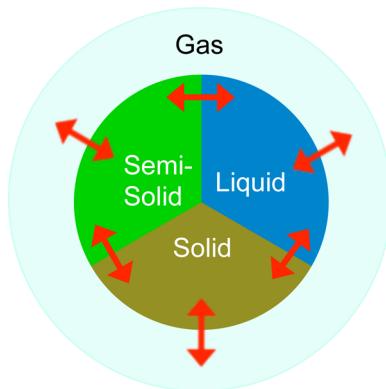


## Multiphase Chemistry at the Atmosphere–Biosphere Interface Influencing Climate and Public Health in the Anthropocene

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### 1. INTRODUCTION AND MOTIVATION

Multiphase chemistry plays a vital role in the Earth system, climate, and health. Chemical reactions, mass transport, and phase transitions between gases, liquids, and solids are essential for the interaction and coevolution of life and climate. Knowledge of the mechanisms and kinetics of these processes is also required to address societally relevant questions of global environmental change and public health in the Anthropocene, that is, in the present era of globally pervasive and steeply increasing human influence on planet Earth.<sup>1</sup> In this work, we review the current scientific understanding and recent advances in the investigation of short-lived health- and climate-relevant

air contaminants (SHCC) and their multiphase chemical interactions at the atmosphere–biosphere interface, including human lungs and skin, plant leaves, cryptogamic covers, soil, and aquatic surfaces. After an overview of different groups of SHCC, we address the chemical interactions of reactive oxygen species and reactive nitrogen species (ROS, RNS), primary biological and secondary organic aerosols (PBA, SOA), as well as carbonaceous combustion aerosols (CCA) including soot, black/elemental carbon, polycyclic aromatic hydrocarbons, and related compounds (PAH, PAC). ROS and RNS interact strongly with other SHCC and are central to both atmospheric and physiological processes and their coupling through the atmosphere–biosphere interface, for example, in the formation and aging of biogenic and combustion aerosols as well as in inflammatory and allergic immune responses triggered by air pollution. Deposition of atmospheric ROS/RNS and aerosols can damage biological tissues, modify surface microbiomes, and induce oxidative stress through Fenton-like reactions and immune responses. The chemical mechanisms and kinetics are not yet fully elucidated, but the available evidence suggests that multiphase processes are crucial for the assessment, prediction, and handling of air quality, climate, and public health. Caution should be taken to avoid that human activities shaping the Anthropocene create a hazardous or pathogenic atmosphere overloaded with allergenic, corrosive, toxic, or infectious contaminants.

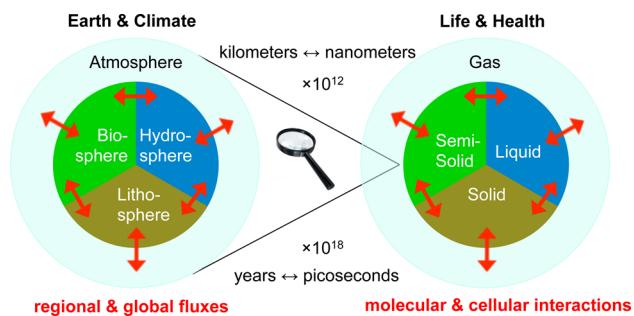
Multiphase chemistry deals with chemical reactions, transport processes, and transformations between gaseous, liquid, and solid matter. These processes are essential for Earth system science and climate research as well as for life and health sciences on molecular and global levels, bridging a wide range of spatial and temporal scales from below nanometers to thousands of kilometers and from less than nanoseconds to years and millennia as illustrated in Figure 1.

From a chemical perspective, life and the metabolism of most living organisms can be regarded as multiphase processes involving gases like oxygen and carbon dioxide; liquids like water, blood, lymph, and plant sap; and solid or semisolid substances like bone, tissue, skin, wood, and cellular membranes. Even primitive forms of life and metabolic activity under anaerobic conditions generally involve multiple liquid and solid or semisolid phases structured by cells, organelles, and membranes.<sup>2</sup> On global scales, the biogeochemical cycling of chemical compounds and elements, which can be regarded as the metabolism of planet Earth, also involves chemical reactions, mass transport, and phase transitions within and

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**Figure 1.** Multiphase processes affecting the Earth system and climate as well as life and public health on regional and global scales.

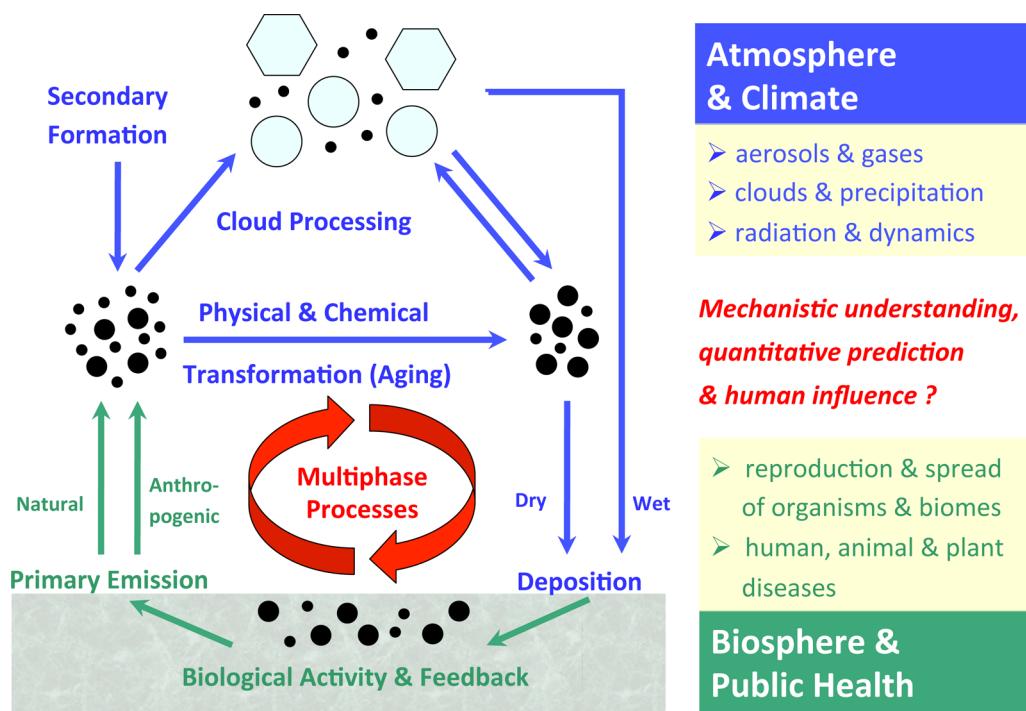
between the atmosphere, biosphere, hydrosphere, and pedosphere/lithosphere.<sup>3</sup>

Particularly important for the coevolution of life and climate on Earth are the exchange and cycling of gases and particles between the biosphere and the atmosphere, which consists largely of biogenic substances, including O<sub>2</sub> and volatile organic compounds as well as primary biological and secondary organic aerosol particles as detailed below. The emission and deposition processes of gases and particles at the atmosphere–biosphere interface as well as the interactions among atmospheric gases, aerosols, clouds, and precipitation are multiphase processes involving air and various liquid, solid, or semisolid condensed phases.<sup>4</sup>

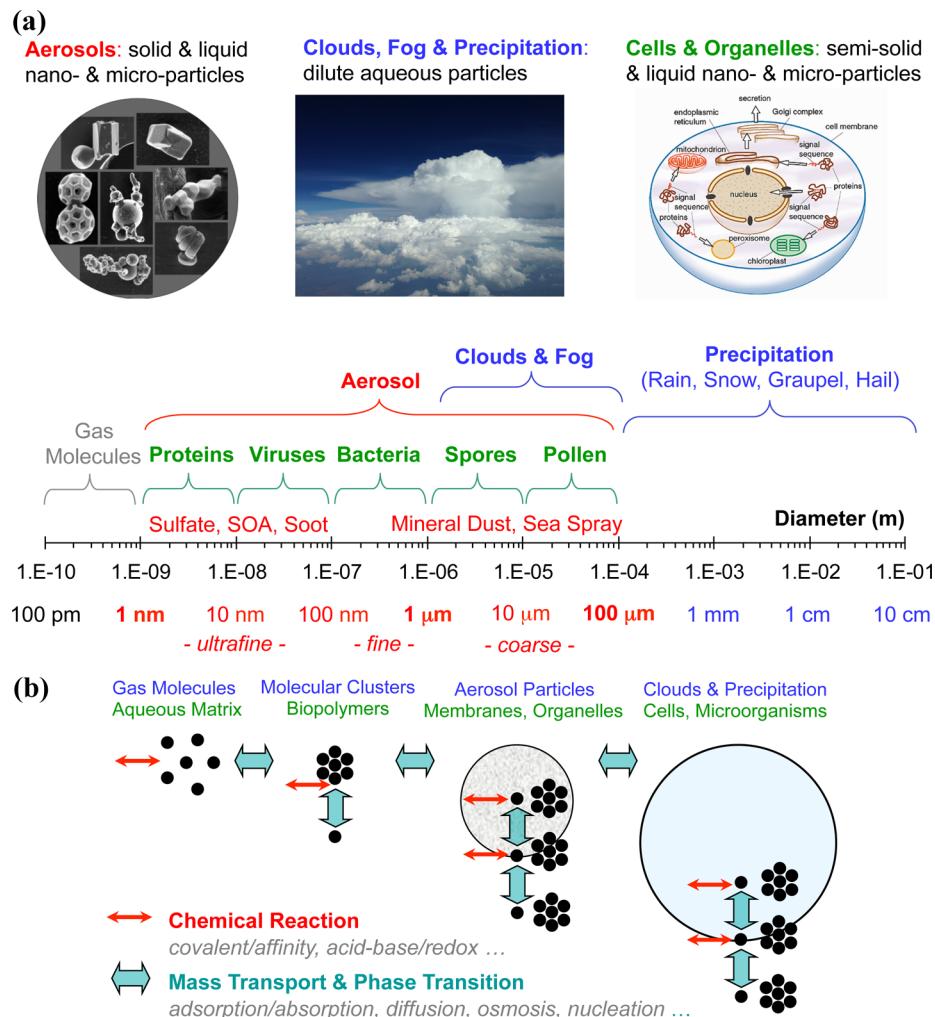
Figure 2 illustrates the cycling and effects of gas molecules and aerosol particles exchanged between the atmosphere and the biosphere. It involves primary emission from natural and anthropogenic sources as well as secondary formation via oxidation of precursors in the atmosphere, whereby the definition of primary and secondary may depend on the temporal and spatial scale of investigation (e.g., the resolution of thermal and chemical processes in exhaust plumes, street

canyons, or vegetation canopies).<sup>5</sup> In the atmosphere, gases and aerosols undergo chemical and physical transformation (“aging”), which includes the interaction with solar radiation and atmospheric oxidants as well as cloud droplets and ice crystals (cloud processing). Recent studies have shown that light can substantially enhance the rate of multiphase chemical processes.<sup>6</sup> Most clouds re-evaporate and release modified aerosols and gases, but when they form precipitation that reaches the Earth’s surface, not only cloud condensation and ice nuclei (CCN/IN) but also other particles and gases are scavenged on the way to the surface and removed from the atmosphere. This process of “wet deposition” is the major sink for atmospheric aerosols; “dry deposition” by convective transport, sedimentation, or diffusion tends to be less important on global scales. Nevertheless, dry deposition on biological surfaces, including human lungs and skin, is highly relevant with respect to local air quality and health effects. After deposition, viable bioparticles can trigger and other particles can interfere with metabolic activity and biological reproduction or disease. These effects may stimulate or suppress further emissions, thus closing a biogeochemical cycle and related feedback loops in the Earth system. The multiphase processes involved in the cycling of gases, aerosols, clouds, and precipitation are important for the evolution, current state, and future development of the atmosphere and climate as well as the biosphere and public health.<sup>4d–f,h,o,7</sup>

Figure 3 provides an overview of the characteristic size range and composition of atmospheric and biological particles as well as the major types and analogies of multiphase chemical processes in the atmosphere and in the biosphere. Aerosol particles are ubiquitous in the atmosphere and affect climate by scattering or absorption of light and serving as nuclei for cloud droplets, ice crystals, and precipitation. The actual abundance, sources, and properties of aerosols, however, are highly variable in space and time, and the quantification of natural and



**Figure 2.** Atmosphere–biosphere exchange and multiphase processing of atmospheric particles and gases [adapted from Fuzzi et al. (2006)<sup>5</sup>].



**Figure 3.** Atmospheric and biological particles. (a) Size range of aerosols, hydrometeors, cells, and organelles. Aerosol picture reprinted with permission from ref 4h. Copyright 2005 John Wiley. Cloud picture reprinted with permission. Copyright 2014 S. Gemsa. Cell structure reprinted with permission from <http://www.nobelprize.org/>. Copyright 1999 the Nobel Assembly at Karolinska Institute. (b) Multiphase chemical transformations in the course of aerosol–cloud–precipitation interactions as well as cellular metabolism and signaling in the biosphere [adapted from Fuzzi et al. (2006)<sup>5</sup>].

anthropogenic aerosol effects on atmospheric processes and climate is among the largest uncertainties in the prediction of future climate change.<sup>4e,h,o,5,7a,c,e,g,8</sup> Moreover, the effects of air particulate matter on public health are major concerns in air quality research and management, but the drivers and mechanisms of these effects are not fully understood.<sup>4d,h,9</sup>

Inhalation and deposition of aerosol particles and atmospheric oxidants can cause oxidative stress and damage to cellular proteins, lipids, and DNA as detailed in the following sections of this Review. Epidemiological studies have shown a clear correlation between air pollutants and adverse health effects including cardiovascular, respiratory, and allergic diseases.<sup>10</sup> Ambient and indoor air pollution by air particulate matter and ozone are among the most prominent leading risk factors for the global burden of disease.<sup>11</sup> Recent model studies with health impact functions suggest global premature mortality rates of about 186 000 yr<sup>-1</sup> from lung cancer, 773 000 yr<sup>-1</sup> from other respiratory diseases, and 2 million yr<sup>-1</sup> from cardiovascular diseases caused by exposure to fine particulate matter (PM2.5) and ozone.<sup>12</sup>

Numerous studies indicate that anthropogenic air pollution has led to a massive increase of aerosol and oxidant

concentrations in the lower atmosphere, that is, at the atmosphere–biosphere interface. For example, the average mixing ratios of ozone in continental background air have increased by factors of 2–4 from around 10–20 ppb from the beginning of the 19th century to 30–40 ppb in the 21st century,<sup>13</sup> and the number and mass concentrations of aerosol particles in polluted urban air are typically by 1–2 orders of magnitude higher than in pristine air of remote continental regions ( $\sim 10^2$ – $10^3$  cm<sup>-3</sup> and  $\sim 1$ – $10$   $\mu$ g m<sup>-3</sup> vs  $\sim 10^3$ – $10^5$  cm<sup>-3</sup> and  $\sim 10$ – $100$   $\mu$ g m<sup>-3</sup>).<sup>4d,f,8g,12,14</sup> The strong increase of these and many other air contaminants like greenhouse gases and reactive nitrogen species caused by human activity is a characteristic feature of global environmental change in the Anthropocene.<sup>1</sup> Anthropogenic air pollution and human influence on the biosphere affect both public health and climate, and this Review attempts to provide an overview of particularly relevant substances and multiphase processes.

