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**Article:**

Burkholder, JB, Abbatt, JPD, Barnes, I et al. (31 more authors) (2017) The Essential Role for Laboratory Studies in Atmospheric Chemistry. *Environmental Science & Technology*, 51 (5). pp. 2519-2528. ISSN 0013-936X

<https://doi.org/10.1021/acs.est.6b04947>

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# The Essential Role for Laboratory Studies in Atmospheric Chemistry

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James B. Burkholder\*, Earth System Research Laboratory, Chemical Sciences Division,  
National Oceanic and Atmospheric Administration, Boulder, CO 80305, USA,  
James.B.Burkholder@noaa.gov

Jonathan P. D. Abbatt\*, Department of Chemistry, University of Toronto, ON, Canada M5S  
3H6, jabbatt@chem.utoronto.ca

Ian Barnes, University of Wuppertal, School of Mathematics and Natural Science, Institute of  
Atmospheric and Environmental Research, Gauss Strasse 20, 42119 Wuppertal, Germany,  
barnes@uni-wuppertal.de

James M. Roberts, Earth System Research Laboratory, Chemical Sciences Division, National  
Oceanic and Atmospheric Administration, Boulder, CO 80305, USA,  
James.M.Roberts@noaa.gov

Megan L. Melamed, IGAC Executive Officer, University of Colorado/CIRES, Boulder, CO  
80309-0216 USA, megan@igacproject.org

Markus Ammann, Laboratory of Environmental Chemistry, Paul Scherrer Institute, Villigen,  
5232, Switzerland, markus.ammann@psi.ch

24 Allan K. Bertram, Department of Chemistry, The University of British Columbia, Vancouver,  
25 British Columbia, Canada V6T 1Z1, bertram@chem.ubc.ca

26 Christopher D. Cappa, Department of Civil and Environmental Engineering, University of  
27 California, Davis, CA 95616, cdcappa@ucdavis.edu

28 Annmarie G. Carlton, Department of Chemistry, University of California, Irvine, CA, 92617,  
29 agcarlto@uci.edu

30 Lucy J. Carpenter, Wolfson Atmospheric Chemistry Laboratories, Department of Chemistry,  
31 University of York, York, United Kingdom, YO10 5DD, lucy.carpenter@york.ac.uk

32 John N. Crowley, Max Planck Institut für Chemie, Mainz, Germany, john.crowley@mpic.de

33 Yael Dubowski, Faculty of Civil and Environmental Engineering Technion, Israel Institute of  
34 Technology, Haifa 32000, Israel, yaeld@tx.technion.ac.il

35 Christian George, Université Lyon 1, F-69626, France; CNRS, UMR5256, IRCELYON, Institut  
36 de recherches sur la catalyse et l'environnement de Lyon, Villeurbanne, F-69626, France,  
37 christian.george@ircelyon.univ-lyon1.fr

38 Dwayne E. Heard, School of Chemistry, University of Leeds, Leeds, LS2 9JT, United Kingdom,  
39 D.E.Heard@leeds.ac.uk

40 Hartmut Herrmann, Leibniz-Institut für Troposphärenforschung (TROPOS), D-04318 Leipzig,  
41 Germany, herrmann@tropos.de

42 Frank N. Keutsch, Department of Chemistry and Chemical Biology, Harvard University,  
43 Cambridge, MA 02128, keutsch@seas.harvard.edu

44 Jesse H. Kroll, Department of Civil and Environmental Engineering, Department of Chemical  
45 Engineering, Massachusetts Institute of Technology, Cambridge, MA 02139, USA,  
46 [jhkroll@mit.edu](mailto:jhkroll@mit.edu)

47 V. Faye McNeill, Chemical Engineering, Columbia University, New York, NY,  
48 [vfm2103@columbia.edu](mailto:vfm2103@columbia.edu)

49 Nga Lee Ng, School of Chemical & Biomolecular Engineering and School of Earth and  
50 Atmospheric Sciences, Georgia Institute of Technology, Atlanta, Georgia, [ng@chbe.gatech.edu](mailto:ng@chbe.gatech.edu)

51 Sergey A. Nizkorodov, Department of Chemistry University of California, Irvine, California  
52 [92697\\_nizkorod@uci.edu](mailto:92697_nizkorod@uci.edu)

53 John J. Orlando, National Center for Atmospheric Research, Atmospheric Chemistry  
54 Observations and Modeling Laboratory, Boulder, CO 80301, [orlando@ucar.edu](mailto:orlando@ucar.edu)

55 Carl J. Percival, School of Earth, Atmospheric and Environmental Sciences, University of  
56 Manchester, Manchester, United Kingdom, [carl.percival@manchester.ac.uk](mailto:carl.percival@manchester.ac.uk)

57 Bénédicte Picquet-Varrault, Laboratoire Interuniversitaire des Systèmes Atmosphériques  
58 (LISA), UMR 7583 CNRS, Universités Paris-Est Créteil et Paris Diderot, Institut Pierre-Simon  
59 Laplace, Créteil Cedex, France, [benedicte.picquet-varrault@lisa.u-pec.fr](mailto:benedicte.picquet-varrault@lisa.u-pec.fr)

60 Yinon Rudich, Department of Earth and Planetary Sciences, Weizmann Institute of Science,  
61 Rehovot 76100, Israel, [yinon.rudich@weizmann.ac.il](mailto:yinon.rudich@weizmann.ac.il)

62 Paul W. Seakins, School of Chemistry, University of Leeds, Leeds, LS2 9JT, United Kingdom,  
63 [P.W.Seakins@leeds.ac.uk](mailto:P.W.Seakins@leeds.ac.uk)

64 Jason D. Surratt, Department of Environmental Sciences and Engineering, University of North  
65 Carolina at Chapel Hill, Chapel Hill, NC 27599, [surratt@unc.edu](mailto:surratt@unc.edu)

66 Hiroshi Tanimoto, National Institute for Environmental Studies, Tsukuba, Ibaraki Japan,  
67 [tanimoto@nies.go.jp](mailto:tanimoto@nies.go.jp)

68 Joel A. Thornton, Department of Atmospheric Sciences, University of Washington, Seattle, WA  
69 98195, [thornton@atmos.washington.edu](mailto:thornton@atmos.washington.edu)

70 Zhu Tong, College of Environmental Sciences and Engineering, Peking University, China,  
71 [tzhu@pku.edu.cn](mailto:tzhu@pku.edu.cn)

72 Geoffrey S. Tyndall, National Center for Atmospheric Research, Atmospheric Chemistry  
73 Observations and Modeling Laboratory, Boulder, CO 80301, [tyndall@ucar.edu](mailto:tyndall@ucar.edu)

74 Andreas Wahner, Institute of Energy and Climate Research, IEK-8: Troposphere,  
75 Forschungszentrum Jülich GmbH, D-52425 Jülich, Germany, [a.wahner@fz-juelich.de](mailto:a.wahner@fz-juelich.de)

76 Charles J. Weschler, Environmental & Occupational Health Sciences Institute, Rutgers  
77 University, Piscataway, NJ 08854, [weschler@rwjms.rutgers.edu](mailto:weschler@rwjms.rutgers.edu)

78 Kevin R. Wilson, Chemical Sciences Division, Lawrence Berkeley National Laboratory,  
79 Berkeley, CA, [krwilson@lbl.gov](mailto:krwilson@lbl.gov)

80 Paul J. Ziemann, Department of Chemistry and Cooperative Institute for Research in  
81 Environmental Sciences, University of Colorado, Boulder, CO, 80309,  
82 [paul.ziemann@colorado.edu](mailto:paul.ziemann@colorado.edu)

83

84 **Abstract**

85 Laboratory studies of atmospheric chemistry characterize the nature of atmospherically relevant  
86 processes down to the molecular level, providing fundamental information used to assess how  
87 human activities drive environmental phenomena such as climate change, urban air pollution,  
88 ecosystem health, indoor air quality, and stratospheric ozone depletion. Laboratory studies have  
89 a central role in addressing the incomplete fundamental knowledge of atmospheric chemistry.  
90 This article highlights the evolving science needs for this community and emphasizes how our  
91 knowledge is far from complete, hindering our ability to predict the future state of our  
92 atmosphere and to respond to emerging global environmental change issues. Laboratory studies  
93 provide rich opportunities to expand our understanding of the atmosphere via collaborative  
94 research with the modeling and field measurement communities, and with neighbouring  
95 disciplines.

