1
Primary and Secondary Sources of Atmospheric Aerosol

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1.1
Introduction

Atmospheric aerosols are suspensions of any substance existing in the solid and/or liquid phase in the atmosphere (except pure water) under normal conditions and having a minimum stability in air assuring an atmospheric lifetime of at least 1 h. Generated by natural sources (i.e., wind-borne dust, sea spray, volcanic debris, biogenic aerosol) and/or anthropogenic activities (i.e., sulfates and nitrates from industrial emissions, wind-forced mineral dust mobilized in areas exploited for agricultural activities, fossil fuel combustion, and waste and biomass burning), aerosol particles range in size from a few nanometers to several tens of microns. As a result of internal cohesive forces and their negligible terminal fall speeds, aerosol particles can first assume sizes appreciably larger than the most common air molecules and subsequently increase to reach sizes ranging most frequently from less than 10^{-3} to no more than 100 μm (Heintzenberg, 1994). Particles with sizes smaller than 20–30 Å (1 Å = 10^{-10} m) are usually classified as clusters or small ions, while mineral and tropospheric volcanic dust particles with sizes greater than a few hundred microns are not considered to belong to the coarse aerosol class, since they have very short lifetimes. Aerosol particles grown by condensation to become cloud droplets are not classified as aerosols, although a cloud droplet needs a relatively small aerosol particle acting as a condensation nucleus for its formation under normal atmospheric conditions. Similarly, precipitation elements such as rain droplets, snowflakes, and ice crystals are not classified as aerosols (Heintzenberg, 1994). Although present in considerably lower concentrations than those of the main air molecules, aerosol particles play a very important role in numerous meteorological, physical, and chemical processes occurring in the atmosphere, such as the electrical conductivity of air, condensation of water vapor on small nuclei and subsequent formation of fog and cloud droplets, acid rains, scattering, and absorption of both incoming solar (shortwave) radiation and thermal terrestrial (longwave) radiation. The interaction processes between atmospheric aerosols and the downwelling and upwelling radiation fluxes of solar

Atmospheric Aerosols: Life Cycles and Effects on Air Quality and Climate, First Edition.
Edited by Claudio Tomasi, Sandro Fuzzi, and Alexander Kokhanovsky.

and terrestrial radiation at the surface play a major role in defining the radiation budget of our planet and, hence, the Earth's climate (Chylek and Coakley, 1974).

To give an idea of the shape of an aerosol particle suspended in dry air, a schematic representation of a particle originating from the aggregation of various kinds of particulate matter fragments is shown in Figure 1.1. It consists of several small unit structures of different chemical composition and origin (soluble acid substances, sodium chloride crystals of marine origin, ammonium sulfates, insoluble carbonaceous matter, insoluble mineral dust, and insoluble organic substances), held together by interparticle adhesive forces in such a way that an aerosol particle behaves as a single unit in suspension. Thus, the same particle often contains distinct homogeneous entities, which are internally mixed to form aggregates of different components.

The insoluble carbonaceous and organic substances often consist of gas-borne particulate matter pieces from incomplete combustion, which predominantly contain carbon and other combustion-produced materials. When the surrounding air relative humidity (RH) increases to reach values higher than 65–70%, the same particle (containing soluble substances) grows gradually by condensation of water vapor to become a water droplet in which pieces of insoluble matter are suspended, as can be seen in the (b) of Figure 1.1 (see also Hänel, 1976), while the various soluble materials reach different solution states as a result of their appreciably differing deliquescence properties. In this way, an internally mixed particle evolves assuming the characteristics of an aggregate consisting of different particulate phases. Figure 1.1 also shows that dry aerosol particles can often exhibit irregular shapes, which can considerably differ from the spherical

