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

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Abstract

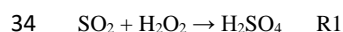
Multiphase chemistry occurs between chemicals in different atmospheric phases, typically involving gas-solid and gas-liquid interactions. The importance of atmospheric multiphase chemistry has long been recognized. Its central role extends from acid precipitation and stratospheric ozone depletion, to 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

31 When *Atmospheric Chemistry and Physics* was launched over two decades ago, Abbatt was invited to submit an

32 article to the inaugural issue, which addressed the kinetics of the multiphase reaction between SO_2 and H_2O_2 on ice

33 surfaces (Clegg and Abbatt, 2001):

34 $SO_2 + H_2O_2 \rightarrow H_2SO_4$ R1

35 This contribution built upon concepts described in a review article published a few years earlier by Ravishankara,

36 which presented a conceptual view of tropospheric heterogeneous and multiphase chemistry (Ravishankara, 1997).

37 Both authors value the opportunity to contribute their opinions on the evolution of our understanding of atmospheric

38 multiphase chemistry. This paper will briefly describe multiphase chemistry, its origins, and the progress made in

39 the past twenty years since the inauguration of *Atmospheric Chemistry and Physics*. It then focuses in depth on

40 future research opportunities and associated challenges. In the 1997 paper, Ravishankara distinguished between

41 heterogeneous and multiphase chemistry based on the extent of diffusion into the bulk. Over the years, it has

42 become clear that diffusion depths vary continuously from solid-like substrates to dilute water solutions. Therefore,

43 in this article, we use the term "multiphase chemistry" to include what was termed heterogeneous chemistry in the

44 late 20th century. For the sake of brevity, the citations in this paper are illustrative and not comprehensive. And so,

45 the reader is directed to in-depth reviews on specific aspects of multiphase chemistry (Ravishankara, 1997; Jacob,

46 2000; Rudich, 2003; Usher et al., 2003; Finlayson-Pitts, 2003; Kolb et al., 2010; George and Abbatt, 2010; Abbatt et

47 al., 2012; Poschl and Shiraiwa, 2015; Burkholder et al., 2017).

48 One underlying theme is that understanding multiphase processes at the molecular level improves our ability to

49 accurately predict atmospheric change, which in turn aids in developing sustainable environmental policy and

50 practices. Positive impacts arise across multiple fields, from climate and air quality to human health and ecology.

51 Another theme is that multiphase chemistry studies are most impactful when closely connected to the entire

52 atmospheric science field, noting the interrelated nature of fundamental chemistry, field measurements, and

53 atmospheric modeling that together constitutes the "three-legged stool" model of our field (see Figure 1) (Abbatt et

54 al., 2014). Multiphase chemistry studies should be conducted to guide, interpret, and encourage field observations,

55 and to quantitatively inform atmospheric models.

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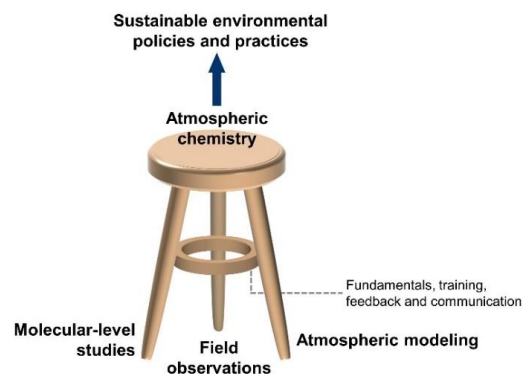
Figure 1: The atmospheric chemistry three-legged stool, where the legs are: i) atmospheric modeling, ii) field

observations, and iii) development of a molecular-level understanding via experimental and theoretical studies 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.

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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.

Despite being very dynamic where the action occurs, with large fluxes of substrate molecules being exchanged

between the phases, one can think of the interface as being a few-molecule-thick "boundary" between the phases at a

given time.

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

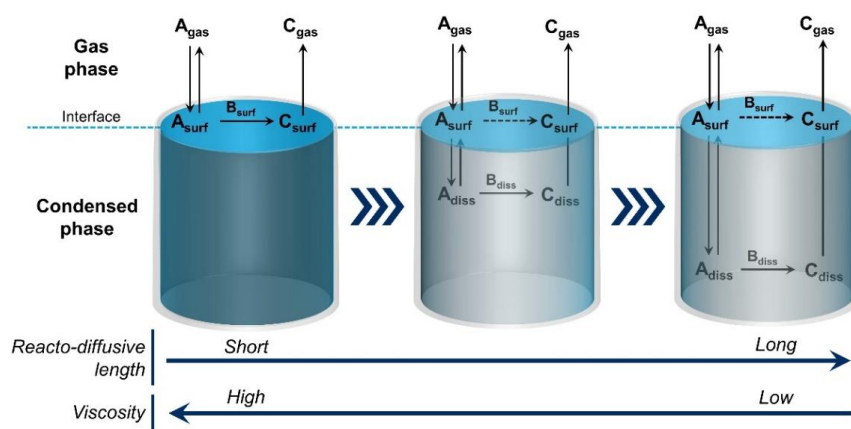
dissolved H_2O_2 to form sulfate (Hoffmann and Edwards, 1975; Penkett et al., 1979). When the substrate is ice