In section 2, we provide an overview of different groups of short-lived health- and climate-relevant air contaminants (SHCC) interacting at the atmosphere–biosphere interface. In section 3, we address in more detail the multiphase chemistry of reactive oxygen and nitrogen species (ROS, RNS)

and other select SHCC at biological surfaces of particular relevance: human lungs and skin, plant leaves and cryptogamic covers (photoautotrophic communities of algae, fungi, lichens, and bryophytes),<sup>15</sup> soil, and aquatic surfaces. In section 4, we conclude with a summary and outlook on key questions and future research perspectives of multiphase chemistry and atmosphere–biosphere interactions in the Anthropocene that may be important for the present state and future development of climate and public health.

## 2. HEALTH- AND CLIMATE-RELEVANT AIR CONTAMINANTS

Recent studies have used the term “short-lived climate pollutants” (SLCP) for atmospheric constituents that affect climate as well as public health with atmospheric lifetimes from days to a few decades.<sup>16</sup> Mitigation strategies of SLCP gain growing attention as it can have substantial cobenefits for health and climate protection,<sup>16</sup> although some investigations suggest that climate benefits from reductions in short-lived forcing agents are smaller than previously estimated.<sup>17</sup> Gas–aerosol interactions substantially alter the relative importance of SLCP emissions and influence the assessment and effects of mitigation policies.<sup>18</sup> While the narrow original definition of SLCP comprised only soot, ozone, hydrofluorocarbons (HFC), and CH<sub>4</sub>,<sup>16a</sup> we follow up on wider definitions<sup>19</sup> and suggest that all short-lived air contaminants relevant for health and climate, including toxic volatile and semivolatile compounds and elements like polycyclic aromatic hydrocarbon (PAH), isocyanic acid (HNCO), and mercury,<sup>20</sup> as well as aerosol particles from biogenic and anthropogenic sources such as primary biological and secondary organic aerosol (PBA/SOA), soil dust,<sup>21</sup> and volcanic ashes, be summarized under the common umbrella term “short-lived health- and climate-relevant air contaminants (SHCC)”. This umbrella term includes but goes beyond legislatively regulated categories of “hazardous air pollutants” and “toxic air contaminants”<sup>22</sup> (see Table 16.15 of Finlayson-Pitts and Pitts (2000)<sup>23</sup>). An overview of these substances, their sources, and their relevance to climate and health is given in Table 1, and in the following subsections we address the multiphase interactions and atmosphere–biosphere exchange of select groups of SHCC: reactive oxygen and nitrogen species; primary biological, secondary organic, and carbonaceous combustion aerosols; and other prominent air contaminants.

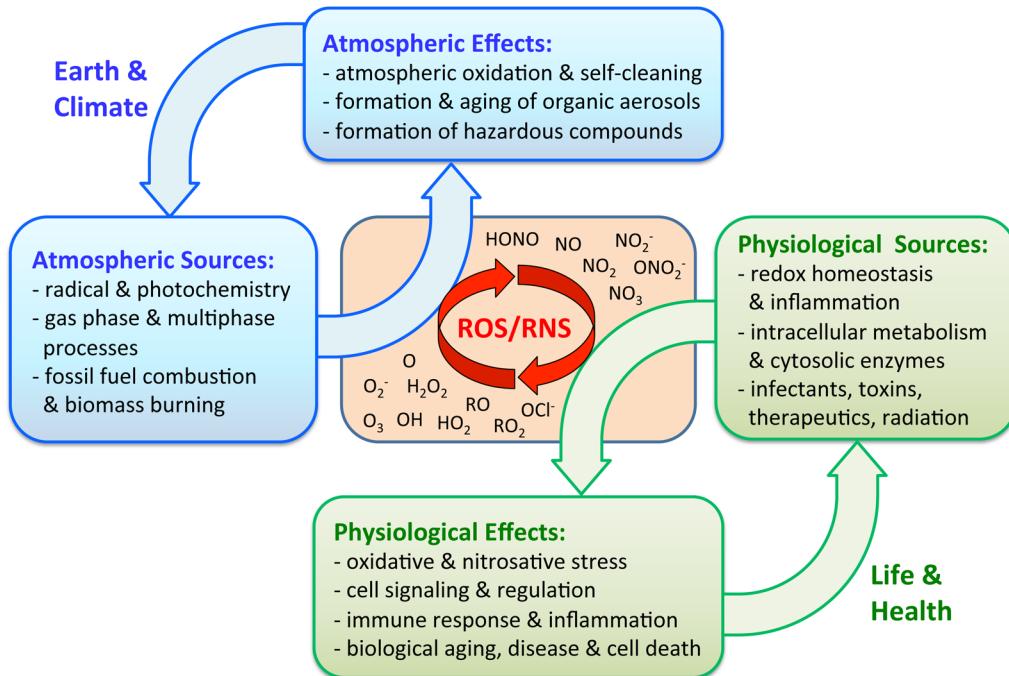
### 2.1. Reactive Oxygen and Nitrogen Species

Reactive oxygen and nitrogen species (ROS and RNS) are key species of both atmospheric and physiological chemistry, and their coupling through the atmosphere–biosphere interface is illustrated in Figure 4. ROS play a central role in the adverse health effects of air pollution, as they can cause oxidative stress, biological aging, cell death, and disease.<sup>9g,24</sup> ROS are produced in a wide range of atmospheric and physiological processes. In physiology and biomedical research, ROS are traditionally defined to include hydroxyl radical ( $\cdot\text{OH}$ ), singlet oxygen ( $^1\text{O}_2$ ), superoxide radical ( $\text{O}_2^{\bullet-}$ ), hydrogen peroxide ( $\text{H}_2\text{O}_2$ ), and ozone ( $\text{O}_3$ ).<sup>24b,25</sup> Extended definitions of ROS comprise a wide range of oxygen-centered and related free radicals, ions, and molecules, including organic peroxy radicals ( $\text{RO}_2^{\bullet}$ ), alkoxy and phenoxy radicals ( $\text{RO}^{\bullet}$ ), ozonides (OZ), organic hydroperoxides (ROOH), organic peroxides (ROOR), and hypochlorite ions ( $\text{OCl}^-$ ).<sup>24b,c,26</sup> In the atmospheric sciences, it is usually more common to speak of (photo)oxidants and radicals,

**Table 1. Overview of Short-Lived Health- and Climate-Relevant Air Contaminants (SHCC) with Major Sources and Relative Importance to Climate and Public Health**

substance classes and species	major sources	climate relevance	health relevance
<b>Reactive Oxygen and Nitrogen Species (ROS, RNS)</b>			
ozone ( $\text{O}_3$ )	photochemistry	++	++
hydroxyl ( $\text{OH}$ )	photochemistry	+	++
hydrogen peroxide ( $\text{H}_2\text{O}_2$ )	photochemistry		++
nitric oxide (NO)	combustion and biogenic emissions	+	++
nitrogen dioxide ( $\text{NO}_2$ )	photochemistry	+	++
nitrous acid (HONO)	photochemistry, biogenic emissions	+	+
nitric acid ( $\text{HNO}_3$ )	gas-phase chemistry	+	
nitrate radical ( $\text{NO}_3$ )	gas-phase chemistry	+	+
<b>Primary Biological Aerosols (PBA)</b>			
bacteria, fungal spores, pollen, etc.	biogenic emissions	+	++
<b>Secondary Organic Aerosols (SOA)</b>			
terpene and isoprene oxidation products	photochemistry, aqueous chemistry	+	+
alkane and aromatic oxidation products	photochemistry, aqueous chemistry	+	+
<b>Carbonaceous Combustion Aerosols (CCA)</b>			
soot, black/elemental carbon, brown carbon, organic carbon	combustion emissions	++	++
polycyclic aromatic hydrocarbons (PAH), nitro- and oxy-PAH	combustion emissions, photochemistry		++
<b>Other Organic and Inorganic Aerosol Components</b>			
inorganic salts and acids ( $\text{SO}_4^{2-}$ , $\text{NO}_3^-$ , etc.)	photochemistry, marine emissions	++	++
mineral and soil dust	dust emissions	++	++
humic-like substances (HULIS)	photochemistry, biogenic emissions	+	+
volcanic ash	volcanic emissions	+	+
transition metals	combustion and dust emissions		++
industrial nanoparticles	industrial activities		++
radionuclides	nuclear accidents		++
fly ash	combustion emissions		+
<b>Other Gas and Multiphase SHCC</b>			
nitrous oxide ( $\text{N}_2\text{O}$ )	biogenic emissions	++	
methane ( $\text{CH}_4$ )	biogenic emissions	++	
ammonia ( $\text{NH}_3$ )	biogenic emissions	+	+
hydrofluorocarbon (HFC)	industrial activities	++	
nonmethane volatile organic compounds (NMVOC)	biogenic and combustion emissions	+	+
carbon monoxide (CO)	combustion emissions	+	+
sulfur dioxide ( $\text{SO}_2$ )	photochemistry, combustion, and volcanic emissions	+	+
dimethyl sulfide (DMS)	marine biogenic emissions	++	
hydrogen sulfide ( $\text{H}_2\text{S}$ )	biogenic and volcanic emissions		+
isocyanic acid (HNCO)	combustion emissions		++
mercury ( $\text{Hg}$ )	combustion emissions		++

but we use ROS to summarize and describe the same group of reactive chemical species as in the life sciences. In addition, we use the term reactive oxygen intermediates (ROI) referring to ROS that are formed as intermediates upon interaction of O<sub>3</sub> and other atmospheric oxidants with aerosol particles.<sup>27</sup>



**Figure 4.** Reactive oxygen and nitrogen species (ROS, RNS): sources, effects, and interactions at the interface of atmospheric and physiological chemistry, with feedback loops involving the Earth system, climate, life, and health [adapted from Shiraiwa et al. (2011)<sup>27</sup>].

Physiological sources of ROS are redox homeostasis and inflammation as well as a variety of cytosolic enzyme systems and normal intracellular metabolism in mitochondria and peroxisomes, and ROS are widely used in cell signaling and regulation.<sup>24c,28</sup> Biological systems such as the phagocytic NADPH oxidase generate ROS endogenously as a defense against foreign organisms and other environmental challenges,<sup>29</sup> and ROS/RNS formation can be also triggered by exogenous sources such as environmental toxins, chemotherapeutics, ultraviolet light, inflammatory cytokines,<sup>24c</sup> and atmospheric pressure plasmas intended for biomedical purposes.<sup>30</sup> ROS production is usually counteracted by an antioxidant defense system including enzymatic scavengers such as superoxide dismutase (SOD) and catalase and nonenzymatic systems of low molecular mass antioxidants like ascorbate.<sup>31</sup> However, excessive production of ROS can overwhelm antioxidant defenses and trigger or enhance oxidative stress, cell death, biological aging, chronic inflammation, allergies, and various diseases.<sup>9g,24b,c,26a,32</sup>

In the atmosphere, ROS are generated via photochemistry and gas-phase, heterogeneous and multiphase reactions involving atmospheric oxidants and aerosol particles. ROS in the atmosphere play a key role in the chemical transformation of hazardous components.<sup>27</sup> Formation and growth of SOA are triggered by reaction of ozone, OH, and NO<sub>3</sub> radicals with volatile organic compounds, generating an array of carbon- and oxygen-centered free radicals.<sup>33</sup> Different types of ROS are closely coupled by radical chain reactions and cyclic transformation in atmospheric chemistry.<sup>4m,23,34</sup> The coupling and exchange of atmospheric and physiological ROS can proceed through various biosurfaces like the human respiratory tract, skin, plant leaves, and cryptogamic covers. ROS in the atmosphere can deposit to these surfaces causing physiological effects such as oxidative stress (see section 3). Atmospheric ROS are present both in the gas phase and in the particle phase. Most ROS like OH, HO<sub>2</sub>, H<sub>2</sub>O<sub>2</sub>, RO, RO<sub>2</sub>, and epoxides<sup>35</sup>

originate from photolysis of ozone and subsequent radical reactions. Recent studies suggest that stabilized Criegee intermediates from gas-phase reactions between ozone and unsaturated organic compounds are of great importance in atmospheric oxidation.<sup>36</sup> Note that stabilized Criegee intermediates may also play an important role as reactive oxygen intermediates in condensed phase oxidation chemistry as discussed below.<sup>37</sup> Depending on ambient conditions and reactivity, the atmospheric lifetimes of ROS range from less than seconds to more than days.<sup>4f,23,38</sup>

The hydroxyl radical, OH, plays a key role in atmospheric self-cleaning and reacts with most air contaminants, transforming them into water-soluble species that can be efficiently removed by wet deposition.<sup>13g,39</sup> Thus, OH is also frequently described as the “detergent of the atmosphere”. Because of its very high reactivity, OH exhibits very short lifetimes (seconds) and low mixing ratios (sub-ppt, i.e., <pmol mol<sup>-1</sup>) in the lower atmosphere, and through rapid radical reaction cycles it is tightly coupled with other ROS and RNS like HO<sub>2</sub>, O<sub>3</sub>, NO, NO<sub>2</sub>, HONO, organic oxy- and peroxyradicals, etc.<sup>4f,23,40</sup>

Hydrogen peroxide, H<sub>2</sub>O<sub>2</sub>, is also ubiquitous in the atmosphere with ppb mixing ratios (nmol mol<sup>-1</sup>) and mass concentrations on the order of 10 ng m<sup>-3</sup>.<sup>23</sup> It is highly water-soluble and undergoes partitioning between the gas phase, deliquesced aerosol particles, cloud, and fog droplets.<sup>41</sup> Several studies have measured the total oxidative capacity of PM in terms of H<sub>2</sub>O<sub>2</sub> in the particle phase with a fluorogenic probe.<sup>42</sup> A series of measurements have been conducted on ROS generation in fine-mode aerosols of laboratory-generated particles, source materials, and ambient particles collected in the Los Angeles area.<sup>43</sup> Particle H<sub>2</sub>O<sub>2</sub> levels were associated with transition metals and active quinones, and H<sub>2</sub>O<sub>2</sub> levels were sensitive to the pH of particle extraction solutions.<sup>43</sup> Laboratory-generated SOA by α-pinene, β-pinene, and toluene contain a substantial amount of H<sub>2</sub>O<sub>2</sub>, which is likely formed by decomposition or hydrolysis of hydroxyhydroperoxides, peroxy-

acids, and related species for  $\alpha$ - and  $\beta$ -pinene SOA, and from redox cycling of quinoid compounds for toluene SOA.<sup>44</sup>