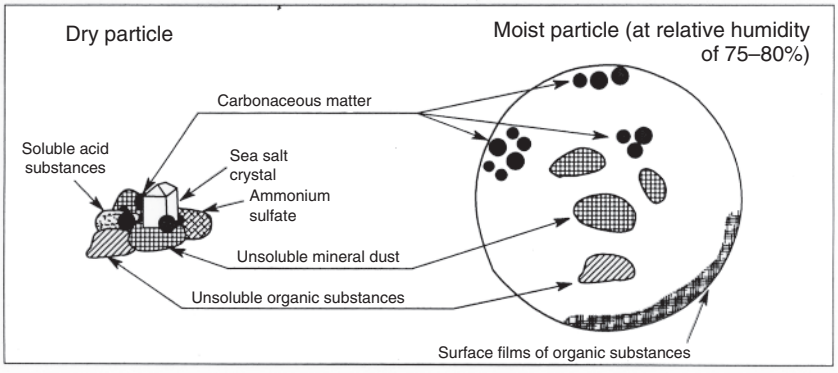

Figure 1.1 Schematic representation of an aerosol particle for dry air conditions (left) and humid air (for relative humidity (RH) = 75–80%) conditions (right), consisting of particulate matter pieces of soluble (i.e., soluble acid substances, sea-salt crystal, ammonium sulfates) and insoluble substances (carbonaceous matter, mineral dust, organic substances), which remain suspended inside the moist particle gradually growing by condensation until becoming a water droplet with soluble salts, acids, and organic compounds. (Adapted from a draft presented by Gottfried Hänel in a seminar given in 1985 at the FISBAT-CNR Institute, Bologna, Italy.)

one. Thus, the size of each real aerosol particle is generally evaluated in terms of an "equivalent" diameter a, for which the volume of such an ideal spherical particle is equal to that of the real particle.

Aerosol particles cover a size range of more than five orders of magnitude, with "equivalent" sizes ranging from 5×10^{-3} to 2.5 μm for fine particles and greater than 2.5 μm for coarse particles (Hinds, 1999). The fine particles include both (i) the so-called Aitken nuclei, having sizes mainly ranging from 5×10^{-3} to 5×10^{-2} μm, and (ii) the so-called "accumulation" particles having sizes ranging from 5×10^{-2} to about 2 μm. In this classification, it is worth mentioning that (i) the nuclei constitute the most important part of the so-called ultrafine particles (which have sizes $<10^{-1}$ μm) and mainly form through condensation of hot vapors during combustion processes and/or nucleation of atmospheric gaseous species to form fresh particles and (ii) the accumulation particles are mainly generated through coagulation of small particles belonging to the nuclei class and condensation of vapors onto existing particles, inducing them to grow appreciably. Consequently, the particle concentration within this size subrange increases, and the accumulation mode becomes gradually more evident, so named because the particle removal mechanisms are poorly efficient in limiting the concentration of such an intermediate-size class of particles. Therefore, such particles have longer residence times than the nuclei, and their number concentration tends to increase through "accumulation" of these particles within such a size class. Among the coarse particles, those having sizes ranging from 10 μm to the previously established upper limit of 100 μm are often called "giant" particles. They mainly contain man-made, sea-salt, and natural dust aerosols, being subject to sufficiently high sedimentation velocities and, hence, very efficiently removed in rather short times.

As shown in Figure 1.2, aerosols with diameters ranging from 10^{-3} to 2×10^{-1} μm can play an important role in cloud and precipitation physics, because water and ice aerosols form cloud droplets and ice crystals with diameters varying mainly from about 2×10^{-2} to more than 10^{3} μm. These growth processes lead to the incorporation of particulate matter into cloud droplets during the formation of precipitation and hence contribute to removing aerosols from the atmosphere through the so-called wet deposition processes.

Aerosols also play a fundamental role in enhancing the electricity characteristics of the atmosphere, mainly due to molecular aggregates carrying an electric charge. These particles are called ions and are divided into (i) small ions, with sizes varying from 3×10^{-4} to no more than 10^{-3} μm, and (ii) large ions, with sizes varying from 10^{-3} to about 5×10^{-1} μm. The presence of these ions determine the electrical conductivity of air. Therefore, their increase in concentration can change the magnitude of the fair weather atmospheric electric field. In the lower atmosphere, ions are mainly produced by cosmic rays and, to a lesser extent, by ionization due to crustal radioactive materials within the surface layer of the atmosphere. Ions are removed from the atmosphere through the combination of ions of opposite sign. Small ions are not much larger than molecules and have electrical mobility (defined as their velocity in an