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 a few degrees below its melting point (Girardet and Toubin, 2001; Abbatt, 2003). The

depth to which a molecule diffuses into the bulk before reacting is called the reacto-diffusive length, which can be

81   very short - comparable to the size of a molecule for processes that only occur at the interface - and larger for

82   reactions also occurring in the bulk (see Figure 2) (Hanson et al., 1994).

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84   **Figure 2:** Multiphase reactions occur at the interface and within the bulk of condensed phase materials. Solids and

85   high-viscosity liquids have short reacto-diffusive lengths, which largely restricts multiphase reactions to the

86   interface. As the viscosity lessens and the reacto-diffusive length deepens, multiphase chemistry can occur deeper

87   into the bulk phase. For reactions that proceed in the bulk, some component of the reaction may also simultaneously

88   occur at the interface (as indicated by the dashed reaction arrow). Figure credit: Zilin Zhou.

89

90   **3 How does multiphase chemistry differ from gas-phase chemistry?**

91   Reactions between closed-shell molecules, though thermodynamically allowed, are slow in the gas phase because of

92   large reaction barriers. However, the rates of multiphase reactions involving the same reactants (or suitably altered

93   versions in the condensed phase) can be larger than in the gas phase, either because the free energy barrier to

94   reaction is lowered or because the concentrations of reactants are enhanced in the condensed phase. To illustrate,

95   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

96   dissolves in water and forms HSO_3^-. Moreover, the gas phase H_2O_2 is efficiently scavenged in clouds, enhancing its

97   concentration for reaction. Thus, aerosol and particles are catalyzing such reactions.

98   Another example is the set of reactions that drive polar stratospheric ozone depletion (Solomon, 1999). Chlorine

99   reservoir compounds such as $ClONO_2$ and HCl do not react rapidly in the gas phase. However, HCl partitions

100   strongly to polar stratospheric clouds by either adsorbing to their surfaces or dissolving within them. For example,

101   sorbed HCl dissociates in ice to form chloride ions, which are reactive with $ClONO_2$. As well, $ClONO_2$ can be

102   protonated in strongly acidic cloud droplets, or it may dissociate to form Cl^+ and NO_3^-. Via mechanisms of this

103   nature, the following net reaction proceeds:

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

105   leading to the formation of Cl_2, which is then released to the gas phase and is readily photolyzed, forming radicals

106   that catalytically participate in gas phase ozone destruction in the Antarctic ozone hole.

107   Another distinguishing feature of multiphase chemistry is that it can lead to the formation of products that do not

108   arise in the gas phase. Consider the acid-catalyzed nucleophilic reactions between sulfate and isoprene-derived

109   epoxydiols that form organo-sulfate molecules and secondary organic aerosol (SOA) (Riva et al., 2019). Water

110   molecules lower the transition state energies of such reactions. The solvent shell, which confines reactant partners

111   via the so-called cage effect, can also promote novel products. For example, the formation of biologically-active

112   secondary ozonides is facile in the condensed-phase ozonolysis of unsaturated fatty esters and triglycerides, arising

113   from reactions of Criegee and carbonyl intermediates that form in the same solvent shell after dissociation of a

114   primary ozonide (Zhou et al., 2019b, 2022). In the gas phase, the solvent shell is (generally) absent, so the carbonyl

115   and Criegee intermediates fly apart, and secondary ozonides do not form so readily.

4 Early studies of atmospheric multiphase chemistry

117   Studies of aerosol and cloud chemistry have proceeded in concert with the development of the wider atmospheric

118   chemistry field with many of the concepts of coupled reactivity and mass transfer initially developed by the process-

119   oriented chemical engineering community (Dankwerts, 1970). Interest in multiphase reactions arose via the

120   profound ways these sparse aerosol particles and cloud droplets can alter gas phase composition. As well, as

121   described in more detail below, it is now evident that the reverse is also important, i.e., the gas phase alters the

122   condensed media with important environmental consequences. Many aerosol and multiphase reaction studies were

123   initially performed to develop parameterizations for atmospheric modeling. Even though this is still a major goal,

124   much more effort is now given to understand the physico-chemical processes, which is essential for predictive

125   capabilities.