96 **Introduction**

97 Atmospheric chemistry is the study of the chemical processes that affect the composition of the  
98 atmosphere, encompassing societally important issues such as air pollution and its related health  
99 and ecosystem effects, as well as climate (see Figure 1). This field unifies the evaluation of both  
100 natural and anthropogenic emissions, measurements of atmospheric composition across a range  
101 of temporal and spatial scales, and assessment of deposition processes that ultimately remove  
102 chemical compounds from the air. All of these processes are impacted by, or may feedback on,  
103 climate change. At the heart of atmospheric chemistry are chemical transformations, often  
104 initiated by sunlight. Molecules absorb solar radiation, leading to a cascade of catalytic and  
105 oxidative reactions. Our molecular-level understanding of atmospheric chemistry has

106 successfully provided the foundations of air quality forecasts including those related to acid  
107 rain,<sup>1</sup> the basis upon which the chemicals responsible for the formation of the Ozone Hole were  
108 banned under the Montreal Protocol and its amendments,<sup>2</sup> and connections between human  
109 impact on atmospheric composition and climate change.<sup>3</sup> The evaluation of geoengineering as an  
110 option to offset global warming also requires a detailed understanding of the underlying  
111 chemistry.<sup>4</sup>

112 Simplified parameterizations of this detailed chemistry developed from well-controlled  
113 laboratory studies, alongside representations of emissions, transport and deposition processes,  
114 are incorporated into computer models to predict atmospheric composition across appropriate  
115 temporal and spatial (local, regional, and global) scales. Comparisons with measurements from  
116 the field are often used to evaluate model predictions or to help identify poorly represented  
117 chemistry. Laboratory studies are an essential bridge between field measurements and models  
118 and provide the basic (or fundamental) physics and chemistry of the underlying mechanisms of  
119 phenomena observed in the field, which is the basis for generating physically meaningful  
120 parameterizations for use in models. This interplay between laboratory studies of chemical,  
121 photochemical, and physical processes, atmospheric modeling, and field measurements has  
122 propelled the field of atmospheric chemistry forward to achieve remarkable success in  
123 understanding the details of stratospheric ozone depletion, acid rain, urban air quality, and the  
124 chemistry of climate-forcing agents, and in informing environmental policies (see Figure 2).

125 The increased emphasis by funding agencies on solutions-driven research, as opposed to  
126 fundamental research of the atmosphere in general, inherently demotivates the development of a  
127 comprehensive understanding of the core processes occurring in the atmosphere. In particular,  
128 attention and resources are funneled into important but highly specific sub-fields, such as

129 detailed studies of the atmospheric impacts of different energy source activities including light  
130 oil extraction, hydraulic fracturing or bitumen extraction.<sup>5</sup> Laboratory studies, together with  
131 field and modeling studies, constitute the long-standing “three-legged stool” of atmospheric  
132 chemistry. The fundamental science aspect of this "three-legged stool" is overshadowed when  
133 the science is solutions-driven, lessening our ability to not only address known environmental  
134 issues such as the relationship of air quality to human health but also our readiness to respond to  
135 unforeseen future environmental threats. Moreover, the complexity of the environment demands  
136 avenues both for discovery-based science and for developing and testing hypotheses under well  
137 controlled and relevant conditions. Despite the successes mentioned above, the atmosphere is far  
138 from being fully understood with regard to changes in composition and the complex interplay of  
139 chemical and physical processes, and thus predictive capabilities are limited. The importance of  
140 fundamental understanding has been stressed previously,<sup>6</sup> most recently in the recent report on  
141 the Future of Atmospheric Chemistry Research prepared for the National Academy of Science in  
142 the United States.<sup>7</sup>

143 This article highlights the significant role of laboratory-based atmospheric chemistry research,  
144 pointing out that our ability to respond to a changing environment and to accurately inform  
145 policy development hinges upon a fundamental molecular-level understanding of these  
146 processes. Additionally, a commitment from universities, research centers, and governmental  
147 agencies to support such research activities in a meaningful way and to promote innovative and  
148 interdisciplinary research is needed at this time of a rapidly changing atmospheric composition.  
149 Several key challenges and opportunities facing the laboratory community are presented in this  
150 article.

151 **Emerging Topics for Future Laboratory Studies in Atmospheric Chemistry**



152 The atmospheric chemistry community has traditionally relied on laboratory studies to determine  
153 the rates and mechanisms of key gas-phase reactions. For example, these processes have been  
154 shown to be important for better understanding ozone depletion in the stratosphere and the  
155 coupled processes of tropospheric oxidant production and organic oxidation that control the  
156 abundance of key climate forcing agents such as methane (CH<sub>4</sub>) and dimethyl sulfide (DMS).<sup>2,8</sup>  
157 There is currently considerable focus on the chemistry that occurs within or on aerosol particles  
158 and cloud droplets, and increasingly on the interface of the atmosphere with the oceans,  
159 biosphere, cryosphere, and indoor environments.<sup>9-13</sup>

160 Recent advances that highlight how far we have come in this field but, more importantly, how  
161 much further we have to go to develop sufficiently accurate and predictive models of the  
162 atmosphere are presented below. The examples below demonstrate that chemistry is central to  
163 our understanding of the field and that we once thought to be understood, such as volatile  
164 organic carbon (VOC) oxidation mechanisms, is much more varied and complex than described  
165 by current mechanisms and included in models.

166 i. Do we understand how organic molecules are oxidized in the atmosphere?

167 Our conceptual view of organic oxidation mechanisms (see Figure 3), central to our  
168 understanding of smog formation, the lifetime of greenhouse gases such as ozone (O<sub>3</sub>) and  
169 methane, and the climate effects of aerosol particles, has been transformed in the past few years.

170 An example of such a transformation is illustrated by recent studies of the chemistry of key  
171 biogenic hydrocarbons, isoprene and the monoterpenes, that are emitted from vegetation.  
172 Although isoprene is the largest source of organic carbon to the atmosphere and even a  
173 component of exhaled human breath, our understanding of how it is oxidized under appropriate

174 radical concentration conditions is still incomplete.<sup>14</sup> Only a short time ago, it was not known  
175 whether it could contribute to aerosol formation, but recent advances now indicate it is an  
176 important aerosol precursor (Figure 3).<sup>15, 16</sup> In particular, when the OH radical adds to one of  
177 isoprene's carbon-carbon double bonds a variety of highly oxidized products form.<sup>17</sup> The radical  
178 intermediates are reactive, and can isomerize leading to efficient autocatalytic oxidation and  
179 multi-functionalised compounds that may form aerosol particles.<sup>18</sup> Similarly,  $\alpha$ -pinene, a  
180 monoterpene, is converted into "highly oxidized molecules (HOMs)" (or "extremely low  
181 volatility organic compounds", ELVOCs) under atmospheric conditions on timescales of seconds  
182 to minutes, with up to 10 oxygen atoms being introduced onto the terpene backbone.<sup>19</sup> These are  
183 all new findings.

184 Major uncertainties in the oxidation mechanisms of VOCs, particularly biogenic VOCs, still  
185 remain. In particular, there is a need to fully establish how much recycling of the OH radical  
186 occurs from isoprene oxidation under different atmospheric conditions. This is especially true in  
187 environments with significant input of molecules from biogenic sources, where the impact of OH  
188 recycling on oxidant levels may be significant.<sup>20</sup> Also important toward understanding the  
189 impacts of VOC oxidation are the roles of epoxides and the HOM compounds in the formation of  
190 biogenic secondary organic aerosol (SOA), new particles,<sup>21</sup> and the potential health effects of  
191 particles containing these highly oxidized species. Indeed, the recognition that such organics  
192 may play a major role in atmospheric particle nucleation and growth processes nicely illustrates  
193 another connection between fundamental physical chemistry processes and atmospheric  
194 behavior.

