*Review*

# AEROSOLS, ORGANIC MATTER, AND IMPACT ON CLIMATE

D. Čeburnis [a,b], J. Ovadnevaitė [a], K. Kvietkus [a], V. Remeikis [a], and V. Ulevičius [a]

[a] *Institute of Physics, Savanorių 231, LT-02300 Vilnius, Lithuania*
E-mail: jurgita@ar.fi.lt
[b] *Department of Experimental Physics, National University of Ireland, Galway, University Road, Galway, Ireland*

Anthropogenic aerosols are intricately linked to the climate system and to the hydrologic cycle. The net effect of aerosols is to cool the climate system by reflecting sunlight. Light-absorbing aerosols can also cool the surface, but warm the atmosphere. All these aerosol processes affect temperature profile of the atmosphere and, along with the role of aerosols as cloud condensation nuclei, affect the hydrologic cycle, through changes in cloud properties and cover as well as precipitation. Aerosol feedback processes have been proved to be difficult to model due to the spatial, temporal, and morphological complexity of aerosols. Continuous observations from satellites, international field experiments, networks of ground-based measurements are required to accurately study aerosol distribution, composition, and transformation mechanisms. Industrialization and expanding population significantly contribute to the organic nature of aerosols. The warming climate has an impact on the Earth's biological activity, which in turn is significantly contributing to the aerosol organic composition. The present paper reviews existing knowledge of organic aerosol, its importance to global climate modelling, and defines priority research areas.

**Keywords:** aerosols, organic matter, climate forcing, biogenic emissions, primary and secondary sources

**PACS:** 91.40.Dr, 42.68.Jg, 92.60 Mt, 92.20 Bk

## 1. Introduction

During the last century, the Earth's surface temperature increased by 0.6 °C, reaching the highest levels in the last millenium [1]. This rapid temperature change is attributed to a shift of less than 1% in the energy balance between absorption of incoming solar radiation and emission of thermal radiation from the Earth's system [2]. Greenhouse gases and aerosols are the most important factors that affect the global climate [1]. Whereas greenhouse gases reduce the emission of thermal radiation to space, thereby warming the surface, aerosols mainly reflect and absorb solar radiation and modify cloud properties, thus cooling the surface (Fig. 1).

Greenhouse gases, such as carbon dioxide ($CO_2$) and methane ($CH_4$), have a lifetime of up to 100 years in the atmosphere and a rather homogeneous distribution around the globe, which makes modelling exercises rather precise and well defined. The main uncertainty of the impact of greenhouse gases on climate comes from insufficient knowledge of natural sources and sinks of greenhouse gases, which makes future forecast imprecise [3].

Tropospheric aerosols, in contrast, exhibit heterogeneous spatial and temporal distribution around the globe, which results from their short lifetime of about a week. Therefore, continuous *in situ* measurements are needed to observe emission and transport of dense aerosol plumes downwind of populated and polluted regions (urban haze), regions with vegetation fires (smoke), deserts (dust), and primary aerosol particles produced over oceans (sea salt). Fine particles have sizes close to wavelengths in the visible light range and are thus expected to have a stronger climatic impact through light scattering than larger particles. Scattering of light by aerosol particles is a direct effect of aerosols on climate. In addition, fine aerosols are transported far from their source regions and their climatic and environmental impact is, therefore, de-localized compared to the emissions areas. Since most aerosols are highly reflective, they raise our planet's albedo, thereby cooling the surface and effectively offsetting greenhouse gas warming between 25 and 50% [4, 5].