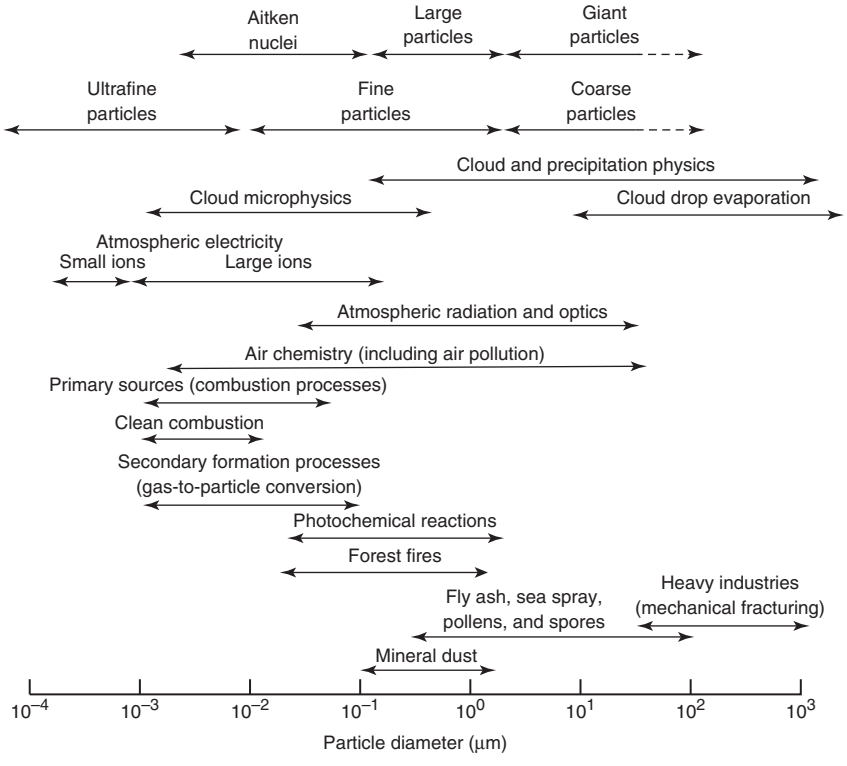

Figure 1.2 Size range of aerosol particles in the atmosphere and their role in atmospheric physics and chemistry.

electric field equal to $1\,V\,m^{-1}$) ranging from about 1 to $2\times10^{4}\,m\,s^{-1}$ at normal temperature and pressure (NTP) conditions. Conversely, the electrical mobility of large ions is very low, generally varying from 3×10^{-8} to $8\times10^{-7}\,m\,s^{-1}$. Thus, the concentration of small ions usually varies from about 40 to $1500\,cm^{-3}$ at sea level, and that of large ions from about $200\,cm^{-3}$ in maritime air to more than $8\times10^{5}\,cm^{-3}$ in the most polluted urban areas. Electrical conductivity of the air is proportional to the product of ion mobility by ion concentration, so it is generally produced by small ions in unpolluted areas. Conversely, the concentration of small ions in polluted urban areas tends to decrease as a result of their capture by both large ions and uncharged aerosols, which all exhibit very high concentrations in highly polluted areas. Consequently, the electrical conductivity of air associated with fair weather atmospheric conditions assumes the lowest values for the highest concentrations of large ions. In this view, the decrease of at least 20% in the electrical conductivity of the air, as observed over the Northern Atlantic Ocean during the twentieth century, is currently attributed to a doubling in the concentration of particles with sizes ranging from 0.02 to 0.2 μm, resulting from the increase in background pollution

conditions measured in North America and Europe (Wallace and Hobbs, 2006).