195 Little is known about the chemistry of these highly oxidized multifunctional reaction products  
196 with respect to subsequent gas-phase and condensed-phase reactivity, photochemistry and light-

197 absorbing properties, hygroscopicity, and volatility. Further complexity arises through the  
198 addition of nitrogen-based functional groups to the molecule via reactions involving nitrogen  
199 oxide radicals, amines, or ammonia. Light-absorbing compounds can result that contribute to the  
200 colored particles collectively referred to as brown carbon aerosol and known to arise from  
201 burning processes. This has implications for climate change given that biomass burning and  
202 wildfire emission sources are expected to increase in the future.<sup>22</sup>

203 Our framework for understanding the multiphase chemistry of when these oxidized products  
204 partition from the gas phase to cloud water or an aerosol particle is in its infancy. Questions that  
205 arise include: How important are radical processes compared to non-radical  
206 nucleophilic/electrophilic, hydrolytic, and addition/condensation reactions?<sup>23</sup> How important are  
207 non-ideal solution effects and how can they best be treated to help interpret field investigations  
208 and improve representations in models?<sup>24</sup> These issues can be addressed with focused laboratory  
209 studies coming out of the coupled atmospheric-physical-analytical chemistry laboratory  
210 communities.

211 ii. What controls the major oxidants in the atmosphere?

212 The chemistry of reactive chemical intermediates – usually radicals – is at the heart of  
213 atmospheric chemistry (see Figure 4). Although our understanding of reaction mechanisms  
214 initiated by conventional oxidants (OH, Cl, O<sub>3</sub>) is extensive, it is not complete and the  
215 importance of new classes of reactive intermediates, e.g. Criegee intermediates, is now being  
216 recognized and studied. For example, although the rapid cycling of OH and HO<sub>2</sub> radicals helps  
217 to drive the chemistry that forms multifunctional organic compounds, the field still does not yet  
218 have closure between measured and modeled OH concentrations in clean environments.<sup>20, 25</sup>

219 Beyond gas-phase OH, there has been a recent surge of activity in the chemistry of other reactive  
220 intermediates, notably nitrate radicals,<sup>26</sup> halogen oxide radicals, Criegee intermediates, and  
221 compounds active as photosensitizers,<sup>27</sup> but our understanding of these processes remains largely  
222 insufficient. The Criegee intermediates present a compelling story, having recently been  
223 observed for the first time via advanced mass spectrometry techniques.<sup>28, 29</sup> These reactive  
224 species are formed when ozone reacts in the gas phase with carbon-carbon double bonds, and  
225 their involvement in the oxidation of key molecules, such as SO<sub>2</sub>, thus affecting new particle  
226 formation, is now being studied.

227 iii. What is the atmospheric importance of multiphase chemistry?

228 Some of the largest uncertainties in our field arise from the chemistry and photochemistry that  
229 takes place involving different interfaces and media, such as aerosol particles and cloud droplets,  
230 that interact with gas phase constituents. Collectively, this is referred to as multiphase chemistry  
231 (see Figure 5).<sup>30</sup>

232 The organic aerosol medium represents a poorly characterized component of particle chemistry  
233 that can affect partitioning of gas-phase organics and can promote chemistry distinctly different  
234 from that which occurs in other chemical environments. Aqueous phase droplets and particles  
235 can enhance the formation of organic aerosol by promoting soluble organic partitioning and  
236 subsequent reactions.<sup>31</sup> Constraining advances in the field of organic particle and mixed organic-  
237 inorganic particle chemistry is our lack of detailed knowledge of the bulk composition, phase  
238 state, morphology, transport properties, interfacial composition, and photochemistry within these  
239 particles under environmental conditions.<sup>27, 32</sup> The development of new methodologies, such as  
240 spectroscopic and mass spectrometric approaches, that are able to better characterize these

241 properties at the individual particle level is a key challenge. In addition, limitations currently  
242 exist in our ability to prepare appropriately complex model substances in the laboratory that  
243 realistically resemble those present in the environment.

244 Since particles can indirectly modify climate by acting as cloud condensation nuclei (CCN) and  
245 ice nuclei (IN) an important area of multiphase chemistry involves studies of the hygroscopicity  
246 of atmospheric particles.<sup>33</sup> Although much has been learned about the relationship of aerosol  
247 particle composition to hygroscopicity and CCN activity, the roles of interfacial processes and  
248 organic substituents are still being resolved.<sup>34, 35</sup> Our understanding of the IN activity of aerosol  
249 particles is even less refined because we lack a first-principles model that can accurately predict  
250 the rates of heterogeneous ice nucleation as a function of aerosol type and composition.<sup>36</sup>

251 The atmosphere interacts with the surfaces of the oceans, vegetation, soil, and indoor  
252 environments. To illustrate such interactions, a long-standing uncertainty in atmospheric  
253 chemistry has been related to non-gas phase sources of HONO, a photolytic source of the OH  
254 radical. To explain measured levels within many boundary layer environments, it is known that  
255 during the day there is a source that forms HONO much faster than the traditional gas-phase  
256 formation route.<sup>37, 38</sup> A variety of mechanisms have been proposed, all involving interfacial  
257 chemistry.<sup>39, 40, 41, 42</sup>

258 A highly interdisciplinary frontier lies with the impacts of atmospheric particles on human  
259 health. Reactive oxygen species (ROS), such as hydrogen peroxide or organic hydroperoxides,  
260 are a class of molecules that can give rise to oxidative stress, the state where the human body's  
261 oxidant and anti-oxidant balance is disrupted.<sup>43</sup> Also important to oxidative stress are molecules  
262 with labile oxidation states and metals that promote the formation of ROS species, either in

263 inhaled atmospheric particles or the body.<sup>44-46</sup> A key question is the degree to which these  
264 molecules drive oxidative stress in the body and whether their sources are endogenous or  
265 exogenous. Moving ahead in this field requires the collaboration of atmospheric chemists and  
266 toxicologists.

## 267 **Challenges and Opportunities in Laboratory Atmospheric Chemistry Research**

### 268 i. Complexity

269 The atmosphere has many chemical constituents and processes interacting in a non-linear  
270 manner. The complexity that arises in the system has typically been addressed by a reductionist  
271 approach where every relevant rate constant and photochemical property is measured. For  
272 example, this approach is used to predict the non-linear response of urban ozone to changing  
273 NO<sub>x</sub> emissions, a relationship now well-recognized in the air pollution control community and  
274 with connections to climate change.<sup>8,47</sup> But other complex interactions are less well understood.  
275 For example, how do chemical interactions with aerosol particles affect the aerosols' ability to  
276 nucleate liquid water and ice clouds? How does the liquid-like layer that exists at the surface of  
277 snow affect the likelihood of molecules being sorbed from the gas phase?<sup>48</sup> How does biology  
278 affect the indoor microbiome and its ability to change VOC levels?<sup>13</sup> And, what is the interplay  
279 between biology and chemistry in controlling the fluxes of climatically-active gases such as  
280 DMS and isoprene to the atmosphere from the ocean and vegetation.<sup>9,12</sup>

281 A reductionist approach is necessary to arrive at the understanding required for confidence in  
282 model predictions of air quality and climate. Nevertheless, it is also fruitful to work with top-  
283 down methods where observations from laboratory experiments conducted under conditions  
284 close to those in the environment can be parameterized for inclusion in models. For example, it

285 is now possible to measure the rate of loss of a gas-phase reactant to ambient aerosol under real-  
286 world conditions.<sup>49</sup> The goal of such experiments is to obtain quantitative closure between the  
287 bottom-up and top-down approaches for assessing the rates of this chemistry.

## 288 ii. Collaboration and Interdisciplinary Research

289 In addition to the value of single-investigator science, atmospheric chemistry has also long  
290 benefited from collaboration. A number of schemes can enhance opportunities for collaboration.  
291 In particular, there is an advantage to incorporating laboratory projects into the funding of large  
292 field campaigns, to facilitate exchange of ideas and experimental techniques from one  
293 community to another. Another approach is for a small number of research teams to work  
294 together to simultaneously study the chemistry in one facility, to examine the nature of complex  
295 processes.<sup>50</sup> Recent advances in theoretical methods mean that quantum chemical calculations  
296 can significantly enhance knowledge gained from laboratory investigations. In addition,  
297 collaborations between laboratory scientists and atmospheric modelers are required to enable  
298 both direct impact-testing of laboratory data and identify areas of research for which remaining  
299 uncertainty has significant repercussions.