However, aerosols containing black carbon are dark and therefore strongly absorb incoming sunlight, both warming the atmosphere and cooling the surface. These two processes occur simultaneously and reduce the atmosphere's vertical temperature gradient, and therefore are expected to cause a decline in
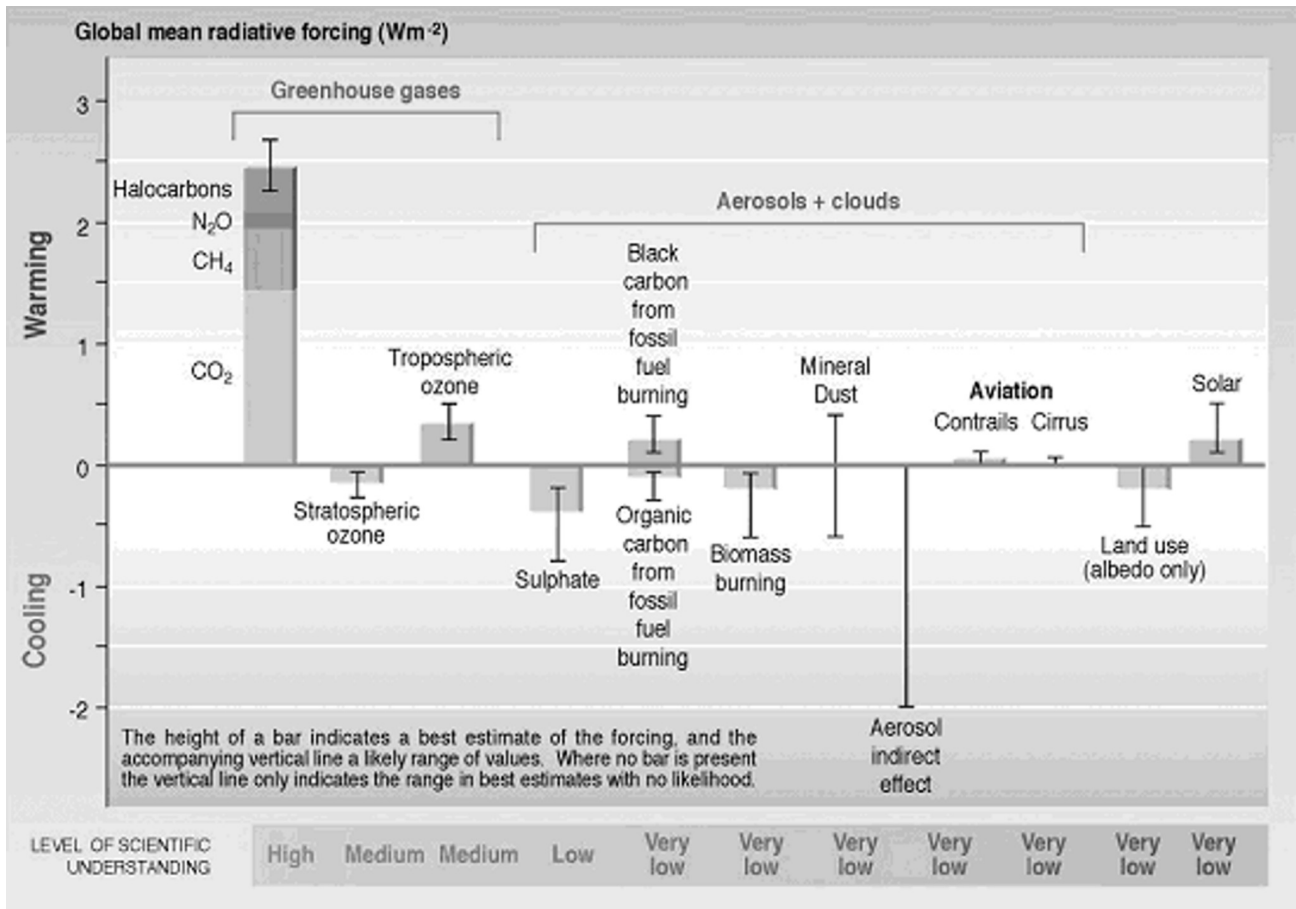
Fig. 1. Anthropogenic and natural forcing of the climate for the year 2000, relative to 1750 [1].