The Fenton reaction,  $\text{Fe}^{2+} + \text{H}_2\text{O}_2$ , is one of the most important and widespread reactions in the multiphase chemistry of ROS and a major source of physiological ROS causing oxidative stress (metal-induced oxidative stress).<sup>46</sup> For example, the Fenton reaction is involved in the pathogenesis of asbestos-induced mesothelial carcinogenesis in the respiratory tract,<sup>47</sup> and it is crucial for the oxidation of sulfur dioxide and organic substances in cloud and fog droplets.<sup>48m,48</sup> Despite its importance, the mechanism of the Fenton reaction and the exact nature of the intermediates involved are still not fully understood. There are two possible first steps in Fenton chemistry: OH radical formation ( $\text{Fe}^{2+} + \text{H}_2\text{O}_2 \rightarrow \text{Fe}^{3+} + \text{OH}^- + \text{OH}$ ) or ferryl ion production ( $\text{Fe}^{2+} + \text{H}_2\text{O}_2 \rightarrow \text{Fe}^{\text{IV}}\text{O}^{2+} + \text{H}_2\text{O}$ ). The relative contribution of the two pathways can strongly influence the overall rates and course of oxidation in atmospheric droplets.<sup>48d</sup> A recent experimental study has shown that the dominant intermediate at pH 7 in water is the ferryl ion, while the formation of OH radicals becomes more important under acidic conditions.<sup>49</sup> Furthermore, Fenton chemistry at the air–water interface was found to predominantly produce ferryl intermediates, and the reaction rates may be substantially enhanced relative to bulk water.<sup>50</sup>

Some biomedical definitions of ROS include reactive nitrogen species (RNS), but in most studies and textbooks of physiological and atmospheric chemistry these are treated separately and comprise  $\text{NO}$ ,  $\text{NO}_2$ ,  $\text{NO}_2^-$ , and  $\text{ONOO}^-$  as well as  $\text{HONO}$  and  $\text{HNO}_3$ . RNS and ROS are tightly coupled through chemical reactions in the atmosphere<sup>4f,23</sup> as well as in the biosphere.<sup>25</sup> For example, nitric oxide (NO) is an important biological signaling molecule involved in cardiovascular regulation<sup>51</sup> as well as in inflammatory processes involving a wide range of ROS/RNS.<sup>32,52</sup> For the atmosphere, NO emissions are a major source of RNS, and NO is a key species in catalytic radical reaction cycles leading to photochemical production or destruction of ozone.<sup>4f,23</sup> Similarly, nitrite ions ( $\text{NO}_2^-$ ), nitrous acid ( $\text{HNO}_2$  alias HONO), and nitric acid ( $\text{HNO}_3$ ) are key species in both the biological and the atmospheric cycling of RNS and their interaction with ROS.

## 2.2. Primary Biological Aerosols

Biogenic aerosols can be regarded as an extension of the atmosphere–biosphere interface and play a vital role in the Earth system.<sup>4d,h,o,53</sup> Primary biological aerosols (PBA), in short bioaerosols, are directly released from the biosphere into the atmosphere and comprise cells, reproductive units, and fragments of viable or dead organisms (bacteria, fungal spores, viruses, pollen, plant debris, etc.).<sup>54</sup> As illustrated in Figure 3, PBA particles span the entire range of atmospheric aerosol particle diameters from nanometers to  $\sim 100 \mu\text{m}$ , whereby the lower limit is given by the size of molecular clusters or macromolecules and the upper limit is related to rapid sedimentation (i.e., larger particles are too heavy to remain airborne for extended periods of time).

PBA are essential for the biological reproduction and geographic spread of many organisms, biomes, and ecosystems, and they can cause or enhance human, animal, and plant diseases. The dispersal of plant, animal, and human pathogens and allergens has major implications for agriculture and public health,<sup>54s,55</sup> and the potential impacts of airborne transmission of genetically modified organisms (GMO) are under discussion.<sup>56</sup> Moreover, PBA can serve as nuclei for cloud

droplets, ice crystals, and precipitation, thus influencing the hydrological cycle and climate (Figure 2).<sup>14b,57</sup> Especially in pristine air over vegetated regions, bioaerosols are likely to be an essential regulating factor in the formation of clouds and precipitation,<sup>14b,58</sup> and bioprecipitation feedback mechanisms may have played an important role in the coevolution of life and climate.<sup>59</sup> For example, strong enhancements in the atmospheric concentration of bioaerosols and ice nuclei (IN) have been observed during and after rain events.<sup>58b,60</sup> Also in marine environments, particulate matter of biological origin may contribute substantially to the abundance of ice nuclei.<sup>61</sup>

While the passive release of PBA particles like anemophilous pollen or dry-discharged fungal spores is mostly wind-driven, some types of PBA particles like actively wet-discharged fungal spores are ejected by emission mechanisms that involve osmotic pressure or surface tension effects.<sup>62</sup> Note that these emission mechanisms involve hygroscopic water uptake by soluble compounds in a way similar to the processes governing the activation of (bio)aerosol particles as condensation nuclei of clouds and fog in the atmosphere.<sup>14b,58c,63</sup>

Recent studies suggest that the average number fluxes of emission and deposition of fungal spores and bacteria over continental regions are on the order of  $10^2 \text{ m}^{-2} \text{ s}^{-1}$ ,<sup>62,64</sup> which reflects an intense and rapid exchange of biological matter and information between atmosphere and biosphere. This exchange also influences the microbiome of ecosystems at the Earth surface, including the atmosphere–biosphere interfaces addressed in sect. 3 (human lungs<sup>65</sup> and skin,<sup>66</sup> plant leaves,<sup>59a,67</sup> and cryptogamic covers<sup>15c,68</sup>) and their chemical composition in response to microbial metabolic activity.

Estimates of global bioaerosol mass emission rates vary widely ( $10\text{--}1000 \text{ Tg yr}^{-1}$ ),<sup>54a</sup> and the regional and temporal variations in the atmospheric abundance and fluxes of emission and transport of different types of bioaerosol particles are poorly constrained.<sup>55a,b,61a,64,69</sup> In most terrestrial environments, PBA constitute a substantial fraction of the atmospheric aerosol load. With regard to number and mass concentration in the coarse particle size range with diameters larger than  $\sim 1 \mu\text{m}$ , PBA typically account for around  $\sim 30\%$  in urban and rural air<sup>4d,54a,58b,70</sup> and up to  $\sim 80\%$  in pristine rainforest air.<sup>14a,b,58c,62,71</sup> The number and mass concentrations of PBA particles over vegetated regions are typically on the order of  $10^4 \text{ m}^{-3}$  and  $1 \mu\text{g m}^{-3}$ , respectively.<sup>54a,58b,62,64b-e,70b,c,72</sup> First estimates suggest that the amount of DNA and biological information in urban fine particulate matter inhaled into the lungs of a human adult is as high as  $1 \mu\text{g}$  or  $10^8$  haploid bacterial genomes per day.<sup>73</sup> This may constitute a massive challenge to the immune system depending on the proportion and nature of pathogenic species. The actual identity, diversity, and abundance of different types of PBA particles, however, as well as their temporal and spatial variability and interactions with other air contaminants are not yet well characterized.

Allergies and associated respiratory diseases are a public health issue of increasing importance,<sup>74</sup> and epidemiological studies have demonstrated that the increasing frequency of pollen induced respiratory allergy correlates with urbanization and high levels of air pollution.<sup>75</sup> Proteins account for up to  $\sim 5\%$  of urban air particulate matter and can be found in coarse biological particles such as pollen grains and also in the fine fraction of air particulate matter with fine fragments of pollen, microorganisms, or plant debris.<sup>76</sup> Pollen grains interact with air pollution associated with  $\text{O}_3$ ,  $\text{NO}_2$ ,  $\text{SO}_2$ , and particulate matter such as soot, exhaust particles, or dust, possibly

enhancing allergic reactions and immunological effects of pollen proteins.<sup>77</sup>

Laboratory and field experiments have shown that O<sub>3</sub> and NO<sub>2</sub> can promote the nitration of protein molecules and peptides in polluted urban air.<sup>27,40d,76b,78</sup> This post-translational modification can enhance the allergenic potential of proteins like the birch pollen allergen Bet v 1,<sup>79</sup> and biomedical data suggest strong links between protein nitration and various diseases.<sup>80</sup> The mechanism of protein nitration by NO<sub>2</sub> and O<sub>3</sub> involves the formation of long-lived reactive oxygen intermediates (ROI), which are most likely tyrosyl radicals (phenoxy radical derivatives of tyrosine).<sup>27,78d,81</sup> Phenoxy radicals can be efficiently stabilized in the condensed phase,<sup>82</sup> which is consistent with the observed long lifetime of the ROI (>10 min). In addition, formation of dimers was observed upon ozone exposure to protein,<sup>78d</sup> which may also proceed through tyrosyl radicals.<sup>81b</sup> In general, post-translational modifications like oxidation, nitration, and oligomerization can enhance the allergenic potential of proteins and may provide a molecular rationale for the enhancement of allergic diseases by air pollution.<sup>4h,26a,76b,78a,79a,80b,83</sup> Thus, adverse health effects of PBA may have been and continue to be exacerbated by human activities changing regional and global air quality in the Anthropocene as further discussed in the final section of this Review.

### 2.3. Secondary Organic Aerosols

SOA account for a major fraction of fine particulate matter in the atmosphere,<sup>8d,h,i,k,84</sup> affecting climate by scattering sunlight and serving as nuclei for cloud droplets and ice crystals.<sup>5,8c,e,j–m,85</sup> They are formed via oxidation of volatile organic compounds (VOC) by atmospheric oxidants such as OH, O<sub>3</sub>, NO<sub>3</sub>, or reactive halogen species,<sup>86</sup> generating myriads of semi- and low-volatile compounds that partition into the particle phase.<sup>33b,87</sup> Prominent SOA precursors include biogenic VOC<sup>88</sup> such as isoprene, monoterpenes (C<sub>10</sub>H<sub>16</sub>: α- and β-pinene, limonene, etc.), and sesquiterpenes (C<sub>15</sub>H<sub>24</sub>) as well as anthropogenic VOC<sup>89</sup> such as alkanes and aromatics. Recent studies have suggested that SOA and biologically produced primary organic aerosols could be distinguished on the basis of chirality and stereochemical speciation.<sup>90</sup>

Organic particles in the atmosphere usually contain also some inorganic salts or acids and water. Such mixtures may follow complex, nonideal behavior including liquid–liquid or liquid–solid phase separation as predicted by thermodynamic models,<sup>91</sup> which has been demonstrated in the laboratory<sup>92</sup> and field studies.<sup>58c,93</sup> Phase-separated organic–inorganic particles can adopt either a partial engulfing morphology or a core–shell morphology, depending on relative differences of the volumes and interfacial tensions between the phases.<sup>92a,f,h</sup>

Until recently, organic aerosol particles in the atmosphere were thought to be liquid and thus internally well-mixed. Depending on temperature and relative humidity, however, organic particles can also adopt amorphous solid or semisolid states. Upon decreasing humidity or temperature, many organic substances tend to form amorphous semisolid or solid (glassy) rather than crystalline phases.<sup>94</sup> Amorphous substances have no long-range atomic order and are classified as solid glasses when their viscosity exceeds 10<sup>12</sup> Pa s. Semisolid substances like rubbers, gels, or ultraviscous liquids have viscosities that are in the range from ~10<sup>12</sup> to ~10<sup>2</sup> Pa s, which is still orders of magnitude higher than the viscosity of liquid water at ambient conditions (~10<sup>-3</sup> Pa s).<sup>4a,94d</sup> Ambient and laboratory-

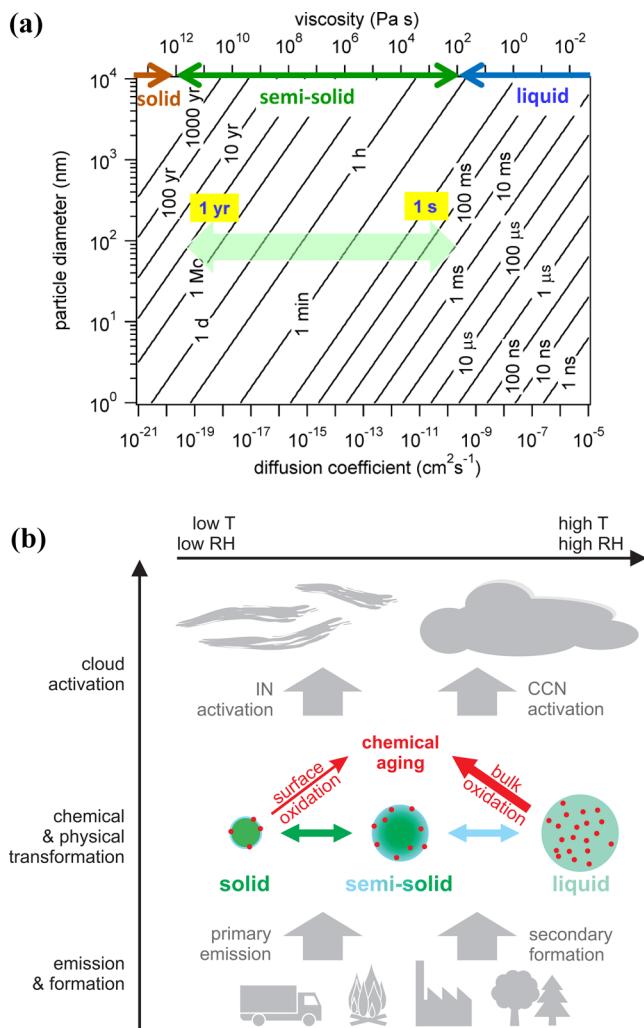
generated organic particles have been observed to bounce off the smooth hard surface of an inertial impactor at low RH, implying a nonliquid state.<sup>95</sup> These observations are consistent with field measurements<sup>96</sup> and laboratory studies<sup>33a,97</sup> observing large amounts of high molecular weight organic compounds (oligomers) that tend to have high glass transition temperatures and form an amorphous solid or semisolid phases.<sup>94c</sup> Viscosity measurements performed in recent studies provide direct evidence that SOA particles formed upon oxidation of α-pinene<sup>98</sup> and isoprene<sup>99</sup> are liquid at high RH and amorphous semisolid or glassy at low RH and ambient temperature.

