ozonides do not form so readily.

Lastly, some multiphase reactants, such as transition metal ions, are absent in the gas phase. More generally, ion-ion and ion-molecule regions play a much greater role in condensed phase chemistry than in tropospheric and stratospheric gas phase chemistry, leading to a wide variety of novel reaction pathways, with R1 being an excellent example.

4 Early studies of atmospheric multiphase chemistry

Studies of aerosol and cloud chemistry have proceeded in concert with the development of the wider atmospheric chemistry field with many of the concepts of coupled reactivity and mass transfer initially developed by the process-oriented chemical engineering community (Dankwerts, 1970). Interest in multiphase reactions arose via the profound ways these sparse aerosol particles and cloud droplets can alter gas phase composition. Also, as described in more detail below, it is now evident that the reverse is important, i.e., the gas phase alters the condensed media with important environmental consequences. Many aerosol and multiphase reaction studies were initially performed to develop parameterizations for atmospheric modeling. Even though this is still a major goal, much more effort is now given to understand the physico-chemical processes, which is essential for predictive capabilities.

The idea of reactive chemistry in hydrometeors goes back to the late 1960s and 1970s and possibly earlier. As in much of science, it is hard to pinpoint a specific paper that expounded this idea. The expectation that the SO_2 pollutant can be oxidized to sulfuric acid in the water droplets in the atmosphere spurred many studies, hypotheses, and definitive results. Early studies examined the potential oxidation of SO_2 in the liquid phase via a variety of oxidants, including O_3, H_2O_2, and NO_2 (Hoffmann and Edwards, 1975; Erickson et al., 1977; Schroeder and Urone, 1978; Penkett et al., 1979). As well, modeling studies showed the feasibility of such oxidation reactions occurring in the atmosphere (Jacob and Hoffmann, 1983; and references therein). The Great Dun Fell experiment observationally established that SO_2 is indeed oxidized in the troposphere via reactions in cloud droplets (Choularton et al., 1997).

Similar multiphase reactions in the stratosphere were sometimes evoked but never pursued with great vigor until the spectacular occurrence and subsequent explanation of the ozone hole (Solomon, 1999) and a multitude of laboratory studies showing that indeed there can be chlorine activation (Molina et al., 1987; Tolbert et al., 1988; Leu, 1988; Hanson and Ravishankara, 1992; and many others).

Additional pioneering atmospheric multiphase chemistry studies arose from aerosol composition measurements conducted over a half-century ago. Specifically, continental aerosol particles always contain a measurable quantity of ammonium, indicating the uptake of gas-phase ammonia to acidic particles (Lee and Patterson, 1969; Kadowaki, 1976). Furthermore, particulate chloride levels in the marine aerosol are depleted relative to their seawater abundance, replaced by sulfate or nitrate (Junge, 1956; Martens et al., 1973). This process was long thought to be the major source of gas-phase chlorine, whereby gaseous HCl is displaced from NaCl particles via the uptake of gas-phase strong acids :

$H_2SO_4 + 2\ NaCl \rightarrow 2\ HCl + Na_2SO_4$ R3

Another early example of halogen chemistry demonstrated that volatile iodine species are released when dissolved oceanic iodide is exposed to either ultraviolet light or ozone (Miyake and Tsunogai, 1963; Garland et al., 1980). This multiphase chemistry is important for the dry deposition of ozone and the release of iodine into the atmosphere (Carpenter et al., 2013):

$O_3 + H^+ + I^- \rightarrow HOI + O_2$ R4

$HOI + I^- + H^+ \rightarrow I_2 + H_2O$ R5

After these early studies, many additional important tropospheric multiphase chemical processes were identified prior to the launch of *Atmospheric Chemistry and Physics.* Some examples, which all address gas-particle and cloud droplet interactions, include: i) the formation of reactive halogen species in the boundary layer (Finlayson-Pitts, 2003; Simpson et al., 2015), ii) the uptake of tropospheric gases by mineral dust, especially nitric acid (Hanisch and Crowley, 2001; Usher et al., 2003), iii) the scavenging of trace gases, such as nitric acid and small oxygenated VOCs, by snow and ice crystals in the free and upper troposphere (Abbatt, 2003), iv) the impact of aqueous cloud and aerosol chemistry on gas phase HO_x levels (Chameides and Davis, 1982; Calvert et al., 1985; Jacob, 1986; Lelieveld and Crutzen, 1991), v) conversion of N_2O_5 to HNO_3 on tropospheric aerosol, with impacts on the NO_x budget (Dentener and Crutzen, 1993), vi) uptake of HO_2 to aerosol (Mozurkewich et al., 1987; Martin et al., 2003), and vii) multiphase conversion of NO_2 to HONO (Finlayson-Pitts et al., 2003). A critical point is that each of these multiphase processes affects the oxidizing capacity of the troposphere, frequently through modification of radical budgets and occurring via gas-aerosol or gas-droplet interactions. For example, these processes initiate oxidation in urban atmospheres through HONO photolysis, drive Arctic boundary layer ozone and mercury depletion via gas-phase halogen chemistry, and modulate the global oxidizing capacity via N_2O_5 or HO_2 loss on aerosol particles. Additional work in the multiphase world at this time involved a wide variety of condensed-phase photochemistry studies, for example involving the interactions of light with nitrate (Zepp et al., 1987), which can lead to the release of NO_x to the gas phase, and with transition metal ion complexes (Faust and Zepp, 1993).