evaporation and cloud formation. In literature they are referred to as semi-direct effect of aerosols on climate. Much more uncertain is the way how aerosols affect clouds and precipitation. Aerosol effects on climate through clouds and precipitation are called the first and the second indirect effect, respectively. Clouds themselves are important regulators of the Earth's radiation budget, because about 60% of the surface is covered with clouds. In polluted regions, the numerous aerosol particles share the condensed water during cloud formation, therefore reducing the cloud droplet size by 20–30%, causing an increase in cloud reflectance of sunlight by up to 25% [2, 6], and cooling the Earth's surface (the first indirect effect or the Twomey effect). The smaller, polluted cloud droplets are less efficient in producing the precipitation thus affecting the precipitation regimes and ultimately the hydrological cycle (the second indirect effect or the cloud life-time effect). To assess the aerosol effect on climate, we first need to distinguish natural aerosols from anthropogenic. The ability of satellites to observe the spatial distribution of aerosols and to distinguish fine particles from the coarse ones can be exploited to separate natural and an-

thropogenic aerosols. *In situ* measurements of aerosol composition and size, as well as models that assimilate the measurements and information on population density and economic activities, are needed to further quantify the anthropogenic aerosol component and to relate it to specific sources [7].

Typical background aerosol comprises primarily produced sea salt particles and secondarily produced particles consisting of non-sea salt (nss) sulphate and organic compounds. Organic material significantly contributes to the total fine aerosol mass at continental mid-latitudes, ∼20–50% [8, 10], and almost 90% in tropical forested areas [9]. Organic compounds that are emitted directly in particulate form are referred to as primary organic aerosol (POA). Many gas-phase organic compounds undergo oxidation in the gas phase to yield products that have sufficiently low vapour pressures so they will partition themselves between the gas and aerosol phases. Such compounds are often referred to as secondary organic aerosol (SOA). More generally, POA and SOA are identified according to the manner by which they enter particulate phase. If organic species enter aerosol phase without prior

chemical reactions, they are called POA regardless of the formation mechanism [10]. However, *in-situ* measurements are often not able to discriminate between POA and SOA, and aerosol calculations or models including knowledge-based description of aerosol components and processes have to be verified against remote sensed techniques. The processes that have to be considered in climate models to account for the organic aerosol (OA) and its climatic impact comprise physics, chemistry, and biology. Physical processes that determine OA mass and size distribution are the emissions of primary OA and SOA precursors, followed by the transport by advection, convection, and diffusion, the mixing with other particles as well as the wet and dry removal in clouds and below clouds. Chemical processes include chemical formation and transformation of OA by homogeneous and heterogeneous reactions on particle surfaces or / and in clouds. Biological processes produce or emit POA and SOA precursors and exhibit significant spatial, temporal, and seasonal variability that should be taken into account in order to quantify physical processes.

## 2. Regional variability of aerosols and direct aerosol effect

Most aerosols are regional in nature owing to their short lifetime, the regional distribution of sources, and the variability in their properties. Seasonal meteorological conditions determine how far aerosols are transported from their sources as well as how they are distributed vertically through the atmosphere. Long-range transport has long been recognized as a key process in determining air quality on a regional scale. Figure 2 shows a few examples of typical aerosol spectra (aerosol volume concentration versus aerosol size) in different regions. Aerosol spectrum usually consists of two modes: accumulation or fine particle mode, which is usually a product of ultra-fine particles processed by clouds (via aqueous chemical reactions within a cloud when soluble gaseous species like sulfur dioxide, sulfuric or nitric acid dissolve into a liquid aerosol particle), and coarse particle mode, which is a product of mechanical processes as the soil erosion, wind blown dust, or bubble bursting at the sea surface. Ultra-fine particle or Aitken mode is always present close to or within natural or anthropogenic sources emitting gaseous precursors needed to form particles through gas-to-particle conversion. However, Aitken mode particles, despite prevailing in their number, do not have impact on cli-

mate until they grow into radiatively active sizes of at least 0.1 $\mu$m in diameter [11].

Aerosol spectral and spatial distribution can be assessed remotely by measuring aerosol optical thickness (AOT). AOT describes attenuation of sunlight by a column of aerosol, and serves as a measure of aerosol column concentration (Table 1). AOT is significantly different across different regions, but less than one would expect from the difference in spectra. This is because AOT is a product of the aerosol size and number. For instance, aerosol size distribution is significantly different in Eastern USA and Southeast Asia (Fig. 2), but AOT is similar in both regions (Table 1). The lowest AOT is over the oceans, where airborne particles are effectively removed from the atmosphere by wet deposition during long transport periods and due to the absence of pollution sources.