Through the Stokes–Einstein equation, the viscosity of an amorphous organic substance can be related to its molecular self-diffusion coefficient, which in turn can be converted into a characteristic *e*-folding time of mass-transport and mixing by molecular diffusion in aerosol particles as shown in Figure 5a. In the size range of the accumulation mode of atmospheric aerosols (particle diameters of the order of ~10<sup>2</sup> nm), which is particularly relevant for SOA, the characteristic time of diffusion varies from fractions of seconds for liquids to years for solids.<sup>4a</sup> Note that the Stokes–Einstein equation may not be applicable for self-diffusion of organic molecules and diffusion of water and small oxidants in highly viscous matrix close to the glass transition temperature.<sup>4a,100</sup> Moreover, the phase state of organic particles in the nucleation mode of atmospheric aerosols (particle diameters around ~20 nm) can be strongly influenced by nanosize effects.<sup>101</sup>

As illustrated in Figure 5b, bulk diffusion can limit the kinetics of mass transport and reaction in amorphous organic aerosol particles and determine if chemical transformation proceeds only at the surface or throughout the particle.<sup>4a</sup> Gas uptake of atmospheric trace gases such as O<sub>3</sub>, NO<sub>3</sub>, OH, NH<sub>3</sub>, and N<sub>2</sub>O<sub>5</sub> and chemical transformation of organic particles can be significantly affected by phase state<sup>4a,102</sup> and mixing state.<sup>103</sup> Aerosol hygroscopic growth and CCN activation can be inhibited,<sup>94d,g,104</sup> and activation pathways of homogeneous or heterogeneous ice nucleation can be influenced substantially.<sup>92c,94e,f,105</sup> Evaporation of SOA particles has been found to proceed slowly upon dilution<sup>106</sup> or heating.<sup>107</sup> Traditionally, gas–particle partitioning of organic substances is considered to be established quasi-instantaneously<sup>108</sup> supported by measurements that are consistent with absorptive partitioning;<sup>109</sup> however, this assumption can be invalid if particles or certain phases therein are semisolid or glassy.<sup>91b,96a,106c,110</sup> Kinetic limitations by mass transfer and transport in the particle phase also affect SOA formation kinetics<sup>95a,111</sup> and particle size distribution dynamics.<sup>112</sup>

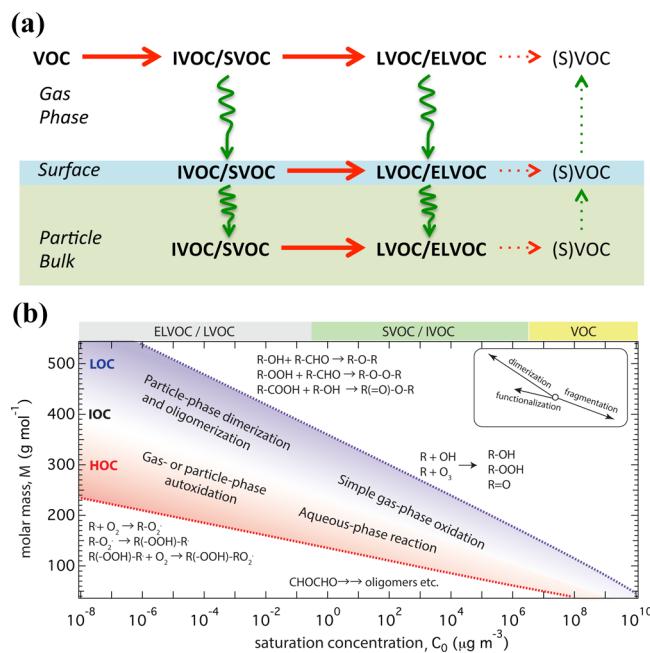
The formation and evolution of SOA are complex multiphase processes, involving a series of chemical reactions and mass transport in the gas phase, at the gas–particle interface, and in the particle phase as outlined in Figure 6a. The chemical kinetics of these processes can be efficiently described and depicted in terms of kinetic regimes and limiting cases as illustrated in Figure 6b. The parameters, which determine the limiting steps and classification, are the main location of chemical reaction, the saturation ratio of the reactants, and the extent of spatial heterogeneity of the gas and particle phases.<sup>113</sup> They can be resolved by kinetic models explicitly describing mass transport and chemical reactions<sup>112e,114</sup> with kinetic input parameters such as reaction rate coefficients,<sup>33a</sup> gas<sup>115</sup> and bulk diffusivities,<sup>4a,92f,105j,116</sup> and accommodation coefficients.<sup>117</sup>

Figure 6b shows a 2D map of SOA evolution with regard to volatility and molar mass along with characteristic reaction



**Figure 5.** Organic aerosols in the atmosphere: oil drops, rubber balls, and glass pearls. (a) Characteristic time of bulk diffusion ( $\tau_{cd}$ ) in liquid, semisolid, and solid particles as a function of diffusion coefficient and particle diameter. In the size range of the atmospheric aerosol accumulation mode (particle diameters around  $\sim 10^2$  nm),  $\tau_{cd}$  in semisolid particles varies from seconds to years (light green arrow). (b) Organic particulate matter in the atmosphere is usually amorphous, and its phase state can vary between liquid, semisolid, and solid (glassy), depending on ambient relative humidity and temperature. Particle phase state, viscosity, and diffusivity play an important role in most aerosol interactions like uptake and partitioning of reactive and condensable gases, chemical transformation and aging, and activation as CCN or IN [from Shiraiwa et al. (2011)<sup>4a</sup>].

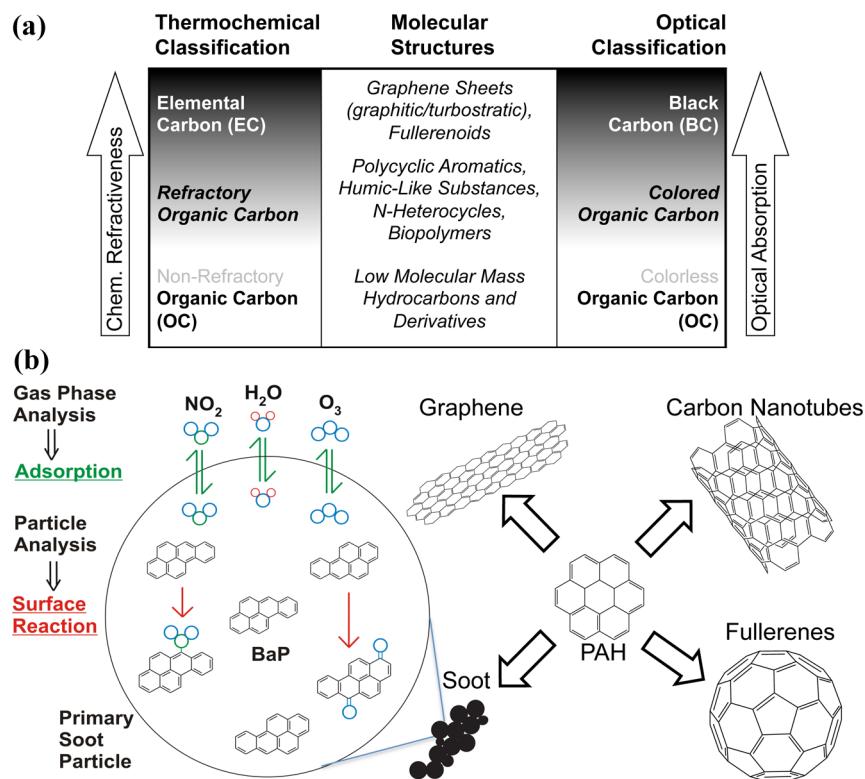
pathways and relevant kinetic regimes.<sup>113a</sup> Volatility or vapor pressure of some of the organic compounds were measured,<sup>118</sup> and if not measured, they can be estimated by several methods.<sup>119</sup> Besides the traditional classification of organic compounds into volatile, semivolatile, and low-volatile (VOC, SVOC, LVOC), the SOA research community also distinguishes further subsets of compounds with intermediate or extremely low volatility (IVOC, ELVOC).<sup>120</sup> Molecular identification of SOA oxidation products shows that the chemical evolution of SOA from a variety of VOC precursors adheres to characteristic “molecular corridors” with a tight inverse correlation between saturation mass concentration ( $C_0$ ) and molar mass ( $M$ ). They are constrained by two boundary lines corresponding to the volatility of *n*-alkanes  $C_nH_{2n+2}$  and



**Figure 6.** Multiphase chemical evolution of secondary organic aerosol (SOA). (a) Schematic outline of sequential and parallel reactions in the gas phase, at the particle surface, and in the particle bulk lead to multiple generations of volatile, semivolatile/intermediate volatile, and low-volatile/extremely low-volatile organic compounds (VOC, SVOC/IVOC, LVOC/ELVOC). Red and green arrows denote chemical reactions and mass transport; solid and dotted lines indicate likely major and minor pathways, respectively. (b) Molecular corridors of SOA evolution in a diagram of molar mass ( $M$ ) versus volatility (saturation mass concentration,  $C_0$ ) of reaction products. Experimental data for different SOA precursors and reaction pathways tend to cluster in high, intermediate, and low O:C corridors (HOC, red shaded area; IOC, white area; LOC, blue shaded area) [adapted from Shiraiwa et al. (2014)<sup>113a</sup>].

sugar alcohols  $C_nH_{2n+2}O_n$ . They illustrate the common range and regular dependence of volatility on the molar mass of organic compounds and reflect that the decrease of volatility with increasing molar mass is stronger for polar compounds.<sup>113a</sup> Many early generation gas-phase oxidation products of alkanes<sup>97a,121</sup> as well as dimers or oligomers fall close to the  $C_nH_{2n+2}$  line, designated as low O:C (LOC) corridor. Oxidation products of isoprene,<sup>35,122</sup> glyoxal and methylglyoxal,<sup>123</sup> and autoxidation products have a high O:C ratio and tend to fall near the  $C_nH_{2n+2}O_n$  line, designated as HOC corridor, while  $\alpha$ -pinene<sup>91b,97b,h,124</sup> and limonene<sup>125</sup> fall into the area characterized by intermediate O:C (IOC) ratios.

As illustrated in the inset in Figure 6b, the three main reaction types of SOA evolution are functionalization, oligomerization, and fragmentation.<sup>8d,126</sup> Single-step functionalization usually leads to a small increase in molar mass and decrease in volatility,<sup>108a</sup> and fragmentation leads to a substantial decrease of molar mass and increase in volatility;<sup>97a,121d,127</sup> these products tend to follow so-called quasi-equilibrium growth.<sup>110b,112g</sup> Dimerization and oligomerization decrease volatility by multiple orders of magnitude;<sup>128</sup> such reactions may be limited by reaction or diffusion in the particle bulk<sup>4a,33a,102f,j,112e,129</sup> or by gas-to-particle mass transfer when the reactions are sufficiently fast catalyzed by acid.<sup>35b,130</sup> Aqueous-phase processing is an efficient pathway for formation of low volatility and semivolatile HOC com-



**Figure 7.** Carbonaceous combustion aerosols (CCA): (a) Composition, properties, and classification with regard to molecular structures, thermochemical refractivity, and specific optical absorption. Adapted with permission from ref 4h. Copyright 2005 John Wiley. (b) Polycyclic aromatic hydrocarbons (PAH) serve as building blocks for soot particles as well as fullerenes, carbon nanotubes, and graphene. Further chemical transformation of PAH like benzo[*a*]pyrene (BaP) on soot in the atmosphere by ROS and RNS like ozone, nitrogen dioxide, and water vapor involves multiple steps of reversible and competitive adsorption as well as surface and bulk reactions, which can be resolved by combined gas and particle analysis. Adapted with permission from ref 83d. Copyright 2002 Mary Ann Liebert, Inc.

pounds<sup>48b,c,123a–d,f–m,131</sup> including light-absorbing compounds like imidazoles.<sup>132</sup> Recently, highly oxidized extremely low volatility organic compounds (ELVOC) have been detected in field and chamber experiments,<sup>133</sup> which may be formed via autoxidation.<sup>134</sup> An apparent volatility of condensing vapors was observed to be indeed very low, and partitioning of ELVOC is most likely limited by mass transfer.<sup>8c,110b,135</sup>

Molar mass and O:C ratio also correlate with the glass transition temperature of organic compounds;<sup>94a,c</sup> hence molecular corridors may allow one to infer change of particle phase state upon aging.<sup>136</sup> Molecular corridors and kinetic regimes facilitate the specification of the rate of progression to higher generation products;<sup>113a</sup> thus, they may serve as a basis for compact representation of SOA evolution in regional and global models of climate and air quality<sup>137</sup> and also for interpretation of model outputs in detailed SOA models.<sup>48f,g,108a,112d,g,114b,120b,g,128,138</sup> Moreover, such representation should be applicable not only for aerosols including biogenic SOA and PBA particles, but also for gas uptake and generic multiphase chemistry at the atmosphere–biosphere interface (e.g., soil, lung, skin, and leaf surfaces).

As SOA particles account for a large fraction of fine particulate matter in the atmosphere, their formation, composition, and phase state are relevant for aerosol effects on climate as well as public health. The detailed chemical composition and phase state appears to have only limited influence on the activation of SOA particles as cloud condensation nuclei (CCN),<sup>139</sup> but it is likely important for the activation of aerosol particles as ice nuclei (IN).<sup>94c,f,105a,f,g,i</sup>

With regard to SOA health effects, substantial amounts of particle-bound ROS are found on biogenic SOA produced from  $\alpha$ -pinene, linalool, and limonene prepared in laboratory chambers.<sup>125c,140</sup> In addition, ROOH and organic peroxide (ROOR) are found to be major constituents of biogenic SOA and aged organic aerosols.<sup>33a,97h,141</sup> SOA containing highly oxidized and high molar mass compounds may induce distinct effects in lung cells.<sup>142</sup> It seems very important to identify and quantify reactive oxygen species and intermediates (ROS/ROI),<sup>27,143</sup> stable free radicals,<sup>38b</sup> and redox-active compounds<sup>144</sup> like quinones and nitrated species as discussed in sections 2.1 and 2.4. In view of the different types of precursor molecules and molecular composition of biogenic SOA (isoprene, terpenes, etc.) and anthropogenic SOA (alkanes, aromatics, etc.), it seems likely that their content of hazardous compounds and health effects are different as well. Moreover, the chemical composition and hazardous potential may also vary with different reaction pathways in the formation and aging of SOA depending on ambient atmospheric conditions.<sup>102g,121a,122f,129,144b,c,145</sup>

## 2.4. Carbonaceous Combustion Aerosols

Soot is the most prominent component of carbonaceous combustion aerosols (CCA) from fossil fuel combustion and biomass burning.<sup>146</sup> It consists primarily of black or elemental carbon (BC, EC) mixed with organic compounds and other substances like sulfate and metal oxides.<sup>147</sup> Traditionally the total carbon (TC) content of air particulate matter is defined as the sum of all carbon contained in the particles, except in the form of inorganic carbonates. TC is usually determined by

thermochemical oxidation and evolved gas analysis ( $\text{CO}_2$  detection), and divided into an organic carbon (OC) fraction and a BC or EC fraction with physicochemical properties that resemble graphitic materials. Measurements of BC and EC are generally based on optical and thermochemical techniques, and OC is operationally defined as the difference between TC and BC or EC. As illustrated in Figure 7a, however, there is no real sharp boundary but a continuous decrease of thermochemical refractivity and specific optical absorption going from graphite-like structures to nonrefractive and colorless organic compounds, respectively. Both BC and EC consist of the carbon content of the graphite-like material usually contained in soot (technically defined as the black product of incomplete hydrocarbon combustion or pyrolysis) and other combustion aerosol particles, which can be pictured as more or less disordered stacks of graphene layers, fullerenoid structures, and large polycyclic aromatic molecules.<sup>4h,148</sup>