5 Progress in the past twenty years

Two major developments profoundly influenced multiphase chemistry. First was the recognition of the importance of aerosol particles in changing the radiative balance of the Earth system, with impacts on climate. The second was the continued recognition of the deleterious effects of particulate matter on human, animal, and ecosystem health. These two fields, climate change and air quality, have provided the impetus (and resources) for the development of the field. As a result, additional research emphasis in the multiphase chemistry community was given at the turn of the 21st century to assess the impacts that arise on the composition of the particles.

Once inhaled, particles harm human health (Landrigan et al., 2018; Murray et al., 2020), with recent studies implicating the secondary component of the particles in negative health outcomes (Pye et al., 2021). Research in the past two decades has focused strongly on the formation of SOA (Kroll and Seinfeld, 2008; Hallquist et al., 2009;

Ziemann and Atkinson, 2012; Shrivastava et al., 2017b). SOA formation has required better knowledge of the kinetics and mechanisms of gas phase oxidation of SOA precursors (Crounse et al., 2013; Ehn et al., 2014). It has also needed a more complete understanding of gas-particle nucleation processes (Kulmala et al., 2014; Trostl et al., 2016; Xiao et al., 2021), volatility (Pankow, 1994; Donahue et al., 2011), and condensed-phase reactions that occur within aerosol particles. Specifically, volatility and multiphase reactivity can be coupled, as illustrated by the formation of high molecular weight, low volatility species within particles (Kalberer et al., 2004). While such oligomers and highly oxygenated species may also form in the gas phase (Bianchi et al., 2019), they arise via a variety of reactions involving pairs of organic reactants, frequently forming esters and acetals/hemiacetals in the condensed phase (Tobias and Ziemann, 2000; Surratt et al., 2006; DeVault and Ziemann, 2021). These reactions may be acid-catalyzed (Jang et al., 2002). Also, multiphase oxidation by gas-phase oxidants can increase the average oxidation state of organic aerosol particles (Kroll et al., 2011) via a series of reactions that initially functionalize and eventually fragment the component molecules (Moise and Rudich, 2000; Molina et al., 2004; George et al., 2007; Kroll et al., 2009). Oxidation leads to a more soluble particle that increases its rate of wet deposition. In addition to forming organic aerosol via gas-to-particle conversion, they are produced from the evaporation of cloud droplets. Oxidation processes occur within cloud droplets (Herrmann et al., 2015), producing more oxidized organics that yield oxygenated aerosol upon evaporation. Similar reactions, proceeding at much higher organic reactant concentrations, can also occur within the aqueous component of tropospheric aerosol (Blando and Turpin, 2000).

Tightly connected to SOA formation and modification processes are the condensed phase viscosity and phase state, which set mixing times within particles and are dependent on relative humidity and temperature (Virtanen et al., 2010; Koop et al., 2011; Renbaum-Wolff et al., 2013). Organic particles are likely glasses in the cold free troposphere (Shiraiwa et al., 2017b), which may affect SOA formation and growth, and restricts the degree to which heterogeneous oxidation can affect the aerosol composition. The particles are liquids in warm, wet boundary layers, with the full particle volume involved in partitioning with gas phase molecules. The large variation in molecular diffusion coefficients and associated mobility determines where chemical reactions are important in the particles, from two-dimensional processes that occur solely at the gas-particle interface to three-dimensional chemistry with reactivity at the interface and deeper in the bulk (see Figure 2). Overall, diffusion is a key parameter for determining whether a reaction is surface-area-limited or volume-limited (Hanson et al., 1994).