Urban and industrial pollution is mainly characterized by fine hygroscopic particles found downwind of populated regions, where regional increase in AOT is obvious. In China, for instance, economic growth and population expansion increased AOT from 0.38 in 1960 to 0.47 in 1990 [12]. However, the more effective clean-air regulations in Europe and the decline in the economy with the political transition in Eastern European countries in the late 1980s and early 1990s revealed, for example, a lower local planetary albedo due to reduced aerosol loading and related effects on clouds [13, 14].

Smoke from vegetation fires is dominated by fine organic particles with varying concentrations of light-absorbing black carbon. On average, 12% of African smoke AOT (Table 1) is due to absorption by black carbon as a result of fires of thin African grasses [15]. In contrast, only 5% of boreal fire smoke AOT is attributed to absorption by black carbon due to long smouldering stage in forest fires [16]. Smoke is less hygroscopic than regional pollution aerosol, with less AOT associated with water [1].

In remote areas AOT is dominated by coarse particles of either dust or sea spray. Dust is emitted from lakebeds in the Sahara, East Asia, and Saudi Arabia deserts that were flooded in the Pleistocene era [17]. Almost no dust is observed from Australia, where the topography is mostly flat, because the arid regions are old and highly weathered. Dust AOT is dominated by coarse particles with varying concentrations of iron oxide. However, African dust transported to Florida contains high concentrations of fine particles during summer and exceeds local pollution standards on particulate matter. Dust from East Asia, from both natural
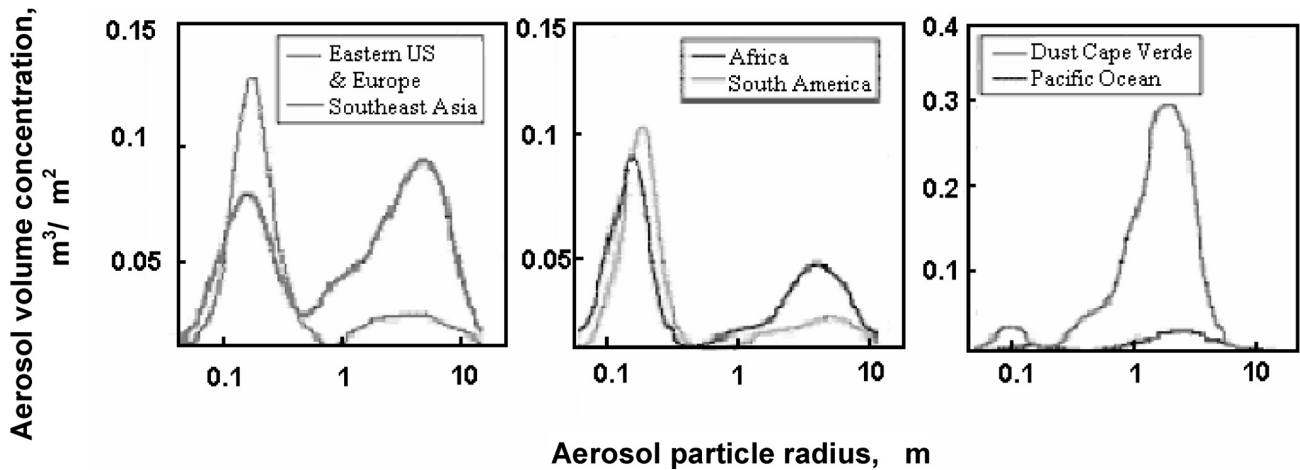
Fig. 2. Typical microphysical aerosol parameters across the globe [7].

Table 1. Aerosol optical thickness (at the wavelength of 0.55 $\mu$m) as a measure of the aerosol column concentration of different aerosol types [7].

| Region | AOT | AOT$_{fine}$, % | % absorption of AOT |
|---|---|---|---|
| *Regional pollution aerosol* | | | |
| Eastern USA and Europe | 0.20 | 94 | 3–6 |
| Southeast Asia | 0.20 | 95 | 12 |
| Central America | 0.30 | 90 | 12 |
| *Biomass burning* | | | |
| Boreal / tropical forest | 0.25–0.45 | 95 | 5–7 |
| Savanna (Africa / South America) | 0.25–0.50 | 92 | 12 |
| *Dust* | | | |
| Sahara / Saudi Arabia | 0.2–0.4 | 25 | 5 |
| *Oceanic* | | | |
| Pacific Ocean | 0.06 | 67 | 2 |
| North Atlantic Ocean[1] | 0.12 | 0.65* | 2 |

[1] data for North Atlantic from Jennings [23].