300 New interdisciplinary opportunities are continuing to arise at the interfaces of atmospheric  
301 chemistry. For example, dedicated lab experiments under controlled conditions will inform us  
302 regarding the nature of biosphere-atmosphere interactions.<sup>51</sup> This, in turn, will help us to better  
303 understand how ecosystem health is affected via atmospheric exposures and will aid in  
304 determining the detailed mechanisms by which vegetation removes oxidants from the  
305 atmosphere. Another example is the chemistry involving atmospheric constituents and the lung-  
306 air interface. Experiments conducted jointly by atmospheric chemists and toxicologists will

307 better establish how airborne particles contribute to increased levels of oxidative stress, a  
308 common hypothesis invoked to explain the epidemiological connections between increased  
309 particulate loadings and negative health outcomes.<sup>52</sup> There is also considerable scope for new  
310 explorations in indoor environments where we spend most of our time and receive most of our  
311 pollutant exposure.<sup>13</sup> Indoor surface-area-to-volume ratios are high, suggestive of the importance  
312 of surface chemistry, and many cleaning and personal care products whose constituents partition  
313 between the gas phase and indoor surfaces are prevalent.<sup>53</sup> Furthermore, the desire to reduce  
314 energy consumption in some modern buildings may lead to less ventilated indoor environments,  
315 and requires better understanding of indoor air chemistry to ensure that such green buildings are  
316 also healthy buildings.

317 For all these opportunities, there is the need for science funding agencies to recognize that such  
318 interdisciplinary research often falls between the cracks of funding programs that are better tuned  
319 to promote the value of disciplinary research.

320

321 iii. The interplay of laboratory experiments with computational and atmospheric modeling  
322 scientists

323 Connecting laboratory experiments to chemical theory remains crucial to the atmospheric  
324 chemistry field. At the molecular level, as computational methods allow for increasingly  
325 complex chemistry to be studied on a computer,<sup>54</sup> interactions between laboratory and  
326 computational chemists are expected to become more common.

327 Individual gas-phase rate or photochemical parameters may be easily incorporated into chemical  
328 transport models and their impact evaluated. However, the challenge of transferring laboratory



329 results from complex systems is not as straightforward. The evaluation of physical and chemical  
330 laboratory data as provided by the NASA JPL (<http://jpldataeval.jpl.nasa.gov/>) and IUPAC  
331 (<http://iupac.pole-ether.fr/>) panels is a particularly important task that supports feedback between  
332 modelers and experimentalists in both directions.<sup>55, 56</sup> Collaborative efforts between laboratory  
333 scientists and process-level atmospheric modelers can ensure accurate parameterizations of  
334 complex chemistry are incorporated into models, and the integrating nature of the models can  
335 provide information about which conditions or timescales need further experimental constraints.  
336 Such collaborations can occur as part of research grants, information collaborations, or special  
337 sessions at international conferences that emphasize the integration of modeling and laboratory  
338 results.

#### 339 iv. The interplay of laboratory experiments with field measurements

340 Many advanced analytical technologies have been developed in the past decade that can be  
341 applied to atmospheric chemistry research, with increasingly sensitive and multiplexing  
342 instruments deployed in both the lab and atmosphere. In order to understand new field  
343 observations the laboratory community is pushed to develop a better understanding of the  
344 associated chemistry and its possible environmental impacts. For example, recent progress has  
345 arisen from the advent of advanced mass-spectrometric techniques that now allow for the  
346 identification and quantification of species present in complex chemical mixtures.<sup>57, 58</sup> Such  
347 instrumentation has been used extensively in new studies of particle nucleation and growth  
348 processes.<sup>21</sup> With a strong demand for sensitive and reliable measurement technologies, the  
349 atmospheric chemistry field fosters developments in advanced measurement technologies. These  
350 advances have led to a recent growth in experimental studies where the relevant chemistry is  
351 studied on genuine atmospheric materials (such as atmospheric aerosol particles) commonly

352 referred to as “field-in-the-lab” or “lab-in-the-field” methods.<sup>49</sup> These approaches are attractive  
353 to test the applicability of laboratory model materials and to provide top-down results that  
354 fundamental-based bottom-up approaches aspire to match.

### 355 **Conclusions**

356 The scientific uncertainties illustrated above are associated with some of the most central  
357 questions in atmospheric chemistry: How are atmospheric molecules transformed, and by what  
358 mechanisms? In what phase does this chemistry occur and on what timescale? How does this  
359 chemistry affect air quality and climate? How do ecosystems affect atmospheric chemistry and  
360 vice versa? How does this chemistry affect the interaction of the atmosphere with other parts of  
361 the environment? These issues and others highlight the ongoing central role for laboratory  
362 studies and a molecular-level understanding of atmospheric chemistry that enable the  
363 development of informed environmental policy.

364 The laboratory studies community is adapting to address far greater scientific complexity than  
365 was apparent only a decade ago, needing to embrace an interdisciplinary and collaborative  
366 research approach while at the same time continuing to focus on the measurement of  
367 fundamental properties of atmospherically relevant molecules and processes. These aspects of  
368 the field are highly attractive to early career scientists who are looking for research experiences  
369 outside of traditional disciplines. Funding agencies can facilitate these efforts, by supporting  
370 fundamental laboratory science and by promoting interdisciplinary and collaborative research  
371 and the interplay between laboratory studies and associated modeling, and field measurement  
372 activities.

373

374 **Acknowledgments**

375 This article arose from discussions at a workshop sponsored by the International Global  
376 Atmospheric Chemistry (IGAC) project (<http://www.igacproject.org>) on “The Future of  
377 Laboratory Studies in Atmospheric Chemistry”. As well, we thank NOAA for hosting the event.  
378 We thank A. Reiser and D.K. Papanastasiou for help with the figures, and B. Christensen with  
379 help during the submission process.