Combustion aerosols produced by forest fires have sizes ranging for the major part from 10^{-3} to 10^{-1} μm, while mineral dust particles generated by soil erosion and wind-forced mobilization present sizes mainly varying from 10^{-1} to no more than 5 μm. Figure 1.2 shows that fly ash, sea spray, pollens, and spores cover all together the size range from 5×10^{-1} to 10^{2} μm, while industrial man-made aerosols fall within the range from 5×10^{1} to more than 10^{2} μm. Chemical processes involve particles mainly generated by air pollution processes, with sizes in general varying from 10^{-3} to 10^{1} μm. More precisely, the aerosol polydispersions of different origins usually cover the following size intervals:

- From less than 10^{-3} to 5×10^{-2} μm, for aerosols generated by primary combustion processes
- From 10^{-3} to 10^{-2} μm, for aerosols produced by clean combustion
- From 10^{-3} to 10^{-1} μm, for secondary aerosols formed through gas-to-particle (g-to-p) conversion processes
- From 5×10^{-2} to about 2 μm, for aerosols originated by photochemical reactions

Aerosol radiative effects on solar and terrestrial radiation are produced more efficiently by particles with sizes ranging from 5×10^{-2} to 5×10^{1} μm, which are able to cause marked scattering and absorption of incoming shortwave radiation at wavelengths varying from 0.4 to 2.2 μm (Charlson *et al*., 1991). As a result of these interactions, nonabsorbing aerosol layers generally produce significant cooling effects, especially when poorly absorbing particles are suspended above low-reflectance surfaces, such as those of the oceanic regions (Bush and Valero, 2002). By contrast, appreciable warming effects can be induced near the surface by strongly absorbing particle layers suspended above bright surfaces, such as those covered by glaciers and snow fields in Greenland and Antarctica (Chylek and Coakley, 1974). The most intense radiative effects are mainly induced directly through scattering and absorption of incoming solar radiation, but appreciable exchanges of infrared radiation between the surface and the atmosphere can occur in the presence of dense aerosol layers near the surface, usually causing rather marked cooling effects within the ground layer. Besides these direct effects induced by aerosol particles on the shortwave and longwave radiation budget of the surface–atmosphere system (Charlson *et al*., 1992; Penner, Dickinson, and O'Neill, 1992), aerosols exert an important influence on climate inducing various indirect effects, which can appreciably modify the size-distribution curves of cloud droplets and ice crystals, enhance the liquid water content (LWC) of clouds, favor longer cloud life, and strongly influence the heterogeneous chemistry of the atmosphere (Schwartz *et al*., 1995; Jensen and Toon, 1997; Lohmann and Lesins, 2002).

The preceding remarks clearly indicate that aerosol is unique in its complexity among the atmospheric constituents and strongly influences the Earth's climate system. Airborne particulate matter is not only generated by particle direct emission mechanisms but can also form from emissions of certain gases that either

condense as particles directly or undergo chemical transformations to gaseous species, which subsequently become particles by condensation. The variety of the morphological, optical, and chemical composition properties of airborne aerosols closely depends on the formation processes of particulate matter and their subsequent aging processes occurring in the atmosphere. Considering only the formation processes of primary and secondary aerosols, the present study describes the various physicochemical processes acting as sources of marine, wind-borne (dust), volcanic, biological, combustion and anthropogenic and/or industrial aerosols, and the chemical reactions leading to the formation of secondary aerosols of both natural and anthropogenic origin. This chapter is divided into the following five sections:

Section 1.1, presenting the primary sources of natural aerosols (mineral dust, sea salt, tropospheric volcanic dust, biogenic aerosols, and forest fire and biomass burning smokes generated by natural processes).

Section 1.2, describing the formation of secondary aerosols of natural origin, like sulfate particles in the troposphere from natural SO_2 and sulfur compounds, natural nitrates from tropospheric nitrogen oxides, organic aerosols from biogenic volatile organic compounds (VOCs), and stratospheric sulfates formed from SO_2 of volcanic or marine origin.

Section 1.3, dealing with the primary sources of anthropogenic aerosols (industrial dust, fossil fuel combustion particles, including carbonaceous (soot) substances, and waste and biomass burning particulate matter).

Section 1.4, describing the main chemical processes forming secondary anthropogenic aerosols (mainly sulfates from SO_2, nitrates from NO_x, and organic aerosols).