Depending on the applied optical or thermochemical methods (absorption wavelength, temperature gradient, etc.), however, BC and EC measurements also include the carbon content of colored and refractory organic compounds, which can lead to substantially different results and strongly limits the comparability and suitability of BC, EC, and OC data for the determination of mass balances and physicochemical properties of air particulate matter. Optical methods for the detection of BC are usually nondestructive and allow (near-)real-time operation, but on the other hand they are particularly prone to misinterpretation. They mostly rely on the assumption that BC is the dominant absorber and exhibits a uniform mass-specific absorption coefficient or cross-section. Although these assumptions may be justified under certain conditions, they are questionable in the context of detailed chemical characterization of aerosol particles. Thus, optically determined BC values should generally be regarded as mass-equivalent values but not as absolute mass or concentration values. Beside different types of graphite-like material, there are at least three classes of atmospherically relevant organic compounds that contribute to the absorption of visible light by air particulate matter: polycyclic aromatics, humic-like substances, and nitrogen-containing organics.<sup>4h,123c,132d,148c,d,149</sup> These are frequently categorized under variants of the umbrella terms of black, brown, or light-absorbing carbon. Improved characterization and distinction of these substance groups can be achieved by advanced optical and microscopic techniques like Raman spectroscopy,<sup>148e,150</sup> single particle soot photometry with laser-induced incandescence,<sup>15f</sup> and electron and X-ray microscopy.<sup>148a,c,152</sup>

CCA contribute to regional and global climate change because of their role in direct, indirect, and semidirect radiative forcing.<sup>4h,o,7f,149a,b,153</sup> Upon emission from combustion sources, fresh soot is initially hydrophobic and mostly externally mixed with nonrefractory compounds; condensation of semivolatile compounds and chemical processing by ozone and other oxidants can alter soot mixing state,<sup>154</sup> influencing the radiative properties,<sup>155</sup> hygroscopic properties, and CCN activity.<sup>104e,153a,156</sup> Note that some studies emphasize the potential importance of simple coatings and core/shell structures, whereas more recent studies suggest that the detailed morphology resulting from coagulation or phase segregation may be more important.<sup>148a,151a,157</sup>

CCA including cigarette smoke and soot from biomass and fossil fuel combustion contain high concentrations of stable free radicals and ROS, which can be directly detected using electron

paramagnetic or spin resonance (EPR/ESR) techniques.<sup>158</sup> Such stable free radicals in combustion particles have been attributed to the existence of various oxygenated compounds including resonantly stabilized free radicals of propargyl, phenoxy, and cyclopentadienyl origins.<sup>158b</sup> Recent electronic structure calculations have suggested that a class of persistent free radicals can be dynamically generated from aromatics with extremely weakly bound carbon–carbon bonds, such as the hybrid derivatives of acenaphthene and hexaphenylethane,<sup>159</sup> leading to formation of stable carbon-centered biradicals.<sup>159</sup>

Cigarette smoke, including “second-hand” smoke inhaled unintentionally and “third-hand” smoke desorbed from indoor surfaces, does not only contain many stable organic compounds that are toxic and carcinogenic such as nicotine nitrosamines, and polycyclic aromatic hydrocarbons.<sup>160,161</sup> The smoke also contains oxidized radicals like semiquinone radicals, which are formed in combustion of hydrocarbon precursors including hydroquinones, catechols, phenols, and benzenes.<sup>158b,162</sup> The observation of semiquinone radicals in cigarette smoke implies that these radicals may be also generated by a variety of sources including biomass fuels and fossil fuels. Indeed large quantities of environmentally persistent free radicals (EPFR) with EPR characteristic signals similar to semiquinone radicals were detected in ambient PM2.5 samples.<sup>158d,163</sup> Gehling and Dellinger<sup>38b</sup> quantified the average bulk concentration of EPFR in PM2.5 collected in Baton Rouge, LA, as on the order of  $10^{16}$ – $10^{18}$  radicals g<sup>-1</sup>. As the radical signal in PM2.5 by EPR was remarkably similar to that of semiquinones in cigarette smoke, such radical concentration was suggested to be expressed as the number of equivalent cigarettes smoked, resulting in 0.4–0.9 equiv cigarettes per day for nonextreme air quality in the United States.<sup>38b</sup>

It has been shown that formation of EPFR occurs when an organic precursor chemisorbs at a redox site of transition metals such as Cu and Fe, subsequently reducing the metal via electron transfer.<sup>82,158b,164</sup> The EPR signals of EPFR in ambient PM2.5 were observed to have three decays: a fast decay, slow decay, and no decay, corresponding to phenoxy radicals, semiquinone radicals, and internal radicals restricted in a solid matrix. On the basis of such measurements, the lifetime of phenoxy radicals is estimated to be on the order of days, whereas those of semiquinone radicals are more than months and even years.<sup>38b</sup>

Polycyclic aromatic hydrocarbons (PAH) are among the most prominent combustion aerosol components relevant for public health as they act as mutagens and carcinogens.<sup>165</sup> They originate mainly from biomass burning, fossil fuel combustion, and motor vehicles,<sup>166</sup> and they can be regarded as molecular building blocks of soot and related carbonaceous materials (Figure 7b).<sup>148e,f,167</sup> While soot particles are formed in bimolecular addition reactions of small unsaturated hydrocarbons (e.g., C<sub>2</sub>H<sub>2</sub>) in the course of incomplete combustion, the formation of fullerenes occurs mainly through unimolecular reactions at high temperatures.<sup>148g,168</sup> Moreover, PAH are starting materials and intermediates in the chemical synthesis of carbon nanotubes, nanoribbons, and graphene layers.<sup>169</sup> PAH consisting of a few fused benzene rings, such as naphthalene, are volatile and reside mainly in the gas phase, whereas PAH containing five or more rings, such as benzo[a]pyrene and coronene, are mostly in the particulate phase. Intermediate PAH, such as 3–4 ring PAH, partition to both phases to a significant extent, depending on temperature, vapor pressure, affinity to soot or elemental carbon, and lipophilicity (solubility

in particulate organic matter), hence strongly influenced by the matrix composition.<sup>4f,170</sup>

PAH are reactive toward atmospheric oxidants including OH, O<sub>3</sub>, NO<sub>3</sub>, and NO<sub>2</sub>, forming various products of oxy- and nitro-PAH such as quinones and epoxides.<sup>165b,171</sup> Photooxidation and subsequent gas-phase reaction of volatile PAH like naphthalene leads to generation of an array of semi- and low-volatile compounds forming SOA.<sup>144a,172</sup> Chemically transformed PAH derivatives, particularly quinones and nitro-derivatives, are often even more toxic than parent PAH.<sup>23,170c,173</sup> Therefore, and because of the potential health effects of ROS/RNS and catalytic metals, emission control technologies for carbonaceous combustion aerosols, which often rely on conditioning and regeneration involving gaseous ROS/RNS or metal catalysts, should be designed in a way that minimizes or avoids emissions of excess ROS/RNS and byproducts like oxy- or nitro-PAH and metals.<sup>4h,167,171c,174</sup> Especially fly ash particles and residues in combustion aerosols contain various transition metals including iron,<sup>175</sup> which is also shown to increase bacterial growth and impair host innate immunity.<sup>176</sup>

Surface and multiphase reactions of soot, PAH, and benzene derivatives with ozone, nitrogen oxides, and other ROS/RNS have been investigated extensively in laboratory experiments and kinetic model studies.<sup>23,102f,117f,127b,165b,177</sup> Reaction kinetics of O<sub>3</sub> and NO<sub>2</sub> with PAH adsorbed on soot and other substrates generally show a nonlinear dependence of the PAH degradation rate on gas-phase oxidant concentration.<sup>167,177i,178</sup> Such behavior can be explained by a so-called Langmuir–Hinshelwood mechanism,<sup>4c,117f,177u,179</sup> in which a molecule adsorbs to the surface for some time followed by surface reaction (Figure 7b). Recent studies suggest that even highly reactive gases such as OH and NO<sub>3</sub> may follow the Langmuir–Hinshelwood mechanism.<sup>4b,102e,h,180</sup> The surface-residence times or desorption lifetimes of O<sub>3</sub> inferred from kinetic data and simple Langmuir–Hinshelwood rate equations are typically in the range 0.01–10 s.<sup>177s,178</sup> However, according to molecular dynamic simulations<sup>181</sup> and density functional theory (DFT),<sup>182</sup> the desorption lifetime of O<sub>3</sub> on the air–surface interface should be only nanoseconds, which is more than 6 orders of magnitude shorter. This indicates that the DFT simulations address a different state of O<sub>3</sub> adsorption than the reaction kinetic experiments. Such discrepancies are reconciled in a kinetic model including decomposition of weakly bound O<sub>3</sub> (i.e., physisorption) into a state of stronger binding to the surface (i.e., chemisorption).<sup>27</sup> The dissociation products are molecular oxygen that desorb back to the gas phase<sup>183</sup> and a reactive oxygen intermediate (ROI), such as a chemisorbed oxygen atom<sup>177i,184</sup> bound to the delocalized  $\pi$ -electrons of an aromatic surface (epoxide-like structure) as suggested by DFT calculations.<sup>182</sup> Model calculations suggest that ROI can be long-lived with chemical lifetimes exceeding 100 s.<sup>27</sup>

It has been demonstrated that there is a strong similarity between the heterogeneous reaction kinetics of ozone on a wide variety of surfaces, including soot, 1-hexadecene, PAH adsorbed on a variety of surfaces,<sup>179b,185</sup> metal oxides, mineral dust, and bioparticles (proteins). This suggests that the ozone loss may proceed through a common reaction pathway on such surfaces, likely involving formation of long-lived ROI. The formation of atomic oxygen and peroxides was invoked to explain the kinetics of O<sub>3</sub> decomposition on mineral dust.<sup>186</sup> The ozonolysis of olefins involves ozonides or stabilized

Criegee intermediates.<sup>37</sup> The chemical identity of O<sub>3</sub>-generated ROI probably varies with the nature of the substrate, but their formation energetics of decomposition and reaction of O<sub>3</sub> may be similar for many substrates. The chemical lifetime of ROI needs to be long to explain the Langmuir–Hinshelwood reaction kinetics observed upon interaction of O<sub>3</sub> with aerosol particles.

Beyond the similarities of ozone reactions with different types of atmospheric aerosol surfaces, it is instructive to realize that heterogeneous and multiphase reactions between carbonaceous aerosol particles and gaseous oxidants exhibit similar kinetic and mechanistic features also at greatly different temperature levels. For example, the gasification of soot and PAH by O<sub>2</sub> and NO<sub>2</sub> during high-temperature conditioning and regeneration of diesel vehicle exhaust filter systems exhibits kinetic features similar to those of the reaction of soot and PAH with O<sub>3</sub> and NO<sub>2</sub> at ambient temperatures.<sup>167,171c,177s</sup> Note that such analogies are consistent with the traditional understanding of the gas-phase oxidation of volatile organic compounds in high-temperature combustion processes and low-temperature atmospheric chemistry, which can also be regarded as a kind of “low temperature combustion”.<sup>187</sup>

## 2.5. Other Air Contaminants Linking Atmospheric and Physiological Chemistry

Similar to nitric oxide (NO), also carbon monoxide (CO) and hydrogen sulfide (H<sub>2</sub>S) are compounds that occur as atmospheric trace gases<sup>4f,23</sup> and have important physiological functions.<sup>188</sup> H<sub>2</sub>S and CO range from 0 to 800 ppt and 40–200 ppb, respectively.<sup>4f,189</sup> NO, CO, and H<sub>2</sub>S can freely permeate through cell membranes and are all produced endogenously in the body to exert a variety of well-defined biological functions, such as serving as vasodilators, promoting angiogenesis, and protecting tissues against damage.<sup>18a,190</sup> Their roles in inflammation are very complicated, showing both pro- and anti-inflammatory effects, depending on their concentrations and complex interplay.<sup>18a</sup>

Sulfur dioxide (SO<sub>2</sub>) originates from fossil fuel combustion<sup>191</sup> and oxidation of H<sub>2</sub>S, dimethyl sulfide (DMS), and other organosulfur compounds.<sup>192</sup> Exposure to a high concentration of sulfur dioxide may lead to irritations of the nose, throat, and airways causing coughing, wheezing, or shortness of breath, and eventually respiratory diseases.<sup>193</sup> SO<sub>2</sub> can be efficiently converted to sulfate (SO<sub>4</sub><sup>2-</sup>) by in-cloud aqueous phase oxidation catalyzed by transition metals, modifying aerosol size distribution with important implications for the magnitude of indirect and direct aerosol cooling and the impact of SO<sub>2</sub>.<sup>4f,48a,194</sup>

Isocyanic acid (HNCO) is a toxic compound contained in the smoke of tobacco, combustion, and biomass burning.<sup>18b,195</sup> HNCO can be also emitted in the exhaust of advanced PM-NO<sub>x</sub> abatement technology of selective catalytic reduction catalysts.<sup>196</sup> HNCO is highly water-soluble and largely dissociated into cyanate ions (NCO<sup>-</sup>) at physiological pH. Recent studies suggest that HNCO is dissolved in cloudwater more efficiently than expected on the basis of the effective Henry's law solubility and propose a secondary, photochemical source of HNCO in ambient air.<sup>197</sup> HNCO can react with amine, hydroxyl, and sulphydryl groups to form carbamyl groups. Carbamylation of proteins is a key step for inflammation linked to a number of health effects including cardiovascular disease,therosclerosis, cataracts, and rheumatoid arthritis.<sup>198</sup> Exposure levels of >1 ppb may provide a direct

source of HNCO to humans at levels that may lead to such adverse health effects.<sup>18b</sup> The gas-phase concentration of HNCO can be up to 600 ppb in laboratory biomass fires and up to 200 ppt in ambient air in urban Los Angeles, CA and in Boulder, CO.<sup>18b,195</sup> Global chemical transport modeling has suggested that annual mean surface HNCO concentrations were highest over parts of China (maximum of 470 ppt), but episodic fire emissions gave much higher levels, exceeding 4 ppb in tropical Africa and the Amazon, and exceeding 10 ppb in Southeast Asia and Siberia. These results suggest that large biomass burning events associated with high concentrations of HNCO and aerosol particles could result in deleterious health effects for the human population in these regions.<sup>199</sup>