Multiphase chemistry also leads to the formation of secondary inorganic aerosol. For example, the hydrolysis of N_2O_5 converts NO_x to HNO_3; the gas-particle partitioning of HNO_3 is then controlled by temperature, relative humidity, and ammonia levels. Also, particulate sulfate is rapidly formed in polluted environments through multiphase aqueous chemistry, acting as the major formation mechanism in cloud-free settings (Cheng et al., 2016; Wang et al., 2016). Potential routes for fast sulfate formation in deliquesced particles include: the role of ionic strength in accelerating the rates of specific processes (Liu et al., 2020), fast interfacial chemistry (Liu and Abbatt, 2021), formation of condensed-phase oxidants through the photolysis of particulate nitrate (Zheng et al., 2020; see also Section 6.5), and the role of specific particle-phase reactants, such as organic hydroperoxides (Wang et al., 2019), hydroxymethanesulfonate (Song et al., 2019), and dissolved transition metal ions (Li et al., 2020b). An accurate quantitative assessment of these and other reaction pathways is still developing but far from complete (Liu et al., 2021b).

As noted earlier, the need to better understand aerosol-climate interactions has also motivated multiphase chemistry research in the past twenty years. Atmospheric processing leads to the formation of water-soluble condensed-phase products, such as sulfate or highly oxygenated organic molecules (Jimenez et al., 2009), enhancing the abilities of tropospheric aerosol particles to act as cloud condensation nuclei (CCN) and affecting their ability to scatter light (Cappa et al., 2011; Moise et al., 2015). As well, the optical properties of the fraction of organic aerosol that absorbs near ultraviolet and visible light (i.e., atmospheric 'brown carbon' particles) are subject to change via multiphase oxidation and condensed phase photochemistry (Laskin et al., 2015; Li et al., 2020a; Hems et al., 2021; Schnitzler et al., 2022), potentially involving transition metals (Al-Abadleh and Nizkorodov, 2021). Although the rates of optical property changes remain uncertain, primary brown carbon particles, as formed in wildfires, tend to become less absorbing in the near UV and visible parts of the spectrum on the timescale of days via a variety of multiphase aging mechanisms (Laskin et al., 2015; Hems et al., 2021), i.e., they are 'bleached.' The diminution of light absorption is in accord with field observations (Forrister et al., 2015).

Multiphase chemistry can also affect the properties of ice nucleating particles (INPs) by both gas-solid and liquid-solid interactions, noting that INPs can act in the deposition mode where water vapor forms ice directly on solid substrates and in the immersion mode where a solid particle immersed in supercooled water leads to nucleation (Kanji et al., 2017). For example, mineral dust can have its IN activity decreased by condensation of involatile materials, such as sulfate or by cloud processing (Sullivan et al., 2010b; Kilchhofer et al., 2021), and strong acids can react with carbonate-containing minerals, leading to particles that are less IN-active in the deposition mode but more active in the immersion mode (Sullivan et al., 2010a). Such effects can also arise when different gas and liquid species are exposed to volcanic ashes (Maters et al., 2020; Fahy et al., 2022). Oxidation reactions can also occur so that efficient biological INPs, such as pollen fragments, lose activity upon oxidation by OH radicals, probably by morphological changes of surface proteins and carbohydrates (Gute and Abbatt, 2018). The mechanisms involving all these interactions are very complex. In the case of mineral dusts, immersion INP activity can be changed by surface modification, ion exchange, adsorption of solutes such as ammonium, and acid dissolution (Sihvonen et al., 2014; Kumar et al., 2019; Yun et al., 2021).

6 The future of atmospheric multiphase chemistry studies

6.1 Multiphase chemistry at the interfaces of the atmosphere

There are exciting opportunities for applying the conceptual, instrumental, and modeling tools developed for multiphase chemistry to understand chemistry occurring at the interfaces of the atmosphere with other environmental domains.

Consider the interface of the atmosphere and the ocean, where the sea-surface microlayer (SML) is a thin layer of ocean water that has enhanced concentrations of biological detritus, surface-active compounds, and gel-like substances (Cunliffe et al., 2013). Recognizing that individual molecule surrogates of the SML only capture specific aspects of the chemistry, experimental designs now involve either genuine seawater or water samples with significant biological components (Prather et al., 2013; Schneider et al., 2019). While we know that the SML can affect the composition of primary marine aerosol, an open question is the degree of chemical processing by photosensitization in the SML or by gas-surface heterogeneous oxidation, yielding volatile species that contribute to marine SOA formation (Donaldson and George, 2012; Rossignol et al., 2016; Mungall et al., 2017; Croft et al., 2019).

Another key role of multiphase reactions is in dry deposition processes on the ocean (e.g., see R4 and R5), vegetation, the built environment, and land surfaces (Garland et al., 1980; Fowler et al., 2009; Kavassalis and Murphy, 2017; Tuite et al., 2021). Deposition is a critical step that controls removing chemicals from the atmosphere. Yet, this process is a parameterization in models. Deposition in many environments needs to be predictive, which demands molecular-level understanding and quantification. This process is essentially a multiphase process that should be broken down into physico-chemical steps, which can be independently measured and understood.