380

381 **References**

- 382 1. EPA, Environmental Protection Agency (US), <https://www.epa.gov/acidrain>.
- 383 2. WMO, World Meteorological Organization, Scientific Assessment of Ozone Depletion: 2014.
- 384 *Global Ozone Research and Monitoring Project 2014, Report 55*, 55 pp., Geneva, Switzerland.
- 385 3. IPCC, Climate Change 2013: The Physical Science Basis. *Contribution of Working Group 1 to the*
- 386 *Fifth Assessment Report of the Intergovernmental Panel on Climate Change, Stocker et al. (eds) 2013,*
- 387 *Cambridge University Press, Cambridge, UK and New York, USA.*
- 388 4. Vaughan, N. E.; Lenton, T. M., A review of climate geoengineering proposals. *Climatic Change*
- 389 **2011**, *109*, 745-790.
- 390 5. Liggio, J.; Li, S. M.; Hayden, K.; Taha, Y. M.; Stroud, C.; Darlington, A.; Drollette, B. D.; Gordon,
- 391 M.; Lee, P.; Liu, P., et al., Oil sands operations as a large source of secondary organic aerosols. *Nature*
- 392 **2016**, *534*, 91-95.
- 393 6. Abbatt, J.; George, C.; Melamed, M.; Monks, P.; Pandis, S.; Rudich, Y., New Directions:
- 394 Fundamentals of atmospheric chemistry: Keeping a three-legged stool balanced. *Atmos. Environ.* **2014**,
- 395 *84*, 390-391.
- 396 7. National Academies of Sciences, Engineering, and Medicine. 2016. The Future of Atmospheric
- 397 Chemistry Research: Remembering Yesterday, Understanding Today, Anticipating Tomorrow.
- 398 Washington, DC: The National Academies Press. DOI: 10.17226/235730.
- 399 8. Monks, P. S.; Archibald, A. T.; Colette, A.; Cooper, O.; Coyle, M.; Derwent, R.; Fowler, D.; Granier,
- 400 C.; Law, K. S.; Mills, G. E., et al., Tropospheric ozone and its precursors from the urban to the global scale
- 401 from air quality to short-lived climate forcer. *Atmos. Chem. Phys.* **2015**, *15*, 8889-8973.
- 402 9. Carpenter, L. J.; Nightingale, P. D., Chemistry and Release of Gases from the Surface Ocean.
- 403 *Chem. Rev.* **2015**, *115*, 4015-4034.
- 404 10. Abbatt, J. P. D.; Thomas, J. L.; Abrahamsson, K.; Boxe, C.; Granfors, A.; Jones, A. E.; King, M. D.;
- 405 Saiz-Lopez, A.; Shepson, P. B.; Sodeau, J., et al., Halogen activation via interactions with environmental
- 406 ice and snow in the polar lower troposphere and other regions. *Atmos. Chem. Phys.* **2012**, *12*, 6237-
- 407 6271.
- 408 11. McNeill, V. F.; Grannas, A. M.; Abbatt, J. P. D.; Ammann, M.; Ariya, P.; Bartels-Rausch, T.;
- 409 Domine, F.; Donaldson, D. J.; Guzman, M. I.; Heger, D., et al., Organics in environmental ices: sources,
- 410 chemistry, and impacts. *Atmos. Chem. Phys.* **2012**, *12*, 9653-9678.
- 411 12. Heald, C. L.; Spracklen, D. V., Land Use Change Impacts on Air Quality and Climate. *Chem. Rev.*
- 412 **2015**, *115*, 4476-4496.
- 413 13. Weschler, C. J., Chemistry in indoor environments: 20 years of research. *Indoor Air* **2011**, *21*,
- 414 205-218.
- 415 14. Wennberg, P., Let's abandon the high NO<sub>x</sub> and low NO<sub>x</sub> terminology *IGAC Newsletter* **2013**, *50*,
- 416 3-4.
- 417 15. Surratt, J. D.; Chan, A. W. H.; Eddingsaas, N. C.; Chan, M. N.; Loza, C. L.; Kwan, A. J.; Hersey, S. P.;
- 418 Flagan, R. C.; Wennberg, P. O.; Seinfeld, J. H., Reactive intermediates revealed in secondary organic
- 419 aerosol formation from isoprene. *Proc. Nat. Acad. Sci. U.S.A.* **2010**, *107*, 6640-6645.
- 420 16. Lin, Y. H.; Zhang, Z. F.; Docherty, K. S.; Zhang, H. F.; Budisulistiorini, S. H.; Rubitschun, C. L.; Shaw,
- 421 S. L.; Knipping, E. M.; Edgerton, E. S.; Kleindienst, T. E., et al., Isoprene Epoxydiols as Precursors to
- 422 Secondary Organic Aerosol Formation: Acid-Catalyzed Reactive Uptake Studies with Authentic
- 423 Compounds. *Environ. Sci. Technol.* **2012**, *46*, 250-258.
- 424 17. Paulot, F.; Crouse, J. D.; Kjaergaard, H. G.; Kurten, A.; St Clair, J. M.; Seinfeld, J. H.; Wennberg,
- 425 P. O., Unexpected Epoxide Formation in the Gas-Phase Photooxidation of Isoprene. *Science* **2009**, *325*,
- 426 730-733.

427 18. Crouse, J. D.; Nielsen, L. B.; Jorgensen, S.; Kjaergaard, H. G.; Wennberg, P. O., Autoxidation of  
428 Organic Compounds in the Atmosphere. *J. Phys. Chem. Lett.* **2013**, *4*, 3513-3520.

429 19. Ehn, M.; Thornton, J. A.; Kleist, E.; Sipila, M.; Junninen, H.; Pullinen, I.; Springer, M.; Rubach, F.;  
430 Tillmann, R.; Lee, B., et al., A large source of low-volatility secondary organic aerosol. *Nature* **2014**, *506*,  
431 476-480.

432 20. Stone, D.; Whalley, L. K.; Heard, D. E., Tropospheric OH and HO<sub>2</sub> radicals: field measurements  
433 and model comparisons. *Chem. Soc. Rev.* **2012**, *41*, 6348-6404.

434 21. Trostl, J.; Chuang, W. K.; Gordon, H.; Heinritzi, M.; Yan, C.; Molteni, U.; Ahlm, L.; Frege, C.;  
435 Bianchi, F.; Wagner, R., et al., The role of low-volatility organic compounds in initial particle growth in  
436 the atmosphere. *Nature* **2016**, *533*, 527-530.

437 22. Laskin, A.; Laskin, J.; Nizkorodov, S. A., Chemistry of Atmospheric Brown Carbon. *Chem. Rev.*  
438 **2015**, *115*, 4335-4382.

439 23. Jang, M. S.; Czoschke, N. M.; Lee, S.; Kamens, R. M., Heterogeneous atmospheric aerosol  
440 production by acid-catalyzed particle-phase reactions. *Science* **2002**, *298*, 814-817.

441 24. Herrmann, H.; Schaefer, T.; Tilgner, A.; Styler, S. A.; Weller, C.; Teich, M.; Otto, T., Tropospheric  
442 Aqueous-Phase Chemistry: Kinetics, Mechanisms, and Its Coupling to a Changing Gas Phase. *Chem. Rev.*  
443 **2015**, *115*, 4259-4334.

444 25. Rohrer, F.; Lu, K. D.; Hofzumahaus, A.; Bohn, B.; Brauers, T.; Chang, C. C.; Fuchs, H.; Haseler, R.;  
445 Holland, F.; Hu, M., et al., Maximum efficiency in the hydroxyl-radical-based self-cleansing of the  
446 troposphere. *Nat. Geosci.* **2014**, *7*, 559-563.

447 26. Ng, N. L.; Brown, S. S.; Archibald, A. T.; Atlas, E.; R.C., C.; Crowley, J. N.; Day, D. A.; Donahue, N.  
448 M.; Fry, J. L.; al., F., Nitrate radicals and biogenic volatile organic compounds: oxidation, mechanisms  
449 and organic aerosol. *Atmos. Chem. Phys. Discuss.* **2016**, doi:10.5194/acp-2016-734.

450 27. George, C.; Ammann, M.; D'Anna, B.; Donaldson, D. J.; Nizkorodov, S. A., Heterogeneous  
451 Photochemistry in the Atmosphere. *Chem. Rev.* **2015**, *115*, 4218-4258.

452 28. Taatjes, C. A.; Shallcross, D. E.; Percival, C. J., Research frontiers in the chemistry of Criegee  
453 intermediates and tropospheric ozonolysis. *Phys. Chem. Chem. Phys.* **2014**, *16*, 1704-1718.

454 29. Welz, O.; Savee, J. D.; Osborn, D. L.; Vasu, S. S.; Percival, C. J.; Shallcross, D. E.; Taatjes, C. A.,  
455 Direct Kinetic Measurements of Criegee Intermediate (CH<sub>2</sub>OO) Formed by Reaction of CH<sub>2</sub>I with O<sub>2</sub>.  
456 *Science* **2012**, *335*, 204-207.

457 30. Pöschl, U.; Shiraiwa, M., Multiphase Chemistry at the Atmosphere-Biosphere Interface  
458 Influencing Climate and Public Health in the Anthropocene. *Chem. Rev.* **2015**, *115*, 4440-4475.

459 31. Ervens, B.; Turpin, B. J.; Weber, R. J., Secondary organic aerosol formation in cloud droplets and  
460 aqueous particles (aqSOA): a review of laboratory, field and model studies. *Atmos. Chem. Phys.* **2011**,  
461 *11*, 11069-11102.

462 32. Koop, T.; Bookhold, J.; Shiraiwa, M.; Pöschl, U., Glass transition and phase state of organic  
463 compounds: dependency on molecular properties and implications for secondary organic aerosols in the  
464 atmosphere. *Phys. Chem. Chem. Phys.* **2011**, *13*, 19238-19255.

465 33. Petters, M. D.; Kreidenweis, S. M., A single parameter representation of hygroscopic growth and  
466 cloud condensation nucleus activity. *Atmos. Chem. Phys.* **2007**, *7*, 1961-1971.