126   Early studies of atmospheric multiphase chemistry arose from measurements of aerosol composition conducted over

127   a half-century ago. Specifically, continental aerosol particles always contain a measurable quantity of ammonium,

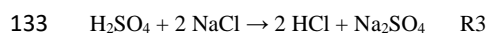
128   indicating the uptake of gas-phase ammonia to acidic particles (Lee and Patterson, 1969; Kadowaki, 1976).

129   Furthermore, particulate chloride levels in the marine aerosol are depleted relative to their seawater abundance,

130   replaced by sulfate or nitrate (Junge, 1956; Martens et al., 1973). This process was long thought to be the major

131   source of gas-phase chlorine, whereby gaseous HCl is displaced from NaCl particles via the uptake of gas-phase

132   strong acids :

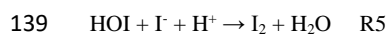
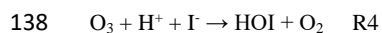
133   $H_2SO_4 + 2\ NaCl \rightarrow 2\ HCl + Na_2SO_4$ R3

134   Another early example of halogen chemistry demonstrated that volatile iodine species are released when dissolved

135   oceanic iodide is exposed to either ultraviolet light or ozone (Miyake and Tsunogai, 1963; Garland et al., 1980).

136   This multiphase chemistry is now recognized as important for the dry deposition of ozone and the release of iodine

137   into the atmosphere (Carpenter et al., 2013):

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

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

140   After such studies, as well as significant research into acid rain formation (e.g., Penkett et al., 1979) and

141   stratospheric ozone depletion (e.g., Solomon, 1999), many additional important tropospheric multiphase chemical

142   processes were identified prior to the launch of *Atmospheric Chemsitry and Physics*: i) the formation of reactive

143   halogen species in the boundary layer (Finlayson-Pitts, 2003; Simpson et al., 2015), ii) the uptake of tropospheric

144   gases by mineral dust, especially nitric acid (Hanisch and Crowley, 2001; Usher et al., 2003), iii) the scavenging of

145   trace gases, such as nitric acid and small oxygenated VOCs, by snow and ice crystals in the free and upper

146   troposphere (Abbatt, 2003), iv) cloud water chemistry, leading to loss of HO_x radicals (Chameides and Davis, 1982;

147   Calvert et al., 1985; Jacob, 1986; Lelieveld and Crutzen, 1991), v) conversion of N_2O_5 to HNO_3 on tropospheric

148   aerosol, with impacts on the NO_x budget (Dentener and Crutzen, 1993), vi) uptake of HO_2 to aerosol (Mozurkewich

149   et al., 1987; Martin et al., 2003), and vii) multiphase conversion of NO_2 to HONO (Finlayson-Pitts et al., 2003). A

150   critical point is that each of these multiphase processes affects the oxidizing capacity of the troposphere, frequently

151   through modification of radical budgets. For example, these processes initiate oxidation in urban atmospheres

152   through HONO photolysis, drive Arctic boundary layer ozone and mercury depletion via gas-phase halogen

153   chemistry, and modulate the global oxidizing capacity via N_2O_5 or HO_2 loss on aerosol particles.

154   **5 Progress in the past twenty years**

155   Two major developments profoundly influenced multiphase chemistry. First was the recognition of the importance

156   of aerosol particles in changing the radiative balance of the Earth system, with impacts on climate. The second was

157   the continued recognition of the deleterious effects of particulate matter on human, animal, and ecosystem health.

158   These two fields, climate change and air quality, have provided the impetus (and resources) for the development of

159   the field. As a result, multiphase chemistry studies did an about-face at the turn of the 21st century as the research

160   focus shifted away from influences on the gas phase to the impacts that arise on the composition of the particles.

161   Once inhaled, particles harm human health (Murray et al., 2020), with recent studies implicating the secondary

162   component of the particles in negative health outcomes (Pye et al., 2021). Research in the past two decades has

163   focused strongly on the formation of SOA (Kroll and Seinfeld, 2008; Hallquist et al., 2009; Ziemann and Atkinson,

164   2012; Shrivastava et al., 2017). SOA formation has required better knowledge of the kinetics and mechanisms of gas

165   phase oxidation of SOA precursors (Crounse et al., 2013; Ehn et al., 2014). It has also needed a more complete

166   understanding of gas-particle nucleation processes (Kulmala et al., 2014; Trostl et al., 2016; Xiao et al., 2021),

167   volatility (Donahue et al., 2011), and condensed-phase reactions that occur within aerosol particles. Specifically,

168   volatility and multiphase reactivity can be coupled, as illustrated by the formation of high molecular weight, low