Indoor environments, with their vast surface area-to-volume ratios, are another example of poorly explored multiphase processes (Morrison, 2008; Abbatt and Wang, 2020; Ault et al., 2020). Contrary to the outdoor environment, where aerosol particles may remain suspended for days to weeks, the indoor air-exchange timescale is on the order of an hour or two. While this lessens the potential for gas-particle chemistry, multiphase chemistry occurs over much longer timescales on fixed indoor surfaces. For example, O_3 is efficiently lost via dry deposition so that its mixing ratios are considerably lower indoors than outdoors (Weschler, 2000). This produces VOCs (Wisthaler and Weschler, 2009) and modifies the composition of sorbed molecules, in some cases forming species more toxic than their precursors (Pitts et al., 1978, 1980; Zhou et al., 2017). It can also lead to the formation of gas-phase OH radicals (Zannoni et al., 2022). This source of OH from alkene ozonolysis is in addition to the generation of OH from photolysis of HONO (Gomez Alvarez et al., 2013), which is partly formed by multiphase chemistry on indoor surfaces. Indoor surfaces are a chemically complex, poorly understood environment, with input from building materials, commercial products, humans, and cooking and cleaning activities. This chemistry is important because humans obtain most of their chemical exposure indoors, not only via inhalation but also through direct dermal uptake and by ingesting dust and contaminated foodstuffs (Li et al., 2019b). Lastly, the light environment indoors can be substantially different than outside, bringing a new twist to multiphase photochemistry (Young et al., 2019).

6.2 Multiphase chemistry and human health

Epidemiological studies have conclusively shown that aerosol particle inhalation harms human health (Pope et al., 2009; Landrigan et al., 2018). For example, it is well-recognized that inflammation occurs upon particle exposure (Brook et al., 2003). The current epidemiology (empirical evidence) does not readily distinguish the specific molecules in the particles and their formation pathways that lead to negative health outcomes, nor the toxicity mechanism at the molecular level. Studies are currently examining oxidative stress, e.g., the reactive oxygen species (ROS) and reactive nitrogen species (RNS), as a mechanism for negative impacts (Shiraiwa et al., 2017a). Although there is debate over whether oxidants are largely endogenous or exogenous (Fang et al., 2022), one hypothesis is that the biochemical balance between oxidants and antioxidants is upset by inhaling harmful species (Miller, 2020). To contribute to this debate, the multiphase chemistry community needs to better describe the chemistry that occurs at the lung-air interface and the composition of respirable aerosol particles, especially the biologically active components that contain reactive functional groups (e.g., epoxides, hydroperoxides), redox-active materials (e.g., quinones), and reactive oxygen species (e.g., peroxides, HO_2/O_2^-). Many of these species are formed by multiphase oxidation processes.

An associated issue is how ultrafine particles influence health. These particles have been shown to be taken directly to the bloodstream and even move to the brain (Oberdorster et al., 2004; Maher et al., 2016). Though the chemistry involved is not the multiphase chemistry discussed here, the interactions of the particle in the liquid phase (i.e., impacting biological systems) are likely important. Many of the lessons learned from studies of multiphase processes are likely applicable to understanding such issues.

Largely unexplored until the recent COVID-19 pandemic is the impact of the atmosphere on airborne and surface-deposited biological pathogens, including bacteria and viruses. Early work in this area included the multiphase chemistry between NO_2 and proteinaceous material, motivated by its potential to drive an allergenic response (Franze et al., 2005; Shiraiwa et al., 2012). Gas phase O_3 has also been examined for its ability to affect the viability of bacteriophages, i.e., microorganisms with a lipid envelope and RNA core similar to the structure of SARS-CoV-2, deposited on surfaces (Tseng and Li, 2008). With the pandemic, research has accelerated into the impact of hygroscopic growth and water content on viral viability within respiratory particle surrogates that consist of viruses embedded in saline droplets containing surfactants, proteins, and carbohydrates. It is important to understand the changes in the acidity of these particles, the mass transfer within them, and the precipitation of salts as the particle water content changes (Lin et al., 2020; Oswin et al., 2022; Huynh et al., 2022).

The recent pandemic led to an emphasis on cleaning surfaces to reduce the potential for infection by fomites, i.e., via contact with contaminated surfaces. While cleaning agents such as chlorine bleach have well-established anti-microbial activity, their multiphase chemistry can release gases and particles that are deleterious to human health (Wong et al., 2017; Mattila et al., 2020). Understanding the multiphase chemistry associated with these cleaning activities and the outcomes of using air cleaners (Collins and Farmer, 2021), is essential for establishing healthy indoor environments.