* Ångström exponent, which is related to a fine particle contribution to AOT.

sources and land use, is elevated to a height of 3–5 km with a pollution layer under it at 0–2 km [18]. On its way to North America, dust deposited in the Atlantic and Pacific Oceans provides key nutrients, such as iron, to oceanic phytoplankton [19]. Oceanic aerosol has been long considered as being composed of coarse sea salt particles produced by bubble bursting in windy conditions and fine sulphate particles produced from oceanic emissions of dimethylsulfide [20]. However, more recently it has been recognized that sea salt contributes to the fine particle content as well [21]. The latest report demonstrated that significant portion of marine aerosol consisted of organic compounds, which will be discussed bellow [22]. Marine aerosol generally absorbs very little sunlight – its AOT is estimated to average 0.06–0.12 in most regions (Table 1), but increases in the windy regions up to 0.4, as in the North Atlantic [23].

Direct aerosol effects on climate are relatively straightforward to calculate because the ways in which various particles scatter and reflect sunlight are well known. Radiative effects resulting from an increase in the concentration of anthropogenic aerosol or greenhouse gases, called radiative forcing, cause a net change in the Earth's absorbed and emitted solar and thermal energy and therefore are the basic ingredients of climate change. Current models of the climate system show a direct relationship between radiative forcing and average global surface temperature, which rises by 0.4–1.2 °C for every 1 W/m$^2$ of forcing [1]. Knowledge of the vertical distribution of aerosols and clouds is needed to calculate the impact of aerosol radiative effects. With the launch of space lidars in the next few years, the global measurements of the aerosol and cloud profiles will be available to assess aerosol radiative forcing [7].
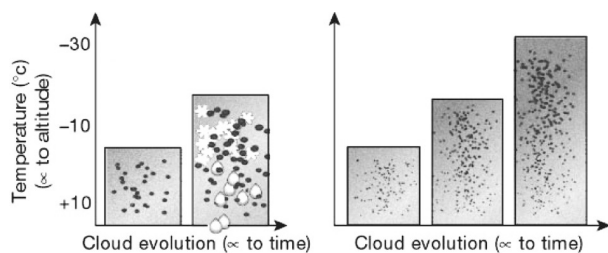
Fig. 3. Schematic diagram of cloud formation in the clean and polluted atmosphere [7].

## 3. Indirect aerosol effects

Each cloud droplet requires an aerosol particle to condense upon, otherwise clouds could not form. Thus the concentration, size, and composition of the aerosols that can act as cloud condensation nuclei (CCN) determine cloud properties, the evolution and development of precipitation. However, availability of moisture, updrafts, and cloud formation are influenced mainly by large-scale dynamic processes. Although natural aerosols are needed to form clouds, it seems that urban haze and smoke aerosols take every opportunity to reduce the formation of precipitation and affect cloud radiative properties at the same time. Figure 3 summarizes both the first and the second indirect aerosol effects. In clean atmosphere (Fig. 3(a)), the cloud droplet size increases with cloud development until it reaches the critical radius of about 15 $\mu$m and the liquid precipitation or glaciation and precipitation takes place, therefore, reducing the cloud lifetime. In polluted clouds, the availability of cloud condensation nuclei decreases the cloud droplet development to the 5–8 $\mu$m radius (Fig. 3(b)). With no precipitation even in super-cooled clouds, they exist longer and are brighter as availability of CCN makes resulting cloud droplets smaller. The first indirect effect or Twomey effect is much better understood, because the second indirect effect or cloud lifetime effect is a consequence of the first indirect effect, therefore the uncertainties associated with the cloud droplet development propagate into the large-scale cloud dynamic processes.

The presence of the light-absorbing aerosol can also affect cloud properties. Heating the lower troposphere by absorption reduces the cloud formation, the effect referred to as a semi-direct effect. Higher atmospheric temperatures reduce convection, thereby causing a decrease in the updraft speed and the amount of liquid water available to form the cloud [24].