Section 1.5, providing the most reliable estimates of the global annual emission fluxes of particulate matter associated with the various primary and secondary formation processes. The estimates were in part taken from the literature of the past 20 years and in part calculated by assuming that they agree with the most realistic evaluations of the global atmospheric mass burdens of the various types of natural and anthropogenic particles.

1.2 A General Classification of Aerosol Sources

Airborne aerosol particles are directly generated by surface sources or through a combination of physical and chemical and sometimes biological processes occurring in the atmosphere and in the adjacent reservoirs. Among these processes, three general types of sources are commonly distinguished:

1. "Bulk-to-particle (b-to-p) conversion," leading to the production of (i) mineral dust particles when the Earth's crust provides the solid base material; (ii) maritime (sea-salt) particles when the liquid base material is constituted by the natural marine water reservoirs; and (iii) biological aerosols when

the particulate solid material is furnished by plants (mainly plant debris and pollens) and animals. It is evident that a variety of physical, chemical, and biological precursors are necessary in all these b-to-p conversion processes for the division of the bulk material into particles before its emission into the atmosphere.

2. "G-to-p conversion" in which condensable vapors lead to either the nucleation of new particles or the condensational growth of existing particles. In these cases, both physical and chemical processes are necessary for the accretion of precursors (most frequently molecules), which are by themselves too small to be initially counted as particles.
3. "Combustion" processes which are assumed to constitute a third typological class of particle sources, even though they are, strictly speaking, a combination of the first two types of formation processes. The main difference between the combustion processes and the b-to-p and g-to-p conversions lies in the high temperatures at which the combustion processes take place, which facilitate the formation of such particles presenting shapes and composition features that cannot be achieved solely through the first two source processes mentioned earlier.

Emitted directly as particles (primary aerosol) through b-to-p conversion processes or originating in the atmosphere through g-to-p conversion processes (secondary aerosol), atmospheric aerosols of both natural and anthropogenic origin present composition characteristics closely dependent on their formation processes, with number concentration generally decreasing rapidly as their sizes gradually increase (Junge, 1963).

1.3 Primary Aerosols of Natural Origin

Significant natural surface sources of primary aerosol particles include the emission of sea spray, release of soil and rock debris (mineral dust) and biogenic aerosols, emission of biomass burning smoke, and injection of volcanic debris at tropospheric altitudes by violent eruptions. A negligible contribution to the overall atmospheric aerosol loading is also given by space, in the form of cosmic aerosols, but these fine particles are deemed to exert only a very weak influence on the aerosol characteristics of the high-altitude atmospheric regions, where particle concentration is always very low. Thus, cosmic rays do not substantially alter the air properties of the low stratosphere and the human environment conditions observed in the troposphere. The aforementioned primary mechanisms that generate the different types of particles formed at the terrestrial surface are each characterized by well-diversified morphological features, chemical composition, optical properties, and deposition patterns. They are described in detail in the following subsections.

1.3.1 Sea-Salt Particles

The oceans constitute the main source of sea-salt aerosols. They are estimated to originate a very large amount of particulate matter per year, including the coarse particles (having sizes $a \geq 2.5\,\mu m$), which are generally be transported over short distances because of their rapid removal due to gravitational settling. Abundance of sea-salt particles is second only to mineral dust in contributing to the overall global particulate mass content of the troposphere (Andreae and Rosenfeld, 2008), in which hygroscopic salts, such as NaCl, KCl, $CaSO_4$, and $(NH_4)_2SO_4$ provide about 3.5% of the overall tropospheric water mass. Relatively high percentage mass concentrations of various salt ions are estimated to furnish relative mass percentages equal to 55.0% Cl^-, 30.6% Na^+, 7.7% $SO_4{}^{2-}$, 3.7% Mg^{2+}, 1.2% Ca^{2+}, 1.1% K^+, and 0.7% due to other ions. Pósfai *et al*. (1995) collected a large variety of sea-salt aerosols during the Atlantic Stratocumulus Transition Experiment/Marine Aerosol and Gas Exchange (ASTEX/MAGE) field campaign undertaken in June 1992 over North Atlantic and studied their morphological characteristics using transmission electron microscopy (TEM) techniques. They found that oceanic aerosols may have different composition features in clean, intermediate, and dirty samples. The major species present in clean samples included NaCl molecules with mixed cations (Na^+, Mg^{2+}, K^+, and Ca^{2+}), sulfate ions, and to a lesser extent $NaNO_3$, presenting uniform composition features of sea-salt mode particles. The excess in sulfate and nitrate concentrations is reasonably due to the oxidation of SO_2 in the sea-salt aerosol water and the reactions of NO_x with NaCl. The same compounds were also found to be present in intermediate samples in which compositional groups characterized by low and high losses of Cl^- ions from sea salt were distinguished, with most Cl^- losses compensated by $NaNO_3$ formation. Several compositional groups were found in the dirty samples, including Na_2SO_4 (with minor contents of Mg, K, and Ca), $(NH_4)_2SO_4$, and silicates, in addition to the particle types present in clean and intermediate samples. The distinct compositional groups monitored in the dirty samples revealed that long-range transport of continental air masses has favored the mixing of aerosols, while ozone oxidation and cloud processing could have contributed to the formation of excess sulfate in such samples.