169   volatility species within particles (Kalberer et al., 2004). While such oligomers and highly oxygenated species may

170   also form in the gas phase (Bianchi et al., 2019), they arise via a variety of reactions involving pairs of organic

171   reactants, frequently forming esters and acetals/hemiacetals in the condensed phase (Tobias and Ziemann, 2000;

172   Surratt et al., 2006; DeVault and Ziemann, 2021). These reactions may be acid-catalyzed (Jang et al., 2002). As

173   well, multiphase oxidation by gas-phase O_3, OH, and NO_3 can increase the average oxidation state of organic

174   aerosol particles (Kroll et al., 2011) via a series of reactions that initially functionalize and eventually fragment the

175   component molecules (George et al., 2007; Kroll et al., 2009). Oxidation leads to a more soluble particle that

176   increases its rate of wet deposition.

177   Tightly connected to SOA formation and modification processes are the condensed phase viscosity and phase state,

178   which set mixing times within particles and are dependent on relative humidity and temperature (Koop et al., 2011;

179   Renbaum-Wolff et al., 2013). Organic particles are likely glasses in the cold free troposphere (Shiraiwa et al., 2017),

180   which slows SOA formation and restricts the degree to which heterogeneous oxidation can affect the aerosol

181   composition. The particles are liquids in warm, wet boundary layers, with the full particle volume involved in

182   partitioning with gas phase molecules. The large variation in molecular diffusion coefficients and associated

183   mobility determines where chemical reactions are important in the particles, from two-dimensional processes that

184   occur solely at the gas-particle interface to three-dimensional chemistry with reactivity both at the interface and

185   deeper in the bulk (see Figure 2). Overall, diffusion is a key parameter for determining whether a reaction is surface-

186   area-limited or volume-limited (Hanson et al., 1994).

187   Multiphase chemistry also leads to the formation of secondary inorganic aerosol. For example, the hydrolysis of

188   N_2O_5 converts NO_x to HNO_3; the gas-particle partitioning of HNO_3 is then controlled by temperature, relative

189   humidity, and ammonia levels. As well, particulate sulfate is rapidly formed in polluted environments through

190   multiphase aqueous chemistry, acting as the major formation mechanism in cloud-free settings (Cheng et al., 2016;

191   Wang et al., 2016). Potential routes for fast sulfate formation in deliquesced particles include: the role of ionic

192   strength in accelerating the rates of specific processes (Liu et al., 2020), fast interfacial chemistry (Liu and Abbatt,

193   2021), formation of condensed-phase oxidants through the photolysis of particulate nitrate (Zheng et al., 2020), and

194   the role of specific particle-phase reactants, such as organic hydroperoxides (Wang et al., 2019),

195   hydroxymethanesulfonate (Song et al., 2019), and dissolved transition metal ions (Li et al., 2020b) An accurate

196   quantitative assessment of these reaction pathways is still developing (Liu et al., 2021b).

197   As noted earlier, the need to better understand aerosol-climate interactions has also motivated multiphase chemistry

198   research in the past twenty years. Atmospheric processing leads to the formation of water-soluble condensed-phase

199   products, such as sulfate or highly oxygenated organic molecules (Jimenez et al., 2009), enhancing the abilities of

200   tropospheric aerosol particles to act as cloud condensation nuclei (CCN) and affecting their ability to scatter light

201   (Cappa et al., 2011; Moise et al., 2015). As well, the optical properties of the fraction of organic aerosol that

202   absorbs near ultraviolet and visible light (i.e., atmospheric 'brown carbon' particles) is subject to change via

203   multiphase oxidation and condensed phase photochemistry (Laskin et al., 2015; Li et al., 2020a; Hems et al., 2021;

204   Schnitzler et al., 2022). Although the rates of optical property changes remain uncertain, primary brown carbon

205   particles, as formed in wildfires, tend to become less absorbing in the near UV and visible parts of the spectrum on

206   the timescale of days via a variety of multiphase aging mechanisms (Hems et al., 2021), i.e., they are 'bleached'.