General knowledge about aerosol effects on clouds and the hydrological cycle is still very rudimentary. The observations of the hydrological cycle are less complete than those of global mean temperature and the physical constraints are weaker, so it will be substantially harder to quantify the range of possible changes in the hydrological cycle [25]. Therefore, clearly more research in terms of field experiments, laboratory studies, and modelling efforts is needed in order to understand and quantify the effect of anthropogenic aerosols on clouds and the hydrological cycle. This is especially important because cloud feedbacks in climate models still represent one of the largest uncertainties. There is still no consensus on whether clouds provide a negative or positive climate feedback in response to a doubling of carbon dioxide. It is largely because of these uncertainties in cloud feedback that the uncertainty range of the increase in the global mean surface temperature in response to doubling of $CO_2$ varies between 1.5 and 4.5 °C.

## 4. Organic aerosol and global climate modelling

During the last decade important advances were made in understanding OA and its behaviour in the atmosphere. Different studies have shown that biogenic volatile organic compounds contribute to the formation of SOA in the atmosphere [26, 27]. Emissions of primary organic particles and also SOA precursors can occur from various sources in the boundary layer and to a lesser extent in the free troposphere. The dry and wet removal processes of OA depend on the water solubility and size of the particles containing these compounds. Finally, the simulation of the climatic impact of these aerosols will additionally require the description (1) of the water uptake by the particles, which depends on their hygroscopic properties, and (2) of the optical properties of the OA, which also depend on the state of mixing of OA components with inorganic components and are needed for the calculation of the extinction of solar radiation [10]. Knowledge of the particle formation and growth has been summarized by Kulmala [28] and results are presented in Fig. 4. Nucleation may involve homogeneous ternary water-sulfuric acid-ammonia mixtures or may be ion-induced. The initial steps of growth include activation of inorganic clusters by soluble organic molecules, the heterogeneous nucleation of insoluble organic vapours on inorganic clusters, and chemical reactions of organic molecules at surfaces of inorganic clusters. Finally, cloud condensation nuclei form through addition of organic and sulfuric acid molecules. The nucleation and growth of new particles seem to occur almost everywhere in the atmosphere. Ion clusters are
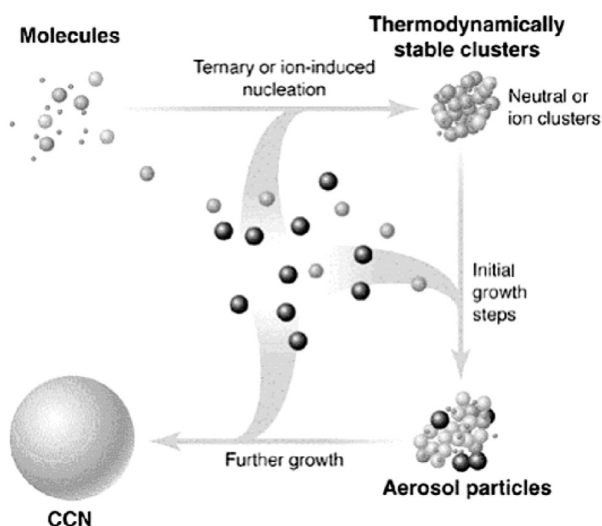
Fig. 4. Schematic presentation of particle formation and growth [28].



Fig. 5. Surface tension lowering by organics in cloud water [30].

always present, and the nucleation of new aerosol particles seems to be controlled by the very initial steps of growth and the competition between condensation and coagulation. Depending on the location, new particle production can more than double the concentration of cloud condensation nuclei over the course of 1 day. Atmospheric new-particle production is thus an essential process that must be understood and included in global and regional climate models.