Lastly, the pandemic prompted a re-examination of an overlooked aspect of our atmosphere that it has an as-yet-unidentified germicidal component referred to as the Open Air Factor (Cox et al., 2021). In particular, it was shown many decades ago that fresh air led to better outcomes for tuberculosis patients and injured World War I soldiers than indoor air. Historically, sending sick people to pristine environments (e.g., the seaside) was a common medical recommendation. It is crucial to determine whether these effects are related in some way to multiphase chemistry occurring between reactive species in the gas phase interacting with biological molecules at the surface of the wounds and lungs.

Each of the above topics provides exciting opportunities for atmospheric chemists to collaborate with the environmental health, medical, and toxicological communities.

6.3 Chemistry of the free troposphere and lower stratosphere

Although the upper troposphere – lower stratosphere region was the focus of much attention in the 1980s and 1990s to understand the changes in ozone levels in these regions, most multiphase chemistry studies are currently conducted at room temperature. There is considerable motivation to re-explore chemistry at colder temperatures, given past work that illustrated the atmospheric impacts of a strongly non-linear dependence of multiphase reactions rates on temperature (Murphy and Ravishankara, 1994) and extensive new observations from the Atom campaigns (Thompson, 2022) that sampled from the boundary layer to the upper troposphere over many latitudes and seasons. As well, there is emerging evidence for organic aerosol in the lower stratosphere, likely arising from wildfire injection, with potential influence on stratospheric ozone (Solomon et al., 2022; Strahan et al., 2022).

Organic aerosol viscosity and phase state depend on the environmental conditions (Koop et al., 2011), with semi-solid and glassy organic particles predicted throughout much of the free troposphere (Shiraiwa et al., 2017b). Aside from those at the gas-particle interface, molecules in highly viscous organic particles are protected from heterogeneous oxidation (Shiraiwa et al., 2011; Zhou et al., 2012; Shrivastava et al., 2017a). Such protection increases the lifetimes of pollutants, e.g., brown carbon chromophores (Schnitzler et al., 2022), and lengthens particles' oxidation timescale and wet deposition lifetime.

In addition to continuing to address the fundamentals of cloud chemistry oxidation processes, the associated chemistry of transition metals, and the production of oxidants within cloud water and via uptake from the gas phase (Herrmann et al., 2015), there is a particular need to also study such processes at cold temperatures, including under supercooled water conditions. When supercooled water is frozen, solutes are excluded from the ice crystals and become highly concentrated at grain boundaries and in liquid and liquid-like layers at the surface, potentially leading to enhanced rates of aqueous phase chemistry. As well, the Reynolds-Workman potential (Workman and Reynolds, 1950), arising at the ice-water interface, can drive chemistry.

A key factor affected by temperature is the solubility of various atmospheric constituents. Simple Henry's law constants and further equilibration steps that determine the overall solubilities are poorly known, especially below room temperature. Most of the data on the solubilities in organics goes back to chemical engineering literature that is more than half a century old. Also, since solubilities vary according to Henry's law equilibria that vary exponentially with temperature, the accurate temperature dependence of solubilities is essential. Acid dissociation constants in organic acids and organic substrates are poorly known, and they determine the overall solubility of a chemical.

6.4 Reactive transformations of organic chemical contaminants

Over forty years ago, it was recognized that multiphase oxidation of chemical contaminants leads to the rapid loss of surface-bound PAHs and the formation of more toxic and potentially carcinogenic products such as nitro-PAHs and oxygenated PAHs (Pitts et al., 1978, 1980). These reactions occur on a variety of surfaces with light, ozone, and NO_2 reactants, some via Langmuir-Hinshelwood mechanisms (Poschl et al., 2001; Mmereki and Donaldson, 2003; Kwamena et al., 2004). Buried PAHs are protected from heterogeneous loss by a crust of unreactive products that accumulates upon them and, when present, within viscous organic aerosol (Zhou et al., 2013, 2019a), enabling the potential for long-range atmospheric transport (Mu et al., 2018). The chemistry of other organic contaminants, including smoking products such as nicotine (Destaillats et al., 2006) and tetrahydrocannabinol (Yeh et al., 2022), a few pesticides (Segal-Rosenheimer and Dubowski, 2007; Finlayson-Pitts et al., 2022), and organophosphate esters (Liu et al., 2021a), has also been recently explored.