467 34. Ruehl, C. R.; Davies, J. F.; Wilson, K. R., An interfacial mechanism for cloud droplet formation on  
468 organic aerosols. *Science* **2016**, *351*, 1447-1450.

469 35. Nozière, B.; Baduel, C.; Jaffrezo, J. L., The dynamic surface tension of atmospheric aerosol  
470 surfactants reveals new aspects of cloud activation. *Nat. Commun.* **2014**, *5*, 7-12.

471 36. Hoose, C.; Mohler, O., Heterogeneous ice nucleation on atmospheric aerosols: a review of  
472 results from laboratory experiments. *Atmos. Chem. Phys.* **2012**, *12*, 9817-9854.

473 37. Kleffmann, J., Daytime sources of nitrous acid (HONO) in the atmospheric boundary layer.  
474 *ChemPhysChem* **2007**, *8*, 1137-1144.

475 38. Li, X.; Rohrer, F.; Hofzumahaus, A.; Brauers, T.; Haseler, R.; Bohn, B.; Broch, S.; Fuchs, H.; Gomm,  
476 S.; Holland, F., et al., Missing Gas-Phase Source of HONO Inferred from Zeppelin Measurements in the  
477 Troposphere. *Science* **2014**, *344*, 292-296.

478 39. George, C.; Strekowski, R. S.; Kleffmann, J.; Stemmler, K.; Ammann, M., Photoenhanced uptake  
479 of gaseous NO<sub>2</sub> on solid-organic compounds: a photochemical source of HONO? *Faraday Discuss.* **2005**,  
480 *130*, 195-210.

481 40. Ye, C. X.; Zhou, X. L.; Pu, D.; Stutz, J.; Festa, J.; Spolaor, M.; Tsai, C.; Cantrell, C.; Mauldin, R. L.;  
482 Campos, T., et al., Rapid cycling of reactive nitrogen in the marine boundary layer. *Nature* **2016**, *532*,  
483 489-491.

484 41. VandenBoer, T. C.; Young, C. J.; Talukdar, R. K.; Markovic, M. Z.; Brown, S. S.; Roberts, J. M.;  
485 Murphy, J. G., Nocturnal loss and daytime source of nitrous acid through reactive uptake and  
486 displacement. *Nat. Geosci.* **2015**, *8*, 55-60.

487 42. Donaldson, M. A.; Bish, D. L.; Raff, J. D., Soil surface acidity plays a determining role in the  
488 atmospheric-terrestrial exchange of nitrous acid. *Proc. Nat. Acad. Sci. U.S.A.* **2014**, *111*, 18472-18477.

489 43. Lushchak, V. I., Free radicals, reactive oxygen species, oxidative stress and its classification.  
490 *Chem.-Biol. Interact.* **2014**, *224*, 164-175.

491 44. Charrier, J. G.; Anastasio, C., On dithiothreitol (DTT) as a measure of oxidative potential for  
492 ambient particles: evidence for the importance of soluble transition metals. *Atmos. Chem. Phys.* **2012**,  
493 *12*, 9321-9333.

494 45. Squadrito, G. L.; Cueto, R.; Dellinger, B.; Pryor, W. A., Quinoid redox cycling as a mechanism for  
495 sustained free radical generation by inhaled airborne particulate matter. *Free Radic. Biol. Med.* **2001**, *31*,  
496 1132-1138.

497 46. Shuster-Meiseles, T.; Shafer, M. M.; Heo, J. E.; Pardo-Levin, M.; Antkiewicz, D. S.; Schauer, J. J.;  
498 Rudich, A.; Rudich, Y., ROS-generating/ARE-activating capacity of metals in roadway particulate matter  
499 deposited in urban environment. *Environ. Res.* **2016**, *146*, 252-262.

500 47. von Schneidmesser, E.; Monks, P. S.; Allan, J. D.; Bruhwiler, L.; Forster, P.; Fowler, D.; Lauer, A.;  
501 Morgan, W. T.; Paasonen, P.; Righi, M., et al., Chemistry and the Linkages between Air Quality and  
502 Climate Change. *Chem. Rev.* **2015**, *115*, 3856-3897.

503 48. Abbatt, J. P. D., Interactions of atmospheric trace gases with ice surfaces: Adsorption and  
504 reaction. *Chem. Rev.* **2003**, *103*, 4783-4800.

505 49. Bertram, T. H.; Thornton, J. A.; Riedel, T. P.; Middlebrook, A. M.; Bahreini, R.; Bates, T. S.; Quinn,  
506 P. K.; Coffman, D. J., Direct observations of N<sub>2</sub>O<sub>5</sub> reactivity on ambient aerosol particles. *Geophys. Res.*  
507 *Lett.* **2009**, *36*.

508 50. Nguyen, T. B.; Crouse, J. D.; Schwantes, R. H.; Teng, A. P.; Bates, K. H.; Zhang, X.; St Clair, J. M.;  
509 Brune, W. H.; Tyndall, G. S.; Keutsch, F. N., et al., Overview of the Focused Isoprene eXperiment at the  
510 California Institute of Technology (FIXCIT): mechanistic chamber studies on the oxidation of biogenic  
511 compounds. *Atmos. Chem. Phys.* **2014**, *14*, 13531-13549.

512 51. Mentel, T. F.; Wildt, J.; Kiendler-Scharr, A.; Kleist, E.; Tillmann, R.; Dal Maso, M.; Fisseha, R.;  
513 Hohaus, T.; Spahn, H.; Uerlings, R., et al., Photochemical production of aerosols from real plant  
514 emissions. *Atmos. Chem. Phys.* **2009**, *9*, 4387-4406.

515 52. West, J. J.; Cohen, A.; Dentener, F.; Brunekreef, B.; Zhu, T.; Armstrong, B.; Bell, M. L.; Brauer, M.;  
516 Carmichael, G.; Costa, D. L., et al., "What We Breathe Impacts Our Health: Improving Understanding of  
517 the Link between Air Pollution and Health". *Environ. Sci. Technol.* **2016**, *50*, 4895-4904.

518 53. Morrison, G., Interfacial chemistry in indoor environments. *Environ. Sci. Technol.* **2008**, *42*, 3494-  
519 3499.

520 54. Vereecken, L.; Glowacki, D. R.; Pilling, M. J., Theoretical Chemical Kinetics in Tropospheric  
521 Chemistry: Methodologies and Applications. *Chem. Rev.* **2015**, *115*, 4063-4114.

522 55. Cox, R. A., Evaluation of laboratory kinetics and photochemical data for atmospheric chemistry  
523 applications. *Chem. Soc. Rev.* **2012**, *41*, 6231-6246.

524 56. Ammann, M.; Cox, R. A.; Crowley, J. N.; Jenkin, M. E.; Mellouki, A.; Rossi, M. J.; Troe, J.;  
525 Wallington, T. J., Evaluated kinetic and photochemical data for atmospheric chemistry: Volume VI -  
526 heterogeneous reactions with liquid substrates. *Atmos. Chem. Phys.* **2013**, *13*, 8045-8228.

527 57. Prather, K. A.; Hatch, C. D.; Grassian, V. H., Analysis of Atmospheric Aerosols. *Annu. Rev. Anal.*  
528 *Chem.* **2008**, *1*, 485-514.