After particles have grown into CCN sizes, they undergo cloud processing. Particles are chemically transformed in clouds via aqueous chemistry reactions and the resulting cloud properties modify climate indirectly. The cloud droplet activation and growth were first described by the classical Kohler theory [29], which predicts supersaturation over a solution droplet of radius $r$:

$$S = \frac{A}{r} - \frac{B}{r^3}, \quad A = \frac{2\sigma M_{\mathrm{w}}}{RT\rho}, \quad B = \frac{3n_{\mathrm{m}}M_{\mathrm{w}}}{4\pi\rho}, \quad (1)$$

where $S$ is the supersaturation, $r$ is the droplet radius, $A$ and $B$ are the constants, $\sigma$ is the surface tension of the droplet, $M_{\mathrm{w}}$ is the molecular weight of water, $R$ is the gas constant, $T$ is the temperature, $\rho$ is the density of the droplet (water), $n_{\mathrm{m}}$ is the number of dissolved moles of solute in the droplet. It is at droplet sizes and solute concentrations below and close to the critical size ($r_{\mathrm{c}} \approx 0.1$ $\mu$m to a few micrometers) that surface tension would most effectively influence whether or not a particle activates and grows into a cloud droplet of typically 5–10 $\mu$m. In the presence of water-soluble atmospheric gases ($HNO_3$, $SO_2$, etc.) the critical supersaturation for the droplet is lowered as the gas condenses into a growing droplet. However, highly sol-
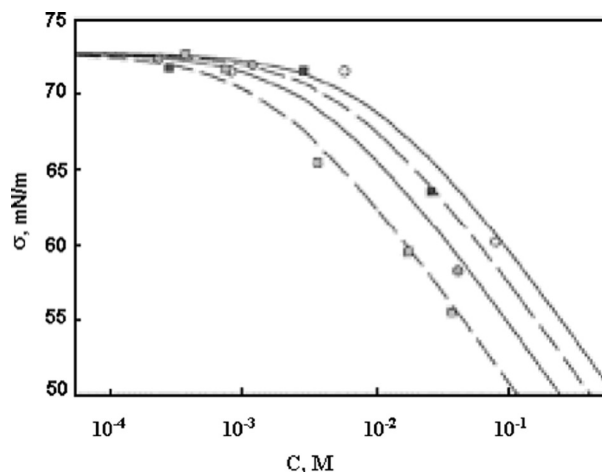
uble gases are not the only compounds that can affect the aerosol activation. The importance of carbonaceous compounds as components of atmospheric aerosol is well established. Many of these compounds are surface-active (Fig. 5) [30] and if, in addition, surface tension is lowered as the substance dissolves, the critical supersaturation is further lowered and the number of particles that can activate increases even more. In general, the lowering of surface tension associated with a dissolving substance has a stronger effect on cloud properties than the fact that the substance itself is only partially soluble, given that most water-soluble organic compounds are surface-active.

Surface tension, however, affects only one part (Kohler term) of the supersaturation equation (1). The other part of the equation includes solubility, molecular weight, and dissociation of compounds, all of which are highly uncertain at the moment. For instance, solubility of inorganic compounds is almost instantaneous, while organic compounds dissolve gradually if they are soluble at all. The molecular weight is even more uncertain parameter, because organic compounds are so diverse that some of them are not identified. A comprehensive work on the estimation of their molecular size has shown that their molecular weights are most likely in the range 200–500 Da [31], but extend up to 1000 Da. The number of dissolved moles of solute in the droplet also remains uncertain, ranging hypothetically from 2 for dicarboxylic acid up to 9 for fulvic acids [32].

Ability of organic substances to significantly modify the cloud droplet population and thus cloud albedo and cloud lifetime warrants active research to provide a full explanation of the relative contribution of SOA and POA to aerosol population. Volatile organic compounds (VOC) are emitted into the atmosphere

from natural sources in marine and terrestrial environments, as well as from anthropogenic sources. On a global basis the emissions of biogenic VOCs, which are emitted mainly by vegetation, are estimated to exceed those from anthropogenic emissions [33]. According to the rough estimates ([10] and references therein), the chemical formation of SOA may be significant as compared to the primary carbonaceous emissions (about 60% on a global scale and even more locally). However, overall estimates of emissions both of primary carbonaceous particles and of gaseous precursors of SOA are different between a factor of 2 and 5.