However, these are largely exceptions, and the multiphase fate of most chemical contaminants, especially thousands of commercial products, has not been examined. Indeed, the atmospheric chemistry and chemical contaminant communities have traditionally not strongly interacted. Although assessment of the gas phase OH reactivity is customarily performed in environmental fate analyses (Li et al., 2019b), many commercial products have sufficiently low volatility such that they reside primarily on surfaces or within particles. It is important to establish whether organic contaminants traditionally viewed as persistent are indeed unreactive with respect to multiphase transformation.

6.5 Understanding the role of light

Many condensed-phase photochemical reactions proceed via indirect mechanisms where a photosensitizing molecule absorbs light, forming reactive species such as HO_2/O_2^- or $^1O_2$ (George et al., 2015). Such chemistry, first identified for natural waters (Canonica et al., 1995), has been implicated in the daytime formation of HONO (George et al., 2005), the photoreactions of brown carbon aerosol (Laskin et al., 2015; Hems et al., 2021), the formation of active halogens (Reeser et al., 2009), and reactivity of black carbon (Monge et al., 2010; Li et al., 2019c). This chemistry has been illustrated using efficient photosensitizing agents, but quantitative assessments of atmospheric importance remain uncertain largely because the character and quantity of atmospheric photosensitizers are not well established. Developing a tighter quantitative connection to the atmosphere will require using more representative photosensitizers, as now being done using marine aerosol components (Ciuraru et al., 2015; Garcia et al., 2021). The wavelengths of interest for the troposphere are in the near UV and visible part of the solar flux.

In addition to indirect sensitization, light can also lead to direct photochemistry. An important finding was that photolysis on ice and snow surfaces was demonstrated to form NO_x in midlatitudes and polar regions (Honrath et al., 1999; Wolff et al., 2002; Domine and Shepson, 2002). This process, which likely proceeds in a wide range of environments, is now referred to as “re-noxification” as it releases NO_x from HNO_3 that has deposited from the atmosphere. Other condensed-phase chemical processes of importance include the formation of oxidants from nitrate and nitrite photolysis (Zepp et al., 1987), photolysis of condensed-phase organic hydroperoxides and other highly oxygenated organics, and the photochemical activity of many transition metal ion complexes (Faust and Zepp, 1993; Weller et al., 2013). It is important to recognize that the absorption spectra and product quantum yields of dissolved species can be different than those in the gas phase (George et al., 2015), with aqueous nitrate a prime example (Benedict et al., 2017). The variable viscosity of organic aerosol matrices can affect photolysis rates, products, and their temperature dependence (Lignell et al., 2014).

6.6 Developments in Field Observational Capabilities

Our ability to characterize atmospheric composition continues to push the field of atmospheric chemistry forward. For multiphase chemistry, advances in analytical mass spectrometry have been transformative. Within the last twenty years, online characterization of aerosol composition has become commonplace (Canagaratna et al., 2007), studies of single particle composition allow us to observe the variability in mixing state and chemical diversity (Zelenyuk and Imre, 2005; Murphy et al., 2006; Prather et al., 2008), and offline filter sampling has progressed from the characterization of a few targeted species to non-targeted analyses using a range of mass spectral ionization methods (Papazian et al., 2022; Ditto et al., 2022). Identifying specific molecular "markers" for organics and functional groups is still somewhat uncertain; developing such identification would be very helpful.

The continued development of analytical techniques will enable increasingly sophisticated characterization of aerosol particles and environmental surfaces, with the opportunity to deploy the same tools in both lab and field settings. However, challenges are arising as well. Despite the rapid development of low-cost sensors, affordable instrumentation for the long-term characterization of aerosol composition in many locations is still lacking. The increasing sophistication of analytical instrumentation also continues to unveil the high degree of chemical complexity present. Whereas high-resolution mass spectrometry yields chemical formulae in real-time, there is often the need to identify chemical structures. This suggests that we should increasingly deploy separation techniques (e.g., chromatography, ion mobility) as front ends to our increasingly sophisticated mass spectrometric techniques (Krechmer et al., 2016; Claflin et al., 2021). There is also value to the expanded use of other classical chemical speciation methods, such as infrared (Russell, 2003) and NMR (Decesari et al., 2007) characterization of aerosol composition collected by filters. While these techniques have low time resolution, they provide complementary quantitative and functional group information and can be inexpensively deployed for long-term analyses in a wide range of environments. Such analyses will also help with the source apportionment of the aerosols.

Aerosol characteristics related to multiphase chemistry can be studied with increasingly sophisticated remote sensing techniques. These approaches have been applied for many years to polar stratospheric clouds, whose composition and phase (via the degree of depolarization of a lidar probe) have been studied (Tritscher et al., 2021). Another example comes from satellite measurements of solid ammonium nitrate particles in the upper troposphere, driven by the Asian monsoon that uplifts ammonia-rich continental air (Hopfner et al., 2019). It is important to determine the role of these particles in ice nucleation and multiphase chemistry.