207   The diminution of light absorption is in accord with field observations (Forrister et al., 2015).

208   Multiphase chemistry can also affect the properties of ice nucleating particles (INPs), by both gas-solid and liquid-

209   solid interactions. For example, mineral dust can have its IN activity decreased by condensation of involatile

210   materials, such as sulfate or by cloud processing (Sullivan et al., 2010b; Kilchhofer et al., 2021), and strong acids

211   can react with carbonate-containing minerals, leading to particles that are less IN-active in the deposition mode but

212   more active in the immersion mode (Sullivan et al., 2010a). Such effects can also arise with the exposure of

213   different gas and liquid species to volcanic ashes (Maters et al., 2020; Fahy et al., 2022). Oxidation reactions can

214   also occur, so that efficient biological INPs, such as pollen fragments, lose activity upon oxidation by OH radicals,

215   probably by morphological changes of surface proteins and carbohydrates (Gute and Abbatt, 2018). The

216   mechanisms involving all these interactions are very complex. In the case of mineral dusts, immersion INP activity

217   can be changed by surface modification, ion exchange, adsorption of solutes such as ammonium, and acid

218   dissolution (Sihvonen et al., 2014; Kumar et al., 2019; Yun et al., 2021).

219

220   **6 The future of atmospheric multiphase chemistry studies**

221   **6.1 Multiphase chemistry at the interfaces of the atmosphere**

222   There are exciting opportunities for applying the conceptual, instrumental, and modeling tools developed for

223   multiphase chemistry to understand chemistry occurring at the interfaces of the atmosphere with other

224   environmental domains.

225   Consider the interface of the atmosphere and the ocean, where the sea-surface microlayer (SML) is a thin layer of

226   ocean water that has enhanced concentrations of biological detritus, surface-active compounds, and gel-like

227   substances (Cunliffe et al., 2013). Recognizing that individual molecule surrogates of the SML only capture specific

228   aspects of the chemistry, experimental designs now involve either genuine seawater or water samples with

229   significant biological components (Prather et al., 2013; Schneider et al., 2019). While we know that the SML can

230   affect the composition of primary marine aerosol, an open question is the degree of chemical processing by

231   photosensitization in the SML or by gas-surface heterogeneous oxidation, yielding volatile species that contribute to

232   marine SOA formation (Donaldson and George, 2012; Rossignol et al., 2016; Mungall et al., 2017; Croft et al.,

233   2019).

234   Another key role of multiphase reactions is in dry deposition processes on the ocean (e.g., see R4 and R5),

235   vegetation, the built environment, and land surfaces (Garland et al., 1980; Fowler et al., 2009; Kavassalis and

236   Murphy, 2017; Tuite et al., 2021). Deposition is a critical step that controls removing chemicals from the

237   atmosphere. Yet, this process is essentially a parameterization in models. Deposition in many environments needs to

238   be predictive, which demands molecular-level understanding and quantification. This process is essentially a

239   multiphase process that should be broken down into physico-chemical steps, which can be independently measured
240   and understood.

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

255   **6.2 Multiphase chemistry and human health**

256   Although the results of epidemiological studies implicate aerosol particle inhalation as harmful to human health
257   (Pope et al., 2009), the mechanistic relationship between atmospheric particle composition and negative health
258   impacts remains elusive. The current epidemiology does not readily distinguish the specific molecules and formation
259   pathways that lead to negative health outcomes, nor the toxicity mechanism. Although there is debate over whether
260   oxidants are largely endogenous or exogenous (Fang et al., 2022), one hypothesis is that the biochemical balance
261   between oxidants and antioxidants is upset by inhaling harmful species (Miller, 2020). To contribute to this debate,
262   the multiphase chemistry community needs to better describe the chemistry that occurs at the lung-air interface and
263   the composition of respirable aerosol particles, especially the biologically active components that contain reactive
264   functional groups (e.g., epoxides, hydroperoxides), redox-active materials (e.g., quinones), and reactive oxygen
265   species (e.g., peroxides, HO_2/O_2^-). Many of these species are formed by multiphase oxidation processes.

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

271   Largely unexplored until the recent pandemic is the impact of the atmosphere on airborne and surface-deposited
272   biological pathogens, including bacteria and viruses. Early work in this area included the multiphase chemistry
273   between NO_2 and proteinaceous material, motivated by its potential to drive an allergenic response (Franze et al.,

274 2005; Shiraiwa et al., 2012). As well, gas phase O_3 has been examined for its ability to affect the viability of

275 bacteriophages, i.e., microorganisms with a lipid envelope and RNA core similar to the structure of SARS-CoV-2,

276 deposited on surfaces (Tseng and Li, 2008). With the pandemic, research has accelerated into the impact of

277 hygroscopic growth and water content on viral viability within respiratory particle surrogates that consist of viruses

278 embedded in saline droplets containing surfactants, proteins, and carbohydrates. It is important to understand the

279 changes in the acidity of these particles, the mass transfer within them, and the precipitation of salts as the particle

280 water content changes (Lin et al., 2020; Oswin et al., 2022; Huynh et al., 2022).