During the Aerosol Characterization Experiment 2 (ACE-2) conducted in summer 1997 over the North Atlantic, Li, Anderson, and Buseck (2003a) found that the major maritime aerosol types include fresh and partly or completely reacted sea salt consisting of NaCl, mixed cations (Na, Mg, K, and Ca), sulfate (Na_2SO_4), and nitrate ($NaNO_3$), confirming the Pósfai *et al*. (1995) evaluations. In addition to the aforementioned marine components, particles of industrial origin, including $(NH_4)_2SO_4$, soot, fly ash, silica, Fe oxide, and $CaSO_4$, were found in the samples, together with minor mineral dust contents. Li, Anderson, and Buseck (2003a) also pointed out that (i) only a sea-salt mass fraction of 0–30% remains unreacted along the Atlantic Ocean coasts of southern Portugal with the anthropogenic aerosol transported from Europe – while the rest was

partly reacted or converted to sulfates and nitrates – and (ii) the sea-salt mass fraction sampled at Punta del Hidalgo (Canary Islands) was much less affected by industrial pollution, with only 5% of the particles that were completely reacted, demonstrating that the dilution of pollution varies considerably as a function of the distance of samplings from sources.

More generally, in the most remote areas of our planet, just above the ocean surface, sea salts are generally found to dominate the mass of both submicrometer and supermicrometer particles. Sea-salt aerosols are generated by various physical processes, especially the rising of entrained air bubbles to the sea surface and the subsequent bursting of such bubbles during whitecap formation, through effectiveness features that strongly depend on wind speed (Blanchard and Woodcock, 1957). These aerosol particles are often the dominant cause of solar light scattering and the main contributor of cloud nuclei in the atmosphere above the most remote oceanic regions, provided that wind speed is high enough and the other aerosol sources are weak (O'Dowd *et al*., 1997). In fact, sea-salt particles can grow considerably as a function of RH due to their usually high hygroscopic properties (Pósfai *et al*., 1998) and often act as very efficient cloud condensation nuclei (CCNs), creating major cloud nucleating effects (O'Dowd *et al*., 1997). Therefore, the characterization of the maritime aerosol production processes occurring at the oceanic surface is of great importance to achieve correct evaluations of their indirect chemical effects in the marine atmosphere, especially those induced by particles with diameters $a < 200$ nm (Leck and Bigg, 2005).