Mineral dusts are lifted into the atmosphere with a global annual flux of ~2000 Tg yr<sup>-1</sup>.<sup>200</sup> They have massive adverse health effects causing a number of lung diseases and pulmonary disorders,<sup>201</sup> and they affect local and global climate by absorbing and scattering solar radiation, and serving as CCN and IN.<sup>7a,57d,e,202</sup> Dust particles can be transported over thousands of kilometers, and recent investigations indicate that Sahara dust can influence cloud formation and precipitation in the Western U.S.<sup>203</sup> The long-range transport, deposition, and accretion of dust particles, possibly associated with microorganisms,<sup>204</sup> can affect the biogeochemical cycles and spread of biomes. For example, Sahara dust delivers key nutrients and contributes to fertilization of the Amazon rainforest.<sup>205</sup> Heterogeneous reactions on dust particles during transport may affect various trace gases including O<sub>3</sub>, NO<sub>x</sub>, NO<sub>3</sub>, N<sub>2</sub>O<sub>5</sub>, SO<sub>x</sub>, HO<sub>x</sub>, and organic compounds.<sup>206</sup> Moreover, the chemical aging of dust particles can lead to the formation of soluble coatings such as nitrate and sulfate,<sup>207</sup> modifying hygroscopicity and the ability to serve as CCN and IN.<sup>208</sup>

The advent of nanotechnology has led to widespread use of nanoparticles in industrial and health applications.<sup>209</sup> Nanoparticles are suspended in air during production and use and become a component in indoor and outdoor environments.<sup>210</sup> There are various kinds of nanoparticles composed of titanium dioxide, carbon, carbon nanotubes, iron oxide, silver, gold, silica, and zinc, most of which are potentially hazardous and toxic due to the small size (<100 nm) of the particles and ability to deposit in the alveoli.<sup>176,211</sup> Nanoparticles have higher surface reactivity and can trigger ROS formation inducing inflammatory response and oxidative stress in alveolar cells and macrophages, which may take up inhaled nanoparticles and respond via release of inflammatory mediators, potentially leading to cell death.<sup>211,212</sup> Iron-containing nanoparticles can induce bacterial growth and biofilm formation, and inhibit antimicrobial peptide function.<sup>213</sup> Coal fly ash, which is a byproduct of combustion and contains various transition metals including iron,<sup>175</sup> is also shown to increase bacterial growth and impair host innate immunity.<sup>176</sup>

Volcanic eruptions are rare events, but once they happen, large amounts of SO<sub>2</sub> and volcanic ash are emitted into the atmosphere.<sup>214</sup> Inhalation of volcanic ash, whose ambient concentration reached more than 10 mg m<sup>-3</sup> in case of eruption of Eyjafjallajökull in Iceland in 2010, can have significant effects on respiratory systems including bronchitis and asthma.<sup>215</sup> For example, Monick et al. found that volcanic ash disrupted pathogen-killing and inflammatory responses in alveolar macrophages, increased bacterial replication, and decreased bacterial killing by antimicrobial peptides, although the ash had little effect on the integrity of alveolar and airway

epithelial cells.<sup>216</sup> Their study suggests that volcanic ash exposure, while not seriously compromising lung cell function, may be able to impair innate immune responses in exposed individuals.<sup>216</sup>

Nuclear accidents like those in Chernobyl in 1986 and in Fukushima in 2011 can lead to the release of gaseous and particulate radioisotopes such as <sup>131,133</sup>I, <sup>89,90</sup>Sr, <sup>134,137</sup>Cs, <sup>132</sup>Te, <sup>103,106</sup>Ru, and <sup>140</sup>Ba.<sup>217</sup> Such radionuclides are mostly associated with fine particulate matter PM2.5,<sup>218</sup> which deposit slowly gravitationally by dry deposition and more efficiently by rainfall with wet deposition, leading to human exposure and contamination of land and water on local regional and global scales.<sup>219</sup>

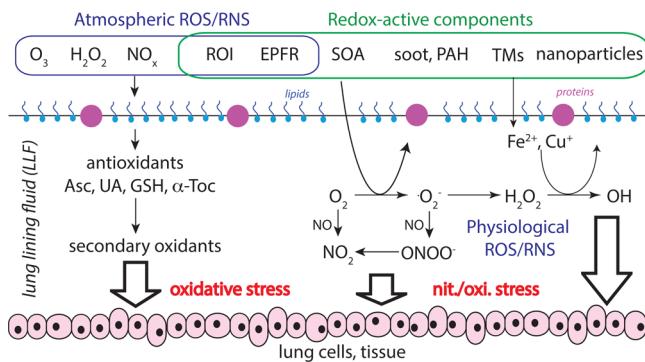
### 3. MULTIPHASE CHEMICAL REACTIONS AT SPECIFIC BIOLOGICAL INTERFACES

#### 3.1. Lung Lining Fluid

Biosurfaces, such as the lung epithelium and the leaves plasmalemma, are naturally protected against ozone by thin fluid films containing antioxidants, which scavenge ozone and ROS before they reach the underlying tissues.<sup>31,220</sup> The surface of the respiratory tract is covered with a lining fluid, extending from the nasal cavity to the alveoli. The lung lining fluid is covered with pulmonary surfactants consisting of a complex mixture of phospholipids, proteins, and cholesterols. The underlying fluid has a two-layer structure: an upper part is a mucus gel phase, which is particularly good at trapping microorganisms and large particles; a lower part is an aqueous sol phase, which bathes the lining epithelial cells.<sup>221</sup> The average thickness of the fluid is about 1–10 μm in the upper airways, about 200–500 nm in the distal bronchoalveolar region, about 15–80 nm in the vast expanse of the alveoli (but can be several micrometers thick in pooled areas).<sup>221,222</sup> Typical estimates of the total lung surface fluid volume in humans are about 15–70 mL.<sup>223</sup> The surface areas of the nasal and alveoli airways are 180 and 885 000 cm<sup>2</sup>, respectively, implying that surface processes may be more important than bulk processes.<sup>31</sup> The fluid also hosts a complex microbial community (lung microbiome) that is influenced by exchange with the environment, including bioaerosol emission and deposition (section 2.2), and influences the chemical composition and biological function of the fluid by metabolic activity.<sup>65</sup>

The epithelial or lung lining fluid (LLF) contains four major low molecular mass antioxidants: ascorbate (vitamin C), uric acid, reduced glutathione (GSH), and α-tocopherol (vitamin E). The concentrations of these oxidants are different in different regions of the respiratory tract. For example, the bronchoalveolar LLF contains 40 (±18) μM ascorbate, 207 (±167) μM uric acid, 109 (±64) μM GSH, and 0.7 (±0.3) μM α-tocopherol.<sup>224</sup> In the nasal cavity, uric acid is by far the most abundant antioxidant (100–400 μM) followed by ascorbate (10–50 μM).<sup>221,224</sup> Uric acid and ascorbate appear to be particularly important antioxidants in both the upper and the lower respiratory tract due to their high concentrations, whereas GSH may be also important in the bronchoalveolar LLF.

As illustrated in Figure 8, ozone may cause oxidative stress to the lung if not scavenged by pulmonary surfactants and antioxidants in the LLF before reaching the lung cells.<sup>225</sup> The reactivity between ozone and antioxidants is in the order uric acid ≈ ascorbate ≫ GSH with the second-order reaction rate



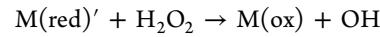
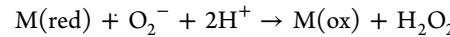
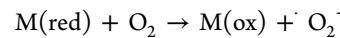
**Figure 8.** Air–lung interface: Reaction of atmospheric ROS and RNS with antioxidants (ascorbate, uric acid, reduced glutathione,  $\alpha$ -tocopherol) contained in the lung lining fluid (LLF) may lead to formation of secondary oxidants. Redox-active components, including reactive oxygen intermediates (ROI), environmentally persistent free radicals (EPFR), soot, polycyclic aromatic hydrocarbons (PAH), transition metals, and nanoparticles, can induce ROS formation *in vivo*, leading to oxidative stress and biological aging.

coefficients at pH = 7.4 of  $(1\text{--}5.8) \times 10^4 \text{ M}^{-1} \text{ s}^{-1}$  for uric acid,  $(1\text{--}5.5) \times 10^4 \text{ M}^{-1} \text{ s}^{-1}$  for ascorbate, and  $5\text{--}57.5 \text{ M}^{-1} \text{ s}^{-1}$  for GSH.<sup>221,226</sup> Note that some previous studies have reported much faster reaction rate coefficient for ascorbic acid and ozone in aqueous solutions with the order of  $10^7 \text{ M}^{-1} \text{ s}^{-1}$ .<sup>227</sup> Recently, Enami et al. have conducted a series of systematic investigations on ozonolysis of antioxidants at the air–water interface with electrospray mass spectrometry experiments.<sup>228</sup> They found that the reactivity of antioxidants including uric acid,<sup>228e</sup> ascorbic acid,<sup>228d</sup> GSH,<sup>228c</sup> and  $\alpha$ -tocopherol<sup>228b</sup> decreases by orders of magnitude when the pH decreases from 7 to 3. The ozonolysis products shift from the innocuous acids at biological pH to persistent and potentially toxic secondary ozonide, epoxide, and ROOR at low pH.<sup>228b,d,e</sup> The mean pH of LLF in healthy subjects is  $\sim 7.8$  but can decrease to pH 4.5, particularly in younger children.<sup>229</sup> LLF can be also locally acidified by inhalation of acidic aerosol particles or by preexistent pathologies such as asthma.<sup>228d,229,230</sup> Therefore, antioxidants, otherwise efficient ozone scavenger under normal physiological conditions, may gradually lose reactivity in LLF layers at lower pH, and the oxidative aggression of ozone and ROS could be still transduced across LLF by secondary oxidants generated in the ozonolysis of antioxidants and surfactant proteins.<sup>225a,228d,229,230</sup>

Reactive nitrogen species (RNS), particularly NO and NO<sub>2</sub> (NO<sub>x</sub>), can cause nitritative stress to the lung.<sup>24b,231</sup> Upon deposition of NO<sub>2</sub> into the LLF, antioxidants catalyze the hydrolytic disproportionation of NO<sub>2</sub> that leads to the formation of nitrate ion and nitrous acid (HONO).<sup>232</sup> Peroxynitrite (ONOO<sup>•</sup>), which can also be formed by reaction between O<sub>2</sub><sup>•</sup> and NO, is an important endogenous nitrating agent that is capable of nitrating tyrosine residues of proteins.<sup>78a,233</sup> Oxidative potential of ONOO<sup>•</sup> may be in part due to the pH-dependent equilibrium with HOONO, which decomposes into OH and NO<sub>2</sub>.<sup>234</sup> ROS and RNS clearly overlap and crosstalk for production, function, and decomposition.<sup>24b</sup>

Exposure of epithelial lung cells to fine particulate matter of diesel exhaust and pollen allergens leads to production and release of cytokines and morphological and functional alterations of cells.<sup>77a,b,235</sup> Decreased macrophage phagocytic activity was observed, when lung cell cultures were exposed to

$\alpha$ -pinene SOA<sup>236</sup> and primary and aged carbonaceous aerosols of diesel exhaust and wood burning.<sup>237</sup> Moreover, particulate matter contains chemical species that are capable of generating ROS upon deposition in the lung.<sup>238</sup> The ability of PM to catalyze ROS generation may be one of the first steps in the induction of oxidative stress as follows:



Here, M represents any components of PM that exhibit redox activity, including combustion generated particles,<sup>239</sup> engine exhaust particles,<sup>158c,240</sup> soot or black carbon particles,<sup>37a,144d,241</sup> transition metals,<sup>48e,242</sup> environmentally persistent free radicals (EPFRs; e.g., semiquinones),<sup>242a,243</sup> humic-like substances (HULIS),<sup>244</sup> and SOA produced from aromatics such as naphthalene.<sup>144a</sup> Oxidation by ozone leads to substantial increase in redox-cycling activity of engine exhaust particles,<sup>240</sup> which may be due to chemical functionalization and formation of ROI at the surface of aerosol particles.<sup>27</sup> Note that some studies have reported that the increase of chemical functionalities of oxidized groups at the soot surface seems to reduce its oxidative stress potential.<sup>144d,241b</sup> In any case, the toxicity and redox-cycling activity of ambient PM can be strongly affected as they chemically age in the atmosphere.<sup>37a,240</sup>

Analyzing fine and coarse PM collected at an urban and rural site in the San Joaquin Valley of California, Anastasio and co-workers investigated the generation of H<sub>2</sub>O<sub>2</sub> and OH and found that iron and copper are particularly important transition metals, which can chemically generate ROS via Fenton-like reactions in a surrogate lung fluid solution.<sup>242a,b,d,f</sup> The Fenton reaction (Fe<sup>2+</sup> + H<sub>2</sub>O<sub>2</sub>) plays fundamental roles *in vivo* and in advanced oxidation processes, but note that the chemical mechanism remains still controversial: it may proceed either via the Haber–Weiss mechanism generating Fe<sup>3+</sup> and OH, or via the Bary–Gorin mechanism generating Fe<sup>IV</sup>=O (ferryl) and H<sub>2</sub>O.<sup>49,50,245</sup> Manganese and vanadium can also produce OH under some conditions, but these metals may not produce significant levels of OH in LLF due to their low ambient concentration; the other transition metals including cobalt, chromium, nickel, zinc, lead, and cadmium do not produce OH at the low soluble metal concentrations expected from ambient PM.<sup>246</sup> In addition to concentration of transition metals, composition and concentration of antioxidants were found to be also critical in ROS production from transition metals, as antioxidants can recycle the oxidized metal back to its reduced form.<sup>246</sup>

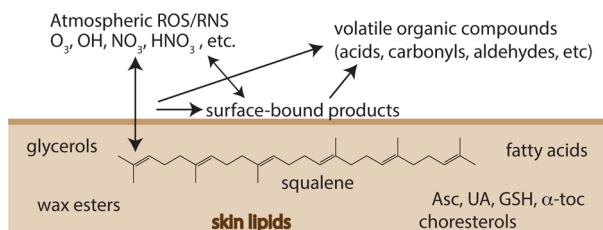
### 3.2. Human Skin

Human skin,<sup>247</sup> hair,<sup>248</sup> and clothing that contains skin oils<sup>249</sup> are further interfaces at which atmospheric oxidants and semivolatile organic compounds,<sup>250</sup> such as PAH and phthalate esters,<sup>112b,251</sup> react with biological tissues and antioxidants. Such interactions may not affect the global atmosphere but are particularly important for indoor air quality,<sup>9d,252</sup> including ozone concentration and deposition in classrooms<sup>253</sup> and aircraft cabins.<sup>249b,254</sup> The deposition velocity of ozone to skin oils is reported in the range of 0.15–0.46 cm s<sup>-1</sup>, which is

significantly higher than to glass, wood floor, and ceiling tile.<sup>249a,253,255</sup>

The outermost surface lipids of adult human skin consist of triacyl glycerols (25%), unesterified fatty acids (25%), wax esters (22%), di- and monoacyl glycerols (10%), and squalene (10%).<sup>256</sup> Similar to LLF, the stratum corneum also contains antioxidants including ascorbate, uric acid, GSH,  $\alpha$ -tocopherol, and cholesterols; however, these are much less abundant as compared to squalene and other saturated fatty acids.<sup>257</sup> Squalene, unsaturated  $C_{30}H_{50}$  with six carbon–carbon double bonds, is found to be the major ozone scavenger at the interface between room air and human envelope.<sup>247</sup> Reaction between squalene and ozone in skin oils leads to the formation of organic compounds (VOC) containing carbonyl, carboxyl, or  $\alpha$ -hydroxy ketone groups such as acetone, 6-methyl-5-hepten-2-one (6-MHO), geranyl acetone, 4-oxopentanal (4-OPA), and secondary ozonides.<sup>247,249a,258</sup> Ozonolysis of unsaturated fatty acids of wax esters and glycerols leads to formation of various kinds of aldehydes such as nonanal and dedecanal.<sup>247,249a</sup> The oxidation products have a wide range of volatility, and semi- and low-volatile products stay on skin and may be skin irritants;<sup>257,259</sup> those with high volatility desorb back to the gas phase, some of which may be respiratory irritants.<sup>260</sup> Some of the products are unsaturated and can further undergo chemical reactions with oxidants forming the array of oxidation products and SOA.<sup>247,261</sup>

Air–skin interactions are schematically summarized in Figure 9. Squalene is also reactive toward other atmospheric ROS/<sup>262</sup>



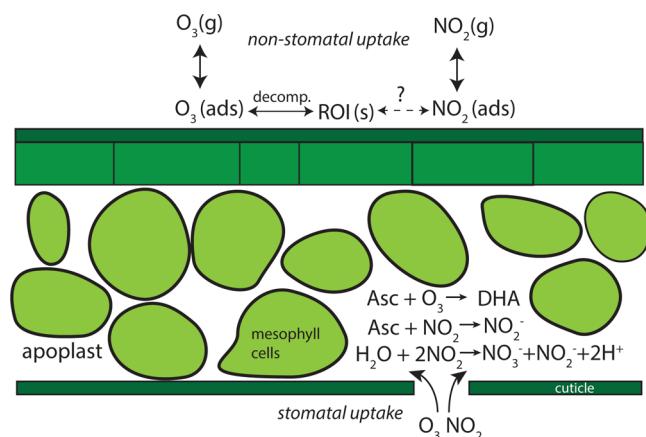
**Figure 9.** Air–skin interface: Reactions of atmospheric ROS and RNS with skin lipids and antioxidants like squalene. Reaction products include gas-phase volatile organic compounds as well as semi- or low-volatile surface-bound products, which may further react with oxidants.