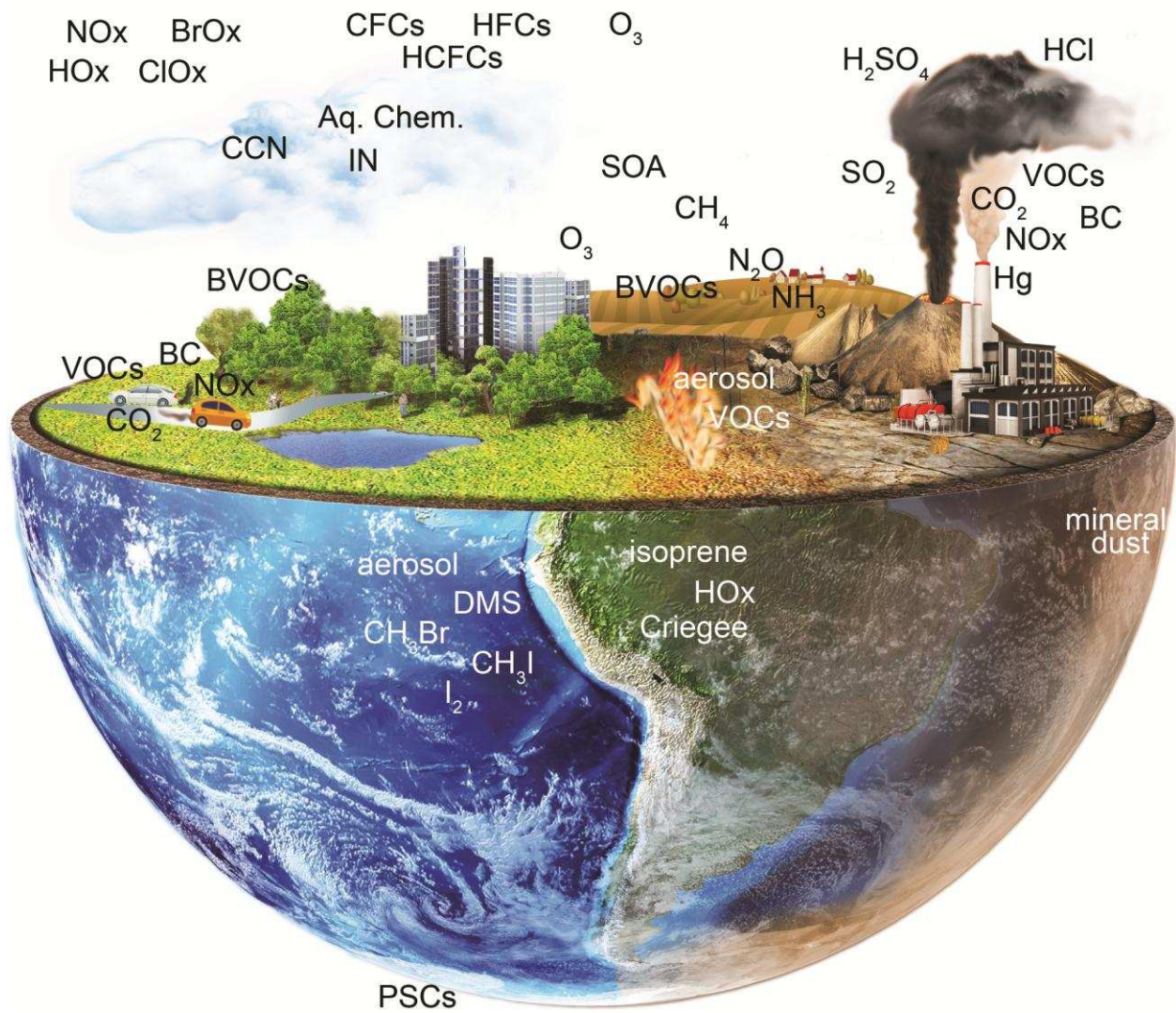
529 58. Lopez-Hilfiker, F. D.; Mohr, C.; Ehn, M.; Rubach, F.; Kleist, E.; Wildt, J.; Mentel, T. F.; Lutz, A.;  
530 Hallquist, M.; Worsnop, D., et al., A novel method for online analysis of gas and particle composition:  
531 description and evaluation of a Filter Inlet for Gases and AEROSols (FIGAERO). *Atmos. Meas. Tech.* **2014**,  
532 *7*, 983-1001.

533

534

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536



538

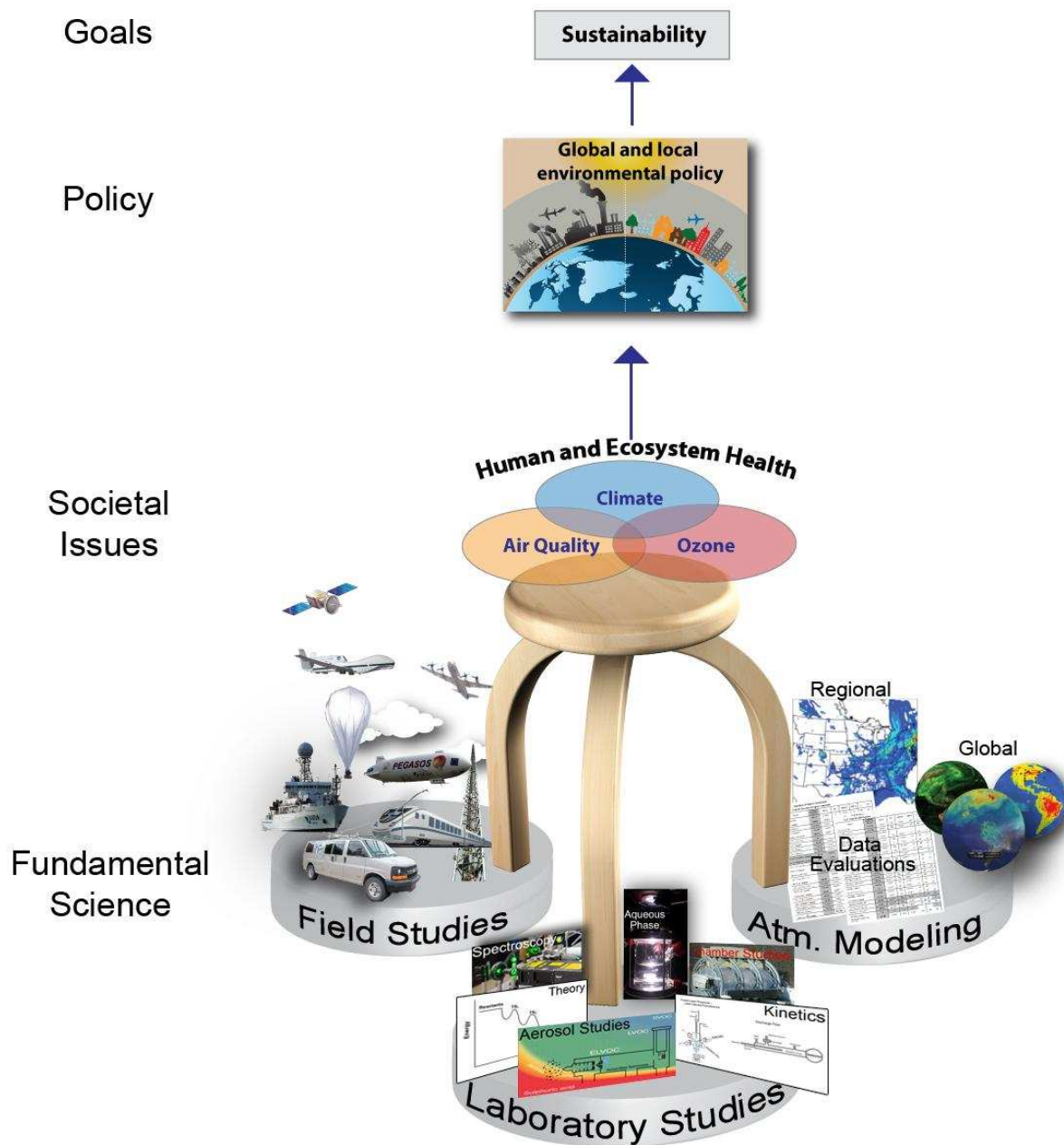
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540 **Figure 1:** A chemical view of the atmosphere highlighting biogenic and anthropogenic  
 541 emissions sources and key atmospheric species.