The maritime particles are ejected into the air through the bubble bursting mechanism occurring at the ocean surface during whitecap formation (Monahan, Spiel, and Davidson, 1986), as can be seen looking at the schematic sequence presented in Figure 1.3. It shows that bubbles with $a \geq 2$ mm first reach the ocean surface (in parts (1)–(3)), each of them ejecting 100–200 film droplets into the air when the upper portion of the air bubble film bursts (see part (4)). These small "film droplets" subsequently evaporate, leaving behind sea-salt particles with $a \leq 0.3$ μm (as can be seen in part (5)). One to five larger drops break away from each jet that forms when a bubble bursts (as shown in part (6)), and these jet drops are thrown about 15 cm up into the air. Some of these drops subsequently evaporate and leave behind sea-salt particles with $a > 2$ μm, containing not only sea salts but also organic compounds and bacteria that are already present in the surface layer of the ocean. This is due to the fact that the surface microlayer of the ocean is enriched in microorganisms, viruses, and extracellular biogenic material, which can enter the atmosphere through such a bubble bursting mechanism. Consequently, sea-salt particles usually contain about 10% organic matter (OM) (Middlebrook, Murphy, and Thomson, 1998), but currently it is not well known whether these biogenic constituents are internally mixed with sea salt or whether they also form agglomerate pools of externally mixed organic particles (Bigg and Leck, 2008).

As a result of the mechanisms described in Figure 1.3, sea-salt particles cover a wide size range from about 0.05 to 10 μm, presenting in general bimodal size-distribution curves with a first mode centered at $a_c \approx 0.1$ μm and consisting of

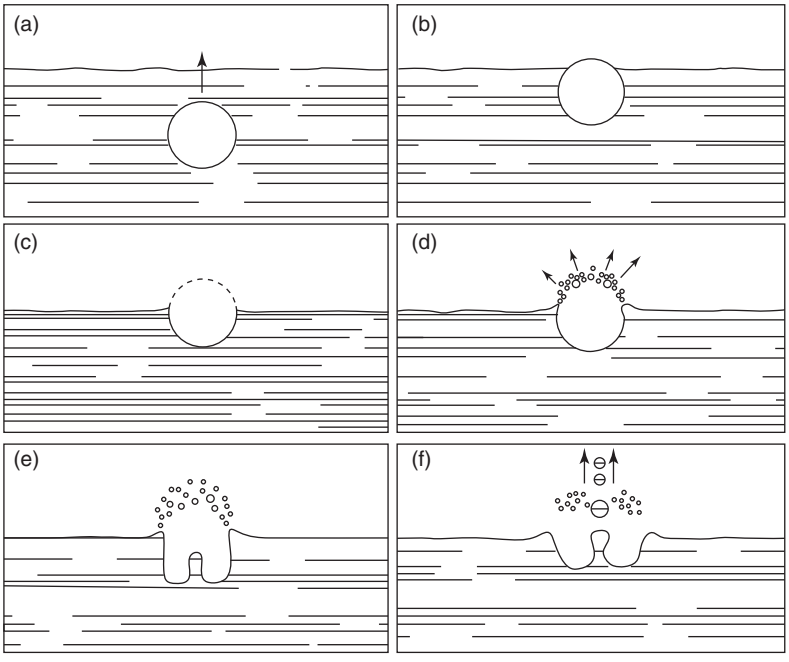

Figure 1.3 Schematic sequence of the various phases through which the film droplets and jet drops are produced when an air bubble bursts at the sea surface: (a) the air bubble is coming to sea surface; (b) the air bubble reaches the surface; (c) the sea water film starts to break; (d) droplets of ~5–30 μm diameter form when the upper portion of the bubble film bursts; (e) film droplets start to evaporate leaving sea-salt particles and other materials in the air; and (f) when the bubble bursts, 1–5 large drops (of sizes equal to about 15% the air bubble diameter) break away from the jet formed during the bubble burst. The time between phases (c) and (f) is ~2 ms.

particles originated from film drops and the second mode centered at $a_c \approx 2.5\,\mu m$ and containing particles forming from the jet drops (Mårtensson *et al*., 2003). These particles exhibit a wide range of lifetime Δt_L, depending on the large variety of sea-salt particle sizes. In fact, the largest droplets fall rapidly to the ground within their area of origin, while only the smallest aerosol particles formed at the ocean surface play a major role in determining maritime aerosol properties on a large scale. The particles are originated by bubble bursting and have sizes ranging approximately from 0.1 to 1 μm, therefore having residence times in the atmosphere long enough to allow sampling in high concentrations even at continental sites (Sinha *et al*., 2008).