RNS including  $OH$ ,  $NO_3$ , and other free radicals.<sup>262</sup> Unsaturated organic species such as squalene, related terpenoids, and unsaturated fatty acids are ubiquitous on various outdoor surfaces, including plant surfaces, soil with plant litter, airborne particles, sea surface layers, and man-made structures.<sup>247</sup> Thus, such reactions at the atmosphere–biosphere interface may affect the budget of  $OH$  and ozone, and hence oxidation capacity of the atmosphere. Reactions at various atmosphere–biosphere interfaces can be also a source of VOC, which may be further oxidized to form SOA.

Human skin is colonized and covered with a diverse collection of microorganisms such as viruses, bacteria and fungi, as well as mites.<sup>66b,c</sup> Most of such skin microbiome are harmless or even beneficial to the host, and they may play an important role in health, disease, and infection of the skin.<sup>66b,263</sup> Deposition of atmospheric ROS/RNS and reactions with skin oils may affect the nutrient source (e.g., fatty acids<sup>264</sup>) of skin microbiome, and consequently it might perturb the host–microbiome relationship and affect variation in the skin microbiome. Such aspects are highly interesting but largely unknown and subject to future studies.

### 3.3. Plant Surfaces and Cryptogamic Covers

The global surface area of plants is on the order of  $2 \times 10^{14} m^2$ <sup>15c,265</sup> and the deposition of ozone and other trace gases to plant leaves is an important process for their removal from the atmosphere, affecting their regional and global budgets.<sup>266</sup> The primary pathway of deposition occurs through uptake by stomata, which typically accounts for 1/3–2/3 of the total deposition in case of ozone.<sup>4e,267</sup> After passing through stomata, trace gases enter the apoplast, which is free diffusional space between mesophyll cells outside the cell walls as illustrated in Figure 10. Ozone diffuses through the apoplast



**Figure 10.** Air–plant interface: Stomatal and nonstomatal uptake of  $O_3$  and  $NO_2$  on plant leaves and reaction with antioxidants and water. For nonstomatal uptake, decomposition of  $O_3$  may lead to the formation of reactive oxygen intermediates (ROI). For stomatal uptake,  $O_3$  reacts primarily with ascorbate (Asc) forming dehydroascorbate (DHA).

and reaches mesophyll and the plasmalemma, which is composed of proteins, lipids, and other unsaturated compounds. Reactions of ozone and such compounds can lead to the formation of secondary oxidants and ROS, resulting in oxidative stress and damage to the plants.<sup>268</sup> Consequently, air pollution with enhanced ozone levels can cause localized cell death, enhanced ethylene emissions, the suppression of photosynthesis, and ultimately reduced tree vitality and forest diebacks, and crop yields, and a diminished diversity of ecosystems.<sup>13b,269</sup>

To prevent cellular damage from oxidative stress, plants have an antioxidative system in the cell walls, which contains millimolar concentrations of antioxidant ascorbic acid.<sup>270</sup> A number of studies have found a strong inverse correlation between plant sensitivity and levels of ascorbic acids within the plant leaf.<sup>271</sup> The primary oxidation product is monodehydroascorbate radical (MDA), which is unstable and disproportionate to dehydroascorbate (DHA).<sup>271c</sup> DHA can be recycled back to ascorbic acid in the cytosol, either via dehydroascorbate reductase in the ascorbate–glutathione cycle at the expense of reduced glutathione (GSH) and NADPH<sup>270c,272</sup> or via the direct reaction of GSH with DHA.<sup>270b</sup> The acidity of plant leaves may also affect the pathways and kinetics of ROS/RNS reaction with antioxidants as outlined in section 3.1. Detailed model simulations, considering degradation and recycling of cell wall ascorbic acid, effects of apoplast pH, and distribution and transport of ascorbic acid in the apoplast, have suggested that ascorbic acid concentration is high enough to scavenge a significant proportion of ozone taken up in the leaf interior

under environmentally relevant conditions; however, there is considerable variation between taxa in the potential degree of protection by ascorbic acid.<sup>270b,d</sup>

In the case of stomatal uptake of NO<sub>2</sub>, NO<sub>2</sub> reacts with water and ascorbic acid, yielding nitrate (NO<sub>3</sub><sup>-</sup>) or nitrite (NO<sub>2</sub><sup>-</sup>), respectively.<sup>273</sup> Reaction with ascorbic acid is the first critical step, and plants with higher leaf ascorbic acid concentration have indeed higher rates of NO<sub>2</sub> uptake.<sup>274</sup> High NO<sub>2</sub> uptake could lead to acidification and accumulation of NO<sub>3</sub><sup>-</sup> and NO<sub>2</sub><sup>-</sup>, some of which are transported from the cell wall solution to the interior of the cell. Measuring such transport rate is difficult, but the activity of the enzyme nitrate reductase can be used as an indication of transport capacity.<sup>275</sup> In addition to such replenishment of the substomatal cavity, the key enzymes of nitrite assimilation and the coordination of nutrient metabolism play crucial roles in determining the total NO<sub>2</sub> deposition.<sup>275a,276</sup> The recent field measurements by eddy covariance confirm the importance of such internal leaf resistance to NO<sub>2</sub>.<sup>277</sup>

Besides stomatal uptake, also nonstomatal uptake has become widely recognized as an important sink for reactive trace gases.<sup>4e,278</sup> It has been shown that nonstomatal flux can contribute up to about one-half of the total ozone flux depending on canopy type and seasons.<sup>267,279</sup> Leaf surfaces are coated with waxes, which are mostly triterpenoids and saturated or oxygenated alkanes without obvious reaction sites for ozone.<sup>280</sup> Some biogenic volatile organic compounds could be dissolved in waxes that may enhance the surface reactivity; however, no enhancement of surface reactions was observed in reaction of ozone and  $\alpha$ -pinene in wax surfaces.<sup>281</sup> Ozone loss on largely inert surfaces such as plant waxes, stainless steel, aluminum, and dust has been reported to proceed with similar kinetic parameters, temperature dependencies, and activation energies.<sup>179b,185,186,281</sup> The kinetic similarities suggest common reaction pathways such as the formation of ROI like atomic oxygen, epoxy, or peroxy groups.<sup>27</sup> Ozone-derived ROI may also react with other reactive species on the surface like adsorbed NO<sub>2</sub> and promote nonstomatal uptake of nitrogen oxides and other ROS/RNS.

Field measurements indicate that nonstomatal uptake of reactive trace gases is influenced by multiple factors including temperature, surface wetness, solar radiation, and wind speed.<sup>4e,279d,282</sup> Ozone deposition increases with temperature and solar radiation,<sup>279d,282a</sup> whereas both inhibition and enhancement of ozone uptake have been observed due to surface wetness.<sup>279b,282b</sup> At low humidity, the adsorption of ozone can be hindered by competitive adsorption and occupation of surface sites by water molecules,<sup>177i,s</sup> and thermal decomposition of ozone may be inhibited by water.<sup>4e</sup> At high humidity, however, deposition may be enhanced due to aqueous reactions in the water film.<sup>279d,282b</sup> Moreover, recent studies suggested that the presence of water film on plant leaves may facilitate efficient uptake of biogenic terpenes and isoprene via protonation, cationic oligomerization, and Fenton oxidation.<sup>283</sup> Such nonstomatal gas uptake and surface reaction mechanisms may also apply for stems and branches of vascular plants as well as the surface of soil organic matter and cryptogamic covers.

Large fractions of terrestrial surfaces including plants and soil are covered by cryptogamic ground covers consisting of cyanobacteria, algae, fungi, lichens, and bryophytes (mosses) in variable proportions.<sup>15a,b</sup> These communities are photoautotrophic and fix large amounts of atmospheric CO<sub>2</sub> and

N<sub>2</sub>.<sup>15c,284</sup> The contribution of cryptogamic covers to global biological nitrogen fixation on land may be as high as ~50%, suggesting that they have an impact on the sequestration of atmospheric CO<sub>2</sub> by terrestrial plants, because this process is often constrained by the availability of fixed nitrogen.<sup>15c</sup> While the fixation of nitrogen by root nodule symbionts usually occurs in bulk soil, cryptogamic covers fix nitrogen at the surface of soil and plants. The difference between surface and bulk fixation is potentially relevant to the bioavailability of the fixed nitrogen as well as for multiphase chemical reactions and the exchange of RNS like HONO at the atmosphere–biosphere interface as outlined above (section 2.1). Mosses can be used as indicators of atmospheric deposition of nitrogen,<sup>285</sup> heavy metals,<sup>286</sup> and selected persistent organic pollutants (POPs) including PAH, dioxins, and furans.<sup>287</sup> Long-term monitoring mosses have been used for assessing the influence of atmospheric deposition on ecosystems and possible effects on humans.<sup>286b,288</sup>

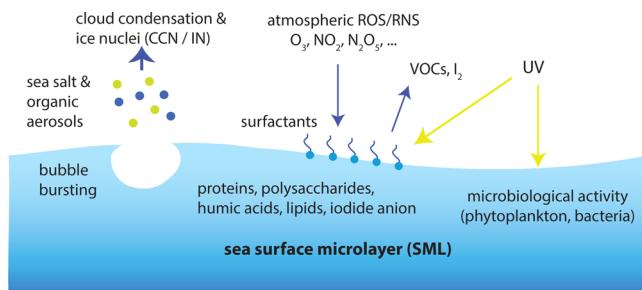
Land-use and climate change are likely to influence the geographic distribution and metabolic activity of cryptogamic covers, which may in turn affect their role in the Earth system and constitute previously unrecognized climate feedback cycles.<sup>15c</sup> In lichens, ROS can be generated by enzymes<sup>289</sup> and enhanced by external factors like desiccation.<sup>290</sup> The generated ROS can be scavenged by antioxidants such as ascorbate,  $\alpha$ -tocopherol, and glutathione.<sup>291</sup> Oxidative stress may arise if the balance between pro- and antioxidative processes is lost.<sup>290,291b</sup> Little is known and further studies are warranted about the rates, chemical mechanism, and effects of emission and deposition of ROS/RNS and other SHCC on cryptogamic covers.<sup>292</sup>

### 3.4. Soil and Aquatic Surfaces

Biotic and abiotic processes release large amounts of greenhouse gases, reactive trace gases, and aerosol particles from soil to the atmosphere, and deposition to soil surfaces is a major sink for atmospheric trace constituents.<sup>4d,e,266g</sup> Air–soil exchange plays a particularly important role in the biogeochemical cycling of nitrogen, and the release of NO from soil microbes has long been recognized as a major source of atmospheric reactive nitrogen.<sup>293</sup> Recent studies have shown that soil may also act as a major source of nitrous acid; HONO accounts for up to ~30% of the primary OH radical production, and field observations indicate large missing sources of HONO on aerosol or terrestrial surfaces.<sup>23,40a,294</sup> Potentially important chemical mechanisms of HONO formation include photolysis of nitric acid (HNO<sub>3</sub>)<sup>295</sup> and heterogeneous reactions of NO<sub>2</sub>, which may be enhanced by organic photosensitizers<sup>6f,h,j-l,296</sup> or catalysts.<sup>297</sup> One of the largest sources could be the release of HONO from biogenic soil nitrite (NO<sub>2</sub><sup>-</sup>), and soil may act as a buffer that can take up nitrous acid from the atmosphere and release it again depending on the ratio between the atmospheric concentration and the effective equilibrium concentration of HONO above soil depending on nitrite concentration, acidity, temperature, and related parameters.<sup>298</sup> Ammonia-oxidizing bacteria in soil may release even more HONO than expected from the acid–base and Henry's law equilibria of the aqueous phase in soil, and the amount of reactive nitrogen emitted as HONO from these sources is comparable with emissions of NO from arid and arable areas.<sup>299</sup> Recent investigations indicate that the surface acidity of soil minerals may influence nitrite speciation in soil, suggesting that up to 70% of global soils are capable of emitting HONO due to their acidic or close

to neutral pH.<sup>300</sup> Nitrous acid may also react with soil carbonates at night and be displaced during the day by air-to-soil transfer of hydrogen chloride and nitric acid, modulating surface acidity and the deposition or emission of nitrous acid.<sup>301</sup> Overall, the soil emissions of both NO and HONO as major sources of atmospheric RNS and ROS depend on soil water content, pH, fertilization, the occurrence of organic matter, and nitrifying and denitrifying microbes. They are a prominent example for the tight coupling of biotic and abiotic multiphase processes at the atmosphere–biosphere interface. Thus, global land-use changes, agricultural activities, and related changes in the chemical composition and microbiome of soil may affect the cycling of reactive nitrogen and the oxidizing capacity of the atmosphere<sup>298,299,302</sup> and may contribute to the effects of air quality on climate and public health in the Anthropocene.