6.7 Developments in Laboratory and Molecular Modeling Techniques

In addition to our ability to conduct field observations, a revolution has occurred in the laboratory's analytical methods. This is most widely apparent in applying sophisticated mass spectrometric techniques, increasingly involving high mass resolution and a range of ionization schemes (Laskin et al., 2013). When coupled with other analytical methods, we can now determine the physico-chemical properties of individual molecules and their mixtures in extreme detail. For example, this approach has been taken to characterize the optical properties of brown carbon aerosol materials (Fleming et al., 2020), the viscosity of organic aerosol (DeRieux et al., 2018), and the structural isomers of complex organic mixtures (Krechmer et al., 2016). There are significant opportunities for additional adoption of techniques from neighboring disciplines. As well, the use of a number of these analytical techniques in both the laboratory and the field will enhance our ability to connect the lab to the field.

Molecular-level chemical models increasingly provide valuable insights into complex multiphase processes. For example, important insights into the nature of the chemistry occurring on polar stratospheric cloud materials were obtained from molecular dynamics modeling (Wang and Clary, 1996; Bianco and Hynes, 2006) and more recent studies have addressed gas-surface interactions and the roles of solvent molecules in small molecular clusters (Gerber et al., 2015; Fang et al., 2019; Yang et al., 2019). Whereas past computational methods only included a few solvent molecules, current dynamics models using state-of-the-art force fields can realistically simulate partitioning, surface adsorption constants, diffusion constants, and vapor pressures, representing an important point of contact to the physical chemistry and chemical physics communities (Tobias et al., 2013). We also note that machine-learning techniques are very recently being applied to molecular dynamics simulations, for example, to describe the interactions of N_2O_5 with liquid water (Galib and Limmer, 2021) and the dissociation of strong acids at aqueous interfaces (de la Puente et al., 2022). For establishing fundamental parameters that are experimentally challenging to measure, such as the likelihood that a collision of a molecule with a particle leads to uptake by the condensed phase (i.e., a mass accommodation coefficient), theoretical methods may be preferable to experiment in some situations.

6.8 Grappling with chemical complexity

Atmospheric aerosol particles and surfaces are morphologically and compositionally complex. This complexity can be enticing from a fundamental chemistry perspective as we disentangle mass transfer, phase separation, and reactivity. However, it can impede the development of an accurate, quantitative description required to inform an atmospheric model. It can also be constraining if we study the detailed chemistry and lose sight of its overall impact on climate, air quality, or ecosystem health.

With enough care, the rate constant for a gas-phase, radical-molecule reaction can be measured with 10-20% accuracy (Cox, 2012). Atmospheric modelers rely upon this confidence level as they assess their predictions. It is humbling to consider the accuracy of the available multiphase kinetics data for the modeling community. Take for example the reaction of N_2O_5 with tropospheric aerosol, which has been long known to impact NO_x/NO_y and active chlorine levels, with a secondary influence on OH, O_3, and CH_4 (Dentener and Crutzen, 1993). Although studies started in the 1980s, new mechanistic insights on N_2O_5 heterogeneous reactivity are still arising (Sobyra et al., 2019; Karimova et al., 2020). Laboratory reactive uptake coefficients for the hydrolysis of N_2O_5 vary over one-to-two orders of magnitude, with larger values reported for aqueous particles composed of sulfate or soluble organics, and lower values for particles with less soluble organics and nitrate (Burkholder et al., 2020). Likewise, uptake coefficients inferred from field measurements or with genuine ambient particles vary by roughly an order of magnitude compared to those measured with laboratory surrogates (Brown et al., 2006; Bertram et al., 2009; Abbatt et al., 2012; Phillips et al., 2016; Tham et al., 2018). The discrepancies between field and lab studies are undoubtedly due to complex and variable particle composition and phase state. Simply put, unlike the case with gas phase reactions, one of the "reactants" in this gas-particle reaction is highly variable. This complexity is exacerbated by the changes in the composition (including acidity), mixing state, and water content of the particle as it resides in the atmosphere. Added complexity arises from the differences in composition that occur in the bulk of particles and droplets compared to their interfacial composition (Wingen and Finlayson-Pitts, 2019).