The average global value of sea-salt particle production in the oceanic regions is estimated to be close to $100\,cm^{-2}\,s^{-1}$, including the large drops formed from windblown spray and foam. As mentioned earlier, the coarse sea-salt particles have rather short lifetimes Δt_L in the air, due to their large sizes. Some scanning electron microscopy (SEM) images of maritime aerosol particles consisting of pure sea-salt (halite) crystals or containing NaCl and other sea-salt cubic crystals alone

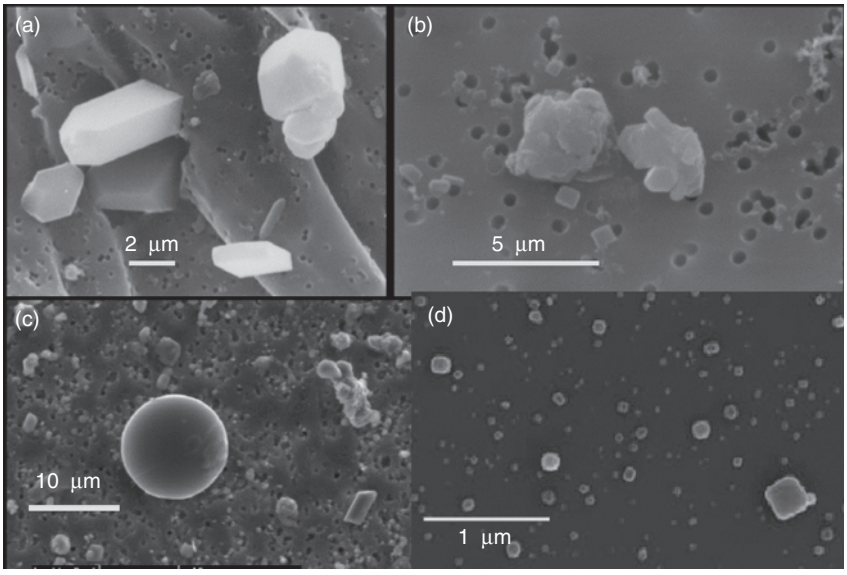

Figure 1.4 SEM images of sea-salt and other maritime particles: (a) particles sampled off the shore of Sardinia (Italy); (b) aggregates of sea-salt (halite) particles and numerous small sea-salt crystals of about 1 μm sizes, with a large-size (~4 μm) particle on the left, containing sea-salt crystals and mineral dust, and a large-size (~4 μm) particle on the right, consisting of various sea-salt crystals; (c) sea-salt and anthropogenic particles sampled off the shore of Malta (Mediterranean Sea), including some sea-salt crystals of cubic shape and a larger irregular-shaped particle (on the right) formed by aggregation of marine particles, together with a large-size spherical particle (having a diameter of ~14 μm) in the middle, presumably due to coal combustion (from the intense ship traffic in the Sicily Channel, near Malta); and (d) several submicron sea-salt cubic crystals sampled near the island of Malta. (Reproduced with permission of Alessandra Bonazza, ISAC-CNR Institute, Bologna, Italy.)

or aggregated with sulfate and nitrate particles are shown in Figure 1.4, as obtained by examining some particulate samples collected at various sites in Sardinia (Italy) and in the Sicily Channel, near the island of Malta. Interesting SEM images of sea-salt cubic crystals of various sizes and aged sea-salt particles have been shown by Sinha *et al*. (2008), obtained for samples collected in the surroundings of Mainz (Germany), that is, in an area very far from the Atlantic Ocean. SEM images of sea-salt particles have also been provided by Li, Anderson, and Buseck (2003a), sampled at Sagres (southern Portugal) and Punta del Hidalgo (Canary Islands, Spain) during the ACE-2, which show evidence of the morphological characteristics of (i) a sea-salt particle consisting of euhedral NaCl with tabular Na_2SO_4, (ii) a particle of euhedral NaCl with mixed-cation sulfate rims, and (iii) a completely converted sea-salt crystal consisting of Na_2SO_4 and $NaNO_3$.

The dry sea-salt particles transported away by winds can very easily form solution droplets in all cases where RH exceeds 65–70%. Ambient gases (e.g., SO_2 and CO_2) are also taken up by these droplets, changing their ionic composition.