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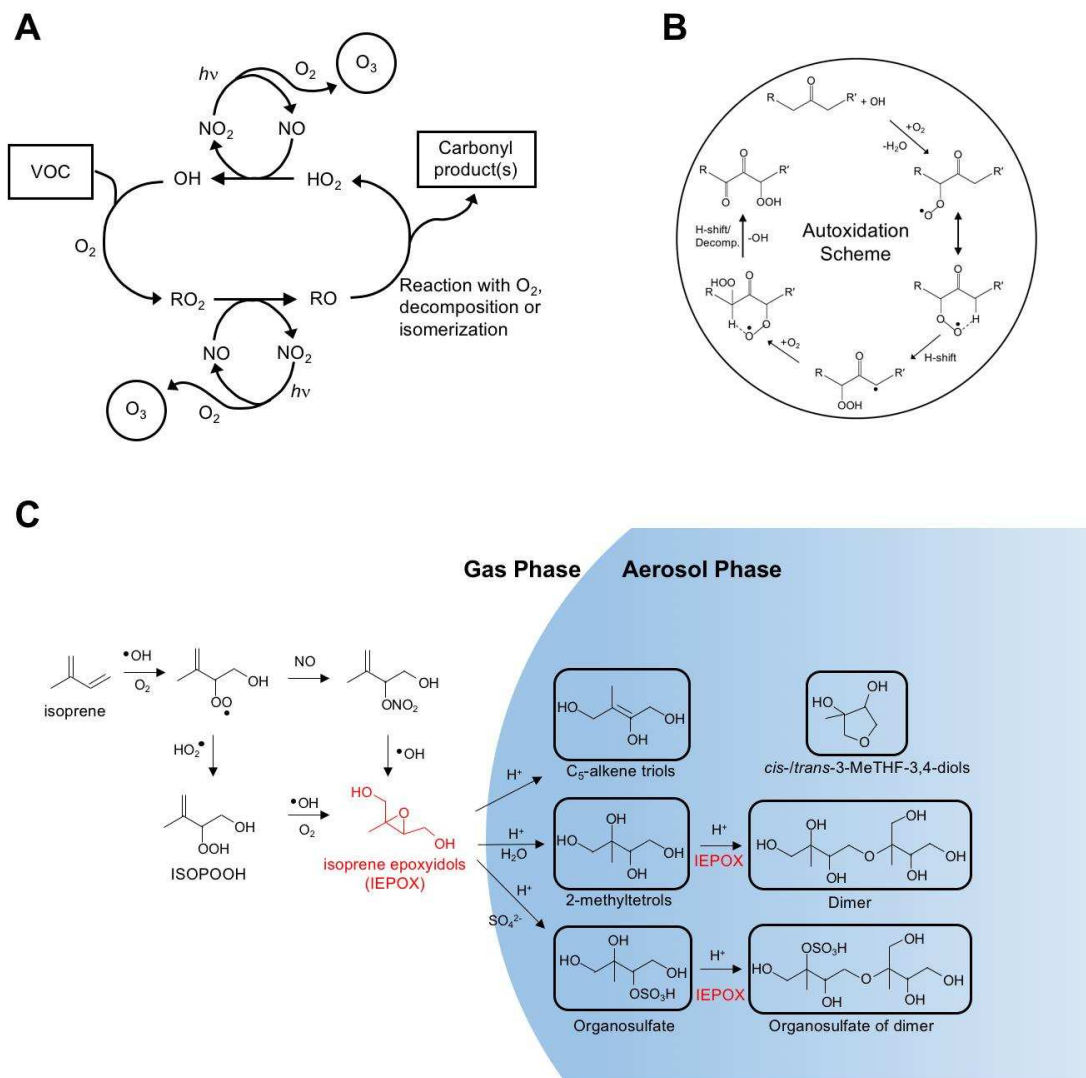




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546 **Figure 2:** The three-legged stool connecting atmospheric chemistry to sustainable policy.

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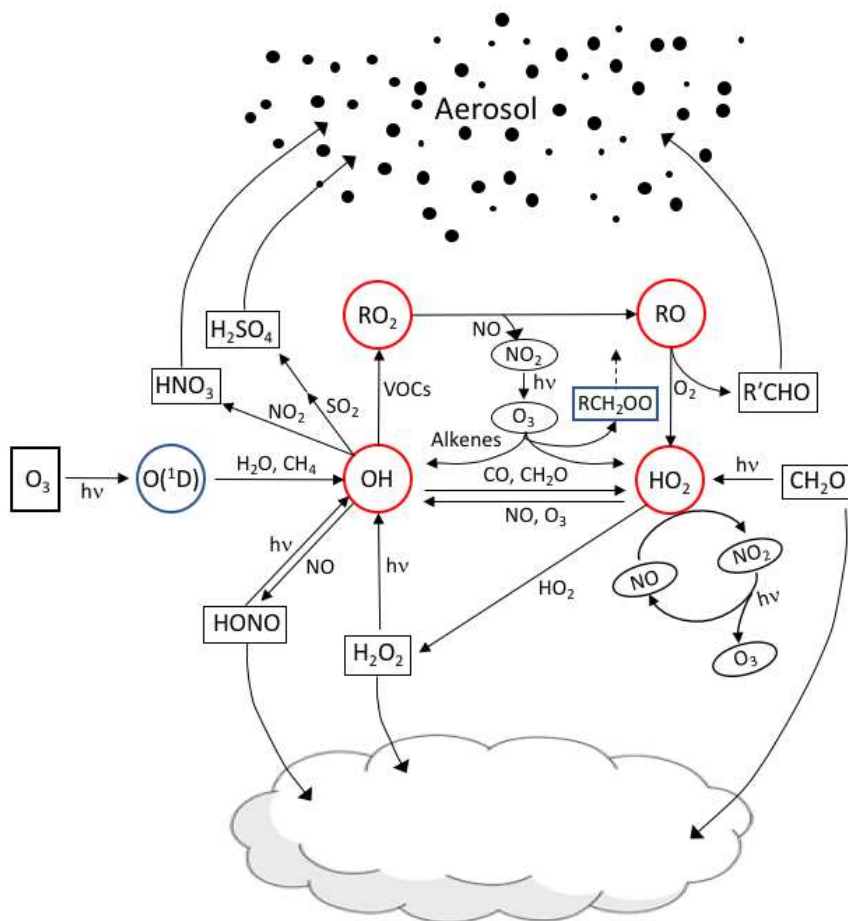


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550 **Figure 3:** Examples of volatile organic compound (VOC) oxidation mechanisms. A.  
 551 Formation of ozone coupled to VOC oxidation in the presence of  $\text{NO}_x$ , B. VOC  
 552 autoxidation scheme adapted from Crouse et al.,<sup>18</sup> C. Isoprene oxidation mechanism  
 553 emphasizing the interplay between chemistry occurring in both the gas and condensed  
 554 phases.

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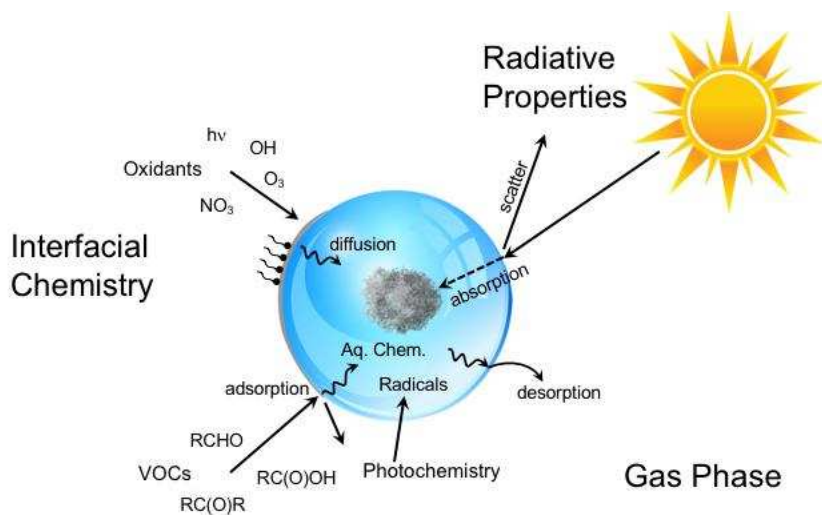
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559 **Figure 4:** Examples of radical formation, cycling, formation of stable products, and  
560 contributions to aerosol and cloud chemistry. Note that radical chemistry also occurs in cloud  
561 droplets and aerosol particles but is not shown.

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567 **Figure 5:** Representation of multiphase processes and radiative properties for a particle or  
568 droplet containing a solid core. Note that other particle morphologies are possible.

569