Likewise, HO_x loss on tropospheric aerosol may significantly impact ozone in high-NO_x atmospheric regimes, as in East Asia. As particulate levels drop in such regions, HO_x abundance and ozone mixing ratios will both rise (Martin et al., 2003; Li et al., 2019a; Ivatt et al., 2022). However, reported HO_2 uptake coefficients vary widely, from research group to research group and from the lab to the field (Burkholder et al., 2020), making modeling predictions highly uncertain.

Both bottom-up and top-down approaches can address chemical complexity. In the traditional bottom-up approach, the effects on the reaction system of step-by-step additions of chemical complexity are evaluated. This leads to a better understanding of the fundamental chemistry needed to develop our predictive abilities. Top-down approaches involve studying chemistry on ambient aerosol particles. This has been done for N_2O_5 and HO_2 aerosol uptake (Bertram et al., 2009; Zhou et al., 2021), for heterogeneous OH oxidation (George et al., 2008), and to characterize SOA formation by using mobile reaction chambers (Jorga et al., 2021). Another top-down method constrains the rates of multiphase chemistry using detailed, simultaneous measurements of gas-phase composition under a range of environmental conditions (Brown et al., 2006). Combining top-down and bottom-up approaches enhances our understanding of the fundamental science while ensuring that parameterizations for atmospheric modeling are accurate.

Models working over a wide range of spatial and temporal scales can help address issues in chemical complexity. As mentioned in the previous section, molecular dynamics calculations are becoming increasingly sophisticated. So too are multiphase kinetics models that can incorporate insights gained at the molecular level into modelling frameworks that aim to couple the gas phase and condensed phases, including bulk reaction kinetics, mass transfer and interfacial processes (Poschl et al., 2007; Tilgner et al., 2013; Woo and McNeill, 2015). A challenge is to couple bulk and interfacial processes correctly. As computing capabilities grow, the complexity of the multiphase and detailed molecular mechanism models that can be incorporated into chemical transport models will also increase. Also, Lagrangian-type models increasingly can model specific field observations (Zaveri et al., 2010). We note that a successful hierarchical approach has arisen in the indoor chemistry community where modeling groups using a wide range of tools, from molecular dynamics to large-scale computational fluid dynamics, interact closely with each other and with experimental scientists (Shiraiwa et al., 2019).

7 Concluding thoughts

- Multiphase chemistry has evolved alongside the wider field of atmospheric chemistry. While initial studies focused on its impacts on the gas phase, the field now addresses how chemistry affects particles. Although modification of aerosol composition has direct relevance to climate and human health, we should not lose sight of the connection of multiphase chemistry to the gas phase composition of the atmosphere.
- We need to understand chemical processes at the molecular level to improve our ability to interpret field observations and predict the nature of a changing atmosphere. Reinforcing an approach based on physico-chemical understanding is necessary for detailed predictions of environmental change.
- There are significant research opportunities for the characterization of the chemistry that occurs at the interface of the atmosphere with the rest of the environment, such as studies of ocean-atmosphere interactions, indoor air, aerosol health effects, atmosphere-cryosphere chemistry from the stratosphere all the way to the snowpack, and pathogen-air interactions.
- With increasingly sophisticated experimental and theoretical tools, atmospheric chemical complexity becomes more apparent. While exciting, this presents challenges and constraints. We should emphasize not only highly detailed, molecular-level measurements but also more widespread and more prolonged aerosol characterization that has less chemical specificity but nevertheless provides valuable insights; there is also a role for both remote sensing measurements and classical analytical techniques in this regard. This is akin to a need to understand and quantify thermal gas phase reactions while also understanding and quantifying microcanonical reactivity.
- Measurements of many fundamental physico-chemical parameters such as solubility, diffusion coefficients, and liquid/solid phase reactivities are sorely needed.
- Multiphase chemistry studies conducted under conditions that match those in the atmosphere, including those of the free troposphere and lower stratosphere, are needed.
- Using the atmosphere as a laboratory to quantify rates of multiphase processes holds promise, with simultaneous measurements of many chemicals and other external parameters becoming more feasible through coordinated field measurements. Designing field studies with an eye toward quantification of the multiphase reactions is beneficial.
- The field of atmospheric chemistry is healthiest when there is extensive communication and feedback between the fundamental chemistry, modeling, and field observation communities (see Figure 1). To keep the three-legged stool balanced and strong, multiphase chemists should interact widely with not only other atmospheric scientists, but also scientists in related fields such as meteorology, climate dynamics, ecology, and human health. This can be accomplished by participation in conferences and seminars that involve science from different legs of the stool, students exploring short-term training opportunities in diverse research groups, and collaborative grants that bring together laboratory and theory, modeling, and field measurement scientists.

Author Contributions

Both authors contributed to writing the manuscript.

Competing Interests

The authors declare they have no conflict of interest.

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