281 The pandemic led to an emphasis on the cleaning of surfaces to reduce the potential for infection by fomites, i.e., via

282 contact with contaminated surfaces. While cleaning agents such as chlorine bleach have well-established anti-

283 microbial activity, their multiphase chemistry can release gases and particles that are deleterious to human health

284 (Wong et al., 2017; Mattila et al., 2020). Understanding the multiphase chemistry associated with these cleaning

285 activities and the outcomes of using air cleaners (Collins and Farmer, 2021), is essential for establishing healthy

286 indoor environments.

287 Lastly, the pandemic prompted a re-examination of an overlooked aspect of our atmosphere that it has an as-yet-

288 unidentified germicidal component referred to as the Open Air Factor (Cox et al., 2021). In particular, it was shown

289 many decades ago that fresh air led to better outcomes for tuberculosis patients and injured World War I soldiers

290 than indoor air. Historically, sending sick people to pristine environments (e.g., the seaside) was a common medical

291 recommendation. It is not clear whether this effect is related to multiphase chemistry at biological surfaces.

292 Each of the above topics provides exciting opportunities for atmospheric chemists to collaborate with the

293 environmental health, medical, and toxicological communities.

6.3 Chemistry of the free troposphere and lower stratosphere

295 Although the upper troposphere – lower stratosphere region was the focus of much attention in the 1980s and 1990s

296 to understand the changes in ozone levels in these regions, most multiphase chemistry studies are currently

297 conducted at room temperature. There is considerable motivation to re-explore chemistry at colder temperatures,

298 given past work that illustrated the atmospheric impacts of a strongly non-linear dependence of multiphase reactions

299 rates on temperature (Murphy and Ravishankara, 1994) and extensive new observations from the Atom campaigns

300 (Thompson, 2022) that sampled from the boundary layer to the upper troposphere over many latitudes and seasons.

301 As well, there is emerging evidence for organic aerosol in the lower stratosphere, likely arising from wildfire

302 injection, with potential influence on stratospheric ozone (Solomon et al., 2022; Strahan et al., 2022).

303 Organic aerosol viscosity and phase state depend on the environmental conditions (Koop et al., 2011), with semi-

304 solid and glassy organic particles predicted throughout much of the free troposphere (Shiraiwa et al., 2017). Aside

305 from those at the gas-particle interface, molecules in highly viscous organic particles are protected from

306 heterogeneous oxidation (Shiraiwa et al., 2011; Zhou et al., 2012). Such protection increases the lifetimes of

307 pollutants, e.g., brown carbon chromophores (Schnitzler et al., 2022), and lengthens particles' oxidation timescale

308 and wet deposition lifetime. In addition, we need to better understand cloud chemistry at cold temperatures,

309   especially under supercooled water conditions. As well, we lack a well-validated model for the retention of

310   molecules in supercooled water droplets as they freeze to form ice; the retention efficiency of dissolved solutes

311   affects the composition of convective outflow from deep cloud systems.

312   A key factor affected by temperature is the solubility of various atmospheric constituents. Simple Henry's law

313   constants and further equilibration steps that determine the overall solubilities are poorly known, especially below

314   room temperature. Most of the data on the solubilities in organics goes back to chemical engineering literature that

315   is more than half a century old. Also, since solubilities vary according to Henry's law equilibria that vary

316   exponentially with temperature, the accurate temperature dependence of solubilities is essential. Acid dissociation

317   constants in organic acids and organic substrates are poorly known, and they determine the overall solubility of a

318   chemical.

6.4 Reactive transformations of organic chemical contaminants

320   Over forty years ago, it was recognized that multiphase oxidation of chemical contaminants leads to the rapid loss of

321   surface-bound PAHs and the formation of more toxic and potentially carcinogenic products such as nitro-PAHs and

322   oxygenated PAHs (Pitts et al., 1978, 1980). These reactions occur on a variety of surfaces with light, ozone, and

323   NO_2 reactants, some via Langmuir-Hinshelwood mechanisms (Poschl et al., 2001; Mmereki and Donaldson, 2003;

324   Kwamena et al., 2004). Buried PAHs are protected from heterogeneous loss by a crust of unreactive products that

325   accumulates upon them and, when present, within viscous organic aerosol (Zhou et al., 2013, 2019a), enabling the

326   potential for long-range atmospheric transport (Mu et al., 2018). The chemistry of other organic contaminants,

327   including smoking products such as nicotine (Destaillats et al., 2006) and tetrahydrocannabinol (Yeh et al., 2022), a

328   few pesticides (Segal-Rosenheimer and Dubowski, 2007; Finlayson-Pitts et al., 2022), and organophosphate esters

329   (Liu et al., 2021a), has also been recently explored.