About 70% of the Earth's surface is covered by oceans, and the air–sea exchange of trace gases and aerosols proceeds through an interface called the sea-surface microlayer (SML; Figure 11).<sup>303</sup> The SML comprises the uppermost tens to



**Figure 11.** Air–sea interface: Multiphase processes at the sea surface microlayer (SML) include the emission of sea spray aerosols via bubble bursting, heterogeneous reactions of atmospheric ROS/RNS with SML constituents (organics, halogens, etc.), and microbiological activity mediated by sunlight.

hundreds of micrometers of the aquatic phase, which can have physicochemical and biological properties distinctly different from those of the underlying water.<sup>304</sup> The SML is a complex mixture of inorganic salts and biogenic organic matter such as proteins, polysaccharides, humic acids, fatty acids, and lipids, which are produced from marine biota as exudates, waste products, or remains after their death.<sup>305</sup> Both salts and organic matter can be released to the atmosphere as sea spray aerosols generated by the bursting of bubbles from breaking waves or rainfall. Chemical composition, size, and mixing state of sea spray aerosols depend on a multitude of factors such as wind speed,<sup>306</sup> the presence of different biological species,<sup>307</sup> dissolved organic compounds and surfactants,<sup>308</sup> and the bubble film thickness upon bursting.<sup>309</sup> Sea spray aerosols smaller than 1  $\mu\text{m}$  are often dominated by organic compounds such as fatty acids,<sup>310</sup> hydroxyacids,<sup>311</sup> polysaccharides, proteins, and organosulfur compounds.<sup>308a,312</sup> For the estimation or prediction of spray organic enrichment, chlorophyll-a has been identified as a suitable biological surrogate that is observable by remote sensing and globally available for use in models.<sup>313</sup> Besides sea spray emissions, the secondary formation of sulfate aerosol and SOA from the oxidation of biogenic dimethyl sulfide and VOC emitted by phytoplankton can influence the abundance and chemical composition of CCN and IN in marine air,<sup>61a,312g,i,314</sup> but the relative contributions, importance, and climate feedback effects

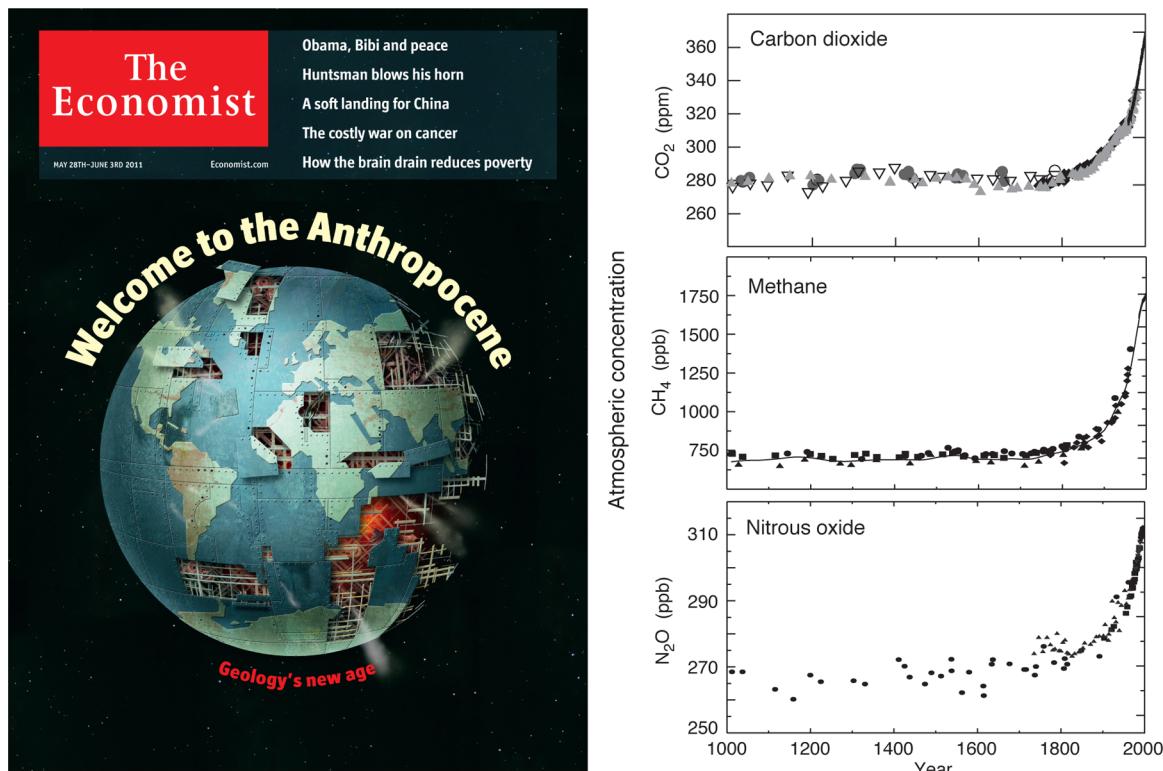
of primary and secondary components of marine aerosols are still under debate.<sup>192a,b,d,315</sup> In any case, changes in the abundance and properties of marine aerosols may influence marine clouds and their radiative effects on the Earth's energy balance and climate on global scales,<sup>8a,316</sup> and technical enhancements of sea spray aerosol production have been discussed with regard to climate geoengineering.<sup>317</sup>

The presence of organic coatings on SML and sea spray aerosol surfaces may influence gas uptake and multiphase chemistry in various ways:<sup>318</sup> (a) Coating blocks interfacial transport inhibiting water evaporation and gas uptake.<sup>103,319</sup> Note that they may not be totally prevented,<sup>320</sup> as alkyl chains can be disordered on surfaces as shown in recent surface-sensitive spectroscopy studies of marine aerosol particles.<sup>321</sup> (b) Coating prevents surface hydrolysis of impinging gas species such as N<sub>2</sub>O<sub>5</sub>.<sup>322</sup> (c) Coating enhances surface concentrations of reagents.<sup>323</sup> (d) Coating constituents react with gas species<sup>324</sup> at SML serving as a significant sink of atmospheric ROS/RNS especially O<sub>3</sub> and NO<sub>2</sub>.<sup>325</sup> Gas uptake of O<sub>3</sub> and NO<sub>2</sub> can be enhanced by light in the presence of photoactive compounds at SML.<sup>61,k,326</sup> Ozone uptake can be significantly enhanced by reaction with iodide leading to emission of gaseous I<sub>2</sub>.<sup>245a,327</sup> Heterogeneous reactions of ozone with lipid components such as polyunsaturated fatty acids and phospholipid<sup>328</sup> at SML and subsequent gas-phase chemistry can lead to formation of reactive carbonyls and a wide variety of oxygenated VOC.<sup>329</sup>

Ocean acidification caused by enhanced CO<sub>2</sub> levels in the atmosphere is a major issue of climate change<sup>330</sup> and will likely also modify the interplay of biology and chemistry in the SML, influencing aerosol and gas-exchange at the air–sea interface. In addition, large-scale changes in the chemical composition and microbiome of oceanic and other aquatic environments may also be induced or enhanced by other anthropogenic interferences such as the release of plastic waste,<sup>331</sup> fuel and oil spills from shipping traffic and accidents, industrial harvesting and mining in the seas, as well as potential marine climate geoengineering activities.<sup>317</sup> All of these factors and related anthropogenic changes may influence the cycling of marine and fresh-water-derived aerosols and their influence on regional and global climate change in the Anthropocene.

#### 4. CONCLUSIONS AND OUTLOOK

A wide range of chemical species undergo multiphase reactions at the atmosphere–biosphere interface. Particularly relevant are the following groups of short-lived health- and climate-relevant air contaminants (SHCC) as reviewed in this work: (1) reactive oxygen and nitrogen species (ROS/RNS) in the gas phase (O<sub>3</sub>, OH, H<sub>2</sub>O<sub>2</sub>, NO<sub>x</sub>, HONO, HNO<sub>3</sub>, etc.) as well as long-lived reactive oxygen intermediates (ROI) and persistent radicals on aerosol particles (tyrosyl, phenoxyl, etc.); (2) primary biological aerosols (PBA) comprising natural bacteria, fungal spores, and pollen as well as chemically or genetically modified organisms or proteins; (3) secondary organic aerosols (SOA) that are formed by oxidation of volatile organic compounds from biogenic and anthropogenic sources and undergo further chemical aging and phase transitions in the atmosphere; and (4) carbonaceous combustion aerosols (CCA) including soot, black/elemental carbon (BC/EC), polycyclic aromatic compounds, and other hazardous species like transition metals. ROS and RNS interact strongly with other SHCC and play a central role in both atmospheric and physiological processes and their coupling through the atmosphere–biosphere inter-



**Figure 12.** Examples of the resonance of the Anthropocene in public media (reprinted with permission from The Economist magazine cover on 28 May 2011; Copyright The Economist Newspaper Limited, London (May 2011)/Jon Berkeley) and of the massive changes of global atmospheric composition related to human activity and climate change over the past centuries reflecting the onset of the Anthropocene. Reprinted with permission from ref 342. Copyright 2001 IPCC.

face, for example, in the formation and aging of biogenic and combustion aerosols as well as in inflammatory and allergic immune responses triggered by air pollution. Biological surfaces usually contain antioxidants such as ascorbate in plant leaves, cryptogamic covers, and lung lining fluids or squalene in skin. Nevertheless, the deposition of atmospheric ROS/RNS and redox- or bioactive aerosol components like soot, transition metals, or micro-organisms can damage biological tissues and induce oxidative stress and affect the microbiome on biological surfaces like human lungs and skin as well as plant leaves, cryptogamic covers, and soil or aquatic surfaces. Such processes can affect higher organisms as well as surface microbiomes, which in turn can influence the chemical composition of atmosphere–biosphere interfaces by metabolic activity. The chemical mechanisms and kinetics are not yet fully understood and far from being captured quantitatively, but multiphase interactions are crucial for both biotic and abiotic processes in the Earth system. Well-designed further experimental investigations and model simulations will be required to unravel the interdependence of chemical and biological agents at the atmosphere–biosphere interface and assess their relative importance for climate and public health on regional and global scales.

With regard to globally pervasive anthropogenic air pollution and environmental change, the term Anthropocene has been established by the Chemistry Nobel Laureate Paul J. Crutzen and colleagues<sup>1b,c,332</sup> to reflect that humanity has become a driving force that determines many characteristics of the present era of Earth history, in particular atmospheric composition, land-use, and biodiversity. The term not only summarizes the perils of anthropogenic air pollution and

climate change, ocean acidification, biodiversity loss, genetic engineering, and geoengineering,<sup>333</sup> but it also highlights the great opportunities for humanity to shape its environment on planet Earth in a conscious and cautious way. At a recent symposium on the Anthropocene,<sup>334</sup> the topic and question “Tropospheric ozone in the Anthropocene: Are we creating a toxic atmosphere?” was addressed by Crutzen’s former student Jack Fishman who referred to the finding that enhanced ozone concentrations in agricultural areas lead to reduced yields of various crops like soy, wheat, cotton, and potato.<sup>13b,334,335</sup>

Since preindustrial times, the ozone background concentrations in populated continental regions appear to have increased by factors around 2–4.<sup>13a,c,336</sup> Many other SHCC have increased in similar ways or even more like aerosol particle number and mass concentrations, which are by 1–2 orders of magnitude higher in polluted urban areas as compared to pristine air in remote continental regions.<sup>4d,h,8g,14a–d</sup> Therefore, and with regard to the assessment, prediction, and handling of environmental change and public health in the Anthropocene, it seems necessary to go one step further and address the general question “Are we creating a hazardous or pathogenic atmosphere, that may severely affect public health through an enhancement of allergenic, corrosive, toxic, and infectious contaminants in indoor and outdoor air?” Such hazardous contaminants include gaseous ROS/RNS like ozone as well as particulate matter like soot and SOA. Recent developments have also raised concerns that industrially produced nanoparticles and chemically or genetically modified organisms and bioaerosols may also have adverse effects on the biosphere and public health.

Figure 12 illustrates the global resonance of the Anthropocene in public media and society, as well as massive changes of global atmospheric composition over the past centuries reflecting the onset of the new epoch. Since around 1800, human activities and climate change have led to a strong increase in the atmospheric concentrations of greenhouse gases and other trace substances like ozone as discussed above. While the atmosphere provides clear indications for global change and the “great acceleration” of global industrial activity in the second half of the 20th century, the dating of epochs in Earth history is usually linked to detectable changes in the stratigraphic record of sediments.<sup>1</sup> Thus, recent studies suggest to specifically date the onset of the Anthropocene by the world’s first nuclear bomb explosion in July 1945 and subsequent nuclear explosions leading to a clearly traceable deposition of radionuclides and decay products in the stratigraphic record of sediments around the world.<sup>337</sup> Of course, human influence on the environment goes back much further than the 20th century and the industrial revolution,<sup>338</sup> but many hundred and thousands of years ago anthropogenic activity and effects were of regional rather than global nature, and the term Paleoanthropocene has been proposed to describe and distinguish earlier periods and lower levels of human influence on planet Earth.<sup>332a</sup>

In climate science as well as in air quality research, it is often challenging to separate natural and human effects and variations. Unperturbed natural conditions are difficult to find or reconstruct from current and recent observations of the environment, which has already undergone massive anthropogenic change on global scales over the past decades and centuries.<sup>1,8a,g,332a,b,f,339</sup> All the more it is important to fully unravel and quantify the multiphase chemical processes and interactions influencing the current state as well as the history and future development of the Earth system, climate, air quality, and public health. This will require further intensified collaboration and interdisciplinary exchange across the fields of chemistry, Earth, and life science, in particular between the scientific communities of atmospheric chemistry and physics, biogeochemistry and ecology, climate and aerosol science, air quality, and health research, including epidemiology and toxicology. For the benefit of society, it will be necessary to transfer and implement the scientific knowledge into successful environmental policies and management, involving social and political scholars and practitioners.<sup>7a,340</sup> This should be pursued in analogy to the successful example of protecting the ozone layer from destruction by chlorofluorocarbons in the late 20th century (scientific discovery followed by intense research; constructive exchange between science, policy, and public; swift mitigation of the problem) rather than the gridlocked struggle for climate protection in the early 21st century (disinformation and lobbying against scientific evidence).<sup>334,341</sup> In this context, it may be helpful to emphasize that greenhouse gases and global warming are just two of many facets of global change, and that the Anthropocene is not only about mitigating negative side-effects of human activities by abstinence but also about actively using scientific knowledge and technology to protect and shape planet Earth for a sustainable development and healthy future of humanity. Thus, the Anthropocene concept may help humanity to realize: We are shaping the planet, so let us get it right.

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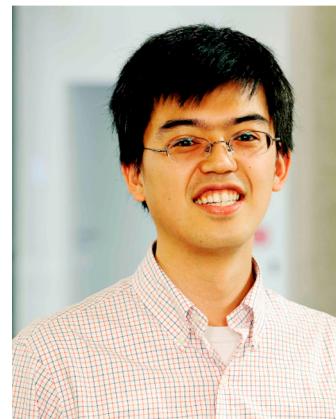
### Notes

The authors declare no competing financial interest.

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