330   However, these are largely exceptions, and the multiphase fate of most chemical contaminants, especially thousands

331   of commercial products, has not been examined. Indeed, the atmospheric chemistry and chemical contaminant

332   communities have traditionally not strongly interacted. Although assessment of the gas phase OH reactivity is

333   customarily performed in environmental fate analyses (Li et al., 2019b), many commercial products have

334   sufficiently low volatility that they reside primarily on surfaces or within particles. It is important to establish

335   whether organic contaminants traditionally viewed as persistent are indeed unreactive with respect to multiphase

336   transformation.

6.5 Understanding the role of light

338   Many condensed-phase photochemical reactions proceed via indirect mechanisms where a photosensitizing

339   molecule absorbs light, forming reactive species such as HO_2/O_2^- or $^1O_2$ (George et al., 2015). Such chemistry has

340   been implicated in the daytime formation of HONO (George et al., 2005), the photoreactions of brown carbon

341   aerosol (Laskin et al., 2015; Hems et al., 2021), the formation of active halogens (Reeser et al., 2009), and reactivity

342   of black carbon (Monge et al., 2010; Li et al., 2019c). This chemistry has been illustrated using efficient

343   photosensitizing agents, but quantitative assessments of atmospheric importance remain uncertain largely because

344   the character and quantity of atmospheric photosensitizers are not well established. Developing a tighter quantitative

345   connection to the atmosphere will require using more representative photosensitizers, as now being done using

346   marine aerosol components (Ciuraru et al., 2015; Garcia et al., 2021). For the troposphere, the wavelengths of

347   interest are in the near UV and visible part of the solar flux.

348   In addition to sensitizing, light can also lead to direct photochemistry in some environments. Photolysis on ice and

349   snow surfaces has been implicated in the formation of NO_x in midlatitudes and polar regions (Honrath et al., 1999;

350   Wolff et al., 2002), and the photolytic lifetimes of condensed phase organic hydroperoxides may determine their

351   atmospheric lifetimes. It is important to recognize the absorption spectra of dissolved molecules can be different

352   than those in the gas phase. Similarly, the quantum yield for the loss of the molecule and the products can be very

353   different than in the gas phase.

6.6 Developments in Observational Capabilities

355   Our ability to characterize atmospheric composition continues to push the field of atmospheric chemistry forward.

356   For multiphase chemistry, advances in analytical mass spectrometry have been transformative. Within the last

357   twenty years, online characterization of aerosol composition has become commonplace (Canagaratna et al., 2007),

358   studies of single particle composition allow us to observe the variability in mixing state and chemical diversity

359   (Murphy et al., 2006; Prather et al., 2008), and offline filter sampling has progressed from the characterization of a

360   few targeted species to non-targeted analyses using a range of mass spectral ionization methods (Papazian et al.,

361   2022; Ditto et al., 2022). Identifying specific molecular "markers" for organics and functional groups is still

362   somewhat uncertain; developing such identification would be very helpful.

363   The continued development of analytical techniques will enable increasingly sophisticated characterization of

364   aerosol particles and environmental surfaces, with the opportunity to deploy the same tools in both lab and field

365   settings. However, challenges are arising as well. Despite the rapid development of low-cost sensors, affordable

366   instrumentation for the long-term characterization of aerosol composition in many locations is still lacking. The

367   increasing sophistication of analytical instrumentation also continues to unveil the high degree of chemical

368   complexity present. Whereas high-resolution mass spectrometry yields chemical formulae in real-time, there is

369   often the need to identify chemical structures. This suggests that we should increasingly deploy separation

370   techniques (e.g., chromatography, ion mobility) as front ends to our increasingly sophisticated mass spectrometric

371   techniques (Krechmer et al., 2016; Claflin et al., 2021). There is also value to the expanded use of other classical

372   chemical speciation methods, such as infrared (Russell, 2003) and NMR (Decesari et al., 2007) characterization of

373   aerosol composition collected by filters. While these techniques have low time resolution, they provide

374   complementary quantitative and functional group information and can be inexpensively deployed for long-term

375   analyses in a wide range of environments. Such analyses will also help with the source apportionment of the

376   aerosols.

377   Aerosol characteristics related to multiphase chemistry can be studied with increasingly sophisticated remote

378   sensing techniques. These approaches have been applied for many years to polar stratospheric clouds, whose

379 composition and phase (via the degree of depolarization of a lidar probe) have been studied (Tritscher et al., 2021).
380 Another example comes from satellite measurements of solid ammonium nitrate particles in the upper troposphere,
381 driven by the Asian monsoon that uplifts ammonia-rich continental air (Hopfner et al., 2019). It is important to
382 determine the role of these particles in ice nucleation and multiphase chemistry.

383 **6.7 Grappling with chemical complexity**

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

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

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

409 Both bottom-up and top-down approaches can address chemical complexity. In the traditional bottom-up approach,
410 the effects on the reaction system of step-by-step additions of chemical complexity are evaluated. This leads to a
411 better understanding of the fundamental chemistry needed to develop our predictive abilities. Top-down approaches
412 involve studying chemistry on ambient aerosol particles. This has been done for N_2O_5 and HO_2 aerosol uptake
413 (Bertram et al., 2009; Zhou et al., 2021), for heterogeneous OH oxidation (George et al., 2008), and by using mobile

414   reaction chambers to characterize SOA formation (Jorga et al., 2021). Another top-down method constrains the

415   rates of multiphase chemistry using detailed, simultaneous measurements of gas-phase composition under a range of

416   environmental conditions (Brown et al., 2006). Combining top-down and bottom-up approaches enhances our

417   understanding of the fundamental science while ensuring that parameterizations for atmospheric modeling are

418   accurate.

419   Increasingly sophisticated chemical models are addressing chemical complexity. Molecular dynamics simulations,

420   coupled with ab initio calculations, are sufficiently advanced to usefully guide and interpret some laboratory studies

421   (Fang et al., 2019; Yang et al., 2019). Whereas past computational methods only included a few solvent molecules,

422   current dynamics models using state-of-the-art force fields can realistically simulate partitioning, surface adsorption

423   constants, diffusion constants, and vapor pressures, representing an important point of contact to the physical

424   chemistry and chemical physics communities (Tobias et al., 2013). For establishing some fundamental parameters

425   that are hard to measure, such as the likelihood that a collision of a molecule with a particle leads to uptake by the

426   condensed phase (i.e., a mass accommodation coefficient), the theory may be a preferable approach to experiment.

427   Multiphase kinetics models incorporate emerging insights from both the lab and theory. A successful approach has

428   arisen in the indoor chemistry community where modeling groups using a wide range of tools, from molecular

429   dynamics to large-scale computational fluid dynamics, interact closely with each other and with experimental

430   scientists (Shiraiwa et al., 2019).

431   **7 Concluding thoughts**

432   • Multiphase chemistry has evolved alongside the wider field of atmospheric chemistry. While initial studies

433   focused on its impacts on the gas phase, the field now addresses how chemistry affects particles. Although

434   modification of aerosol composition has direct relevance to climate and human health, we should not lose sight

435   of the connection of multiphase chemistry to the gas phase composition of the atmosphere.

436   • We need to understand chemical processes at the molecular level to improve our ability to interpret field

437   observations and predict the nature of a changing atmosphere. Such fundamental understanding is unlikely to

438   arise using artificial intelligence – machine learning approaches, which can only extrapolate to future

439   environments using known chemistry and behavior. Reinforcing an approach based on physico-chemical

440   understanding is necessary for detailed predictions of environmental change.

441   • There are significant research opportunities for the characterization of the chemistry that occurs at the interface

442   of the atmosphere with the rest of the environment, such as studies of ocean-atmosphere interactions, indoor air,

443   aerosol health effects, and pathogen-air interactions.

444   • With increasingly sophisticated experimental and theoretical tools, atmospheric chemical complexity becomes

445   more apparent. While exciting, this presents challenges and constraints. We should emphasize not only highly

446   detailed, molecular-level measurements but also more widespread and more prolonged timescale aerosol

447   characterization that has less chemical specificity but nevertheless provides valuable insights; there is also a role

448   for both remote sensing measurements and classical analytical techniques in this regard. This is akin to a need

449   to understand and quantify thermal gas phase reactions while also understanding and quantifying

450   microcanonical reactivity.

451   • Measurements of many fundamental physico-chemical parameters such as solubility, diffusion coefficients, and

452   liquid/solid phase reactivities are sorely needed.

453   • Multiphase chemistry studies conducted under conditions that match those in the atmosphere, including those of

454   the free troposphere and lower stratosphere, are needed.

455   • Using the atmosphere as a laboratory to quantify rates of multiphase processes holds promise, with

456   simultaneous measurements of many chemicals and other external parameters becoming more feasible through

457   coordinated field measurements. Designing field studies with an eye toward quantification of the multiphase

458   reactions is beneficial.

459   • The field of atmospheric chemistry is healthiest when there is extensive communication and feedback between

460   the fundamental chemistry, modeling, and field observation communities. To keep the three-legged stool

461   balanced and strong, multiphase chemists should interact widely with other atmospheric chemists, exploring

462   training and collaborative opportunities that bridge the different legs of the stool (see Figure 1).

463

464   **Author Contributions**

465   Both authors contributed to writing the manuscript.

466   **Competing Interests**

467   The authors declare they have no conflict of interest.

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470

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