Recently a significant marine POA source acting during the period of high biological activity has been identified in the North Atlantic [22]. Further studies based on coherent observations are required to construct reliable POA emission inventories not only from the anthropogenic sources that are most commonly considered in modelling studies but also from biogenic sources, the importance of which remains to be determined. Particular attention should be given to accurate estimation of emissions of biogenic POA currently omitted from climatic modelling prediction. Recent experiments have demonstrated that biogenic POA exhibits significant seasonal changes. The evolution of micro-algae is driven by environmental change, particularly increasing oceanic temperatures [34], thus the production of marine POA represents a newly identified and potentially important component of the marine biota and climate feedback system involving aerosols and clouds. Figure 6 shows seasonal cycle of water-soluble organic carbon (WSOC) over the North Atlantic with a clear maximum occurring during spring through autumn.

Regardless of the origin of OA, carbonaceous compounds are highly modified in the atmosphere. Moreover, recent developments both of aerosol sampling and chemical analysis have resulted in new findings that show the formation of low volatility oligomers and polymers. The separation between the primary and secondary organic aerosol compounds becomes more difficult after the recent detection of oligomers in SOA since compounds that have been considered of primary origin might be chemically produced. Therefore, it becomes extremely difficult to quantify OA considering its sources. Experimental techniques developed for identification of the OA origin would not only help to link OA to the sources, but also make inventories of OA more reliable. One of the promising new techniques in identifying the OA origin may be the carbon isotope ratio measured in atmospheric aerosols. It is well estab-
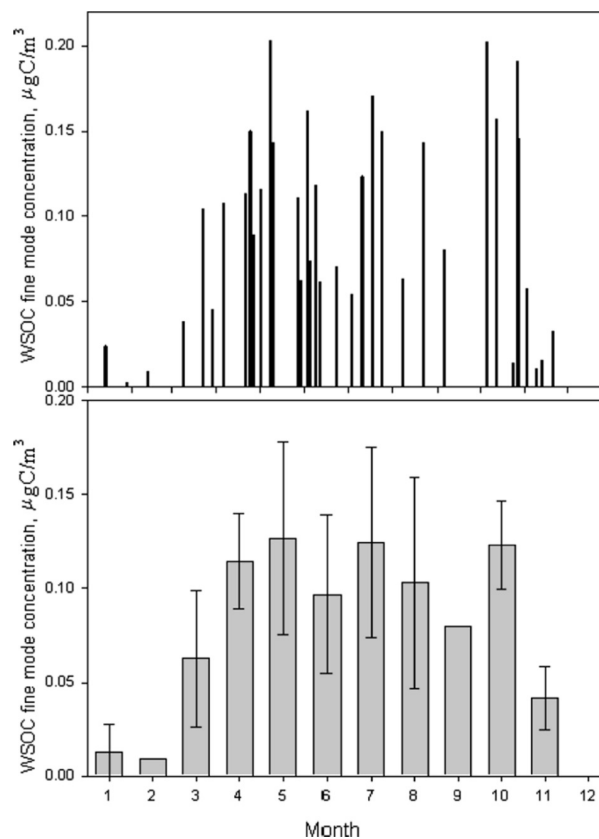


Fig. 6. Seasonal variation of the submicron WSOC concentration. Upper plot represents a time series; lower plot shows average monthly concentration $\pm 1$ standard deviation (except for months with a single value).

lished that the carbon isotope ratio varies significantly depending on whether OA has been produced by biogenic or anthropogenic processes [35]. Both $^{14}C$ and $^{13}C$ should be depleted to different degree in anthropogenic aerosol as compared to biogenic sources. A three end-member mixing model can then be used to estimate the contribution to OA from the fossil fuel, modern terrestrial sources, and marine sources. The conceptual model of the direct transfers of OC in rainfall, presented by Raymond [36], concluded that the net transfer of continental OC to the ocean could be close to zero due to a significant marine to continental flux.

The water uptake by the organic aerosol components and their interactions with the organic salts dissolved in the same aqueous solution changes the aerosol water concentration and the properties of resulting aerosol. Corresponding data for water-soluble organic compounds relevant to atmospheric applications are rather scarce, partly because organic compounds previously have not been considered important to the hygroscopic and cloud-forming properties, and also because organic compounds are more difficult to measure than inorganic compounds, especially the low

molecular weight organics due to their high volatility. Significance of organic compounds can be further stressed by the fact that organic compounds contribute to the water uptake most significantly at low relative humidity, while at high relative humidity the inorganic compounds dominate [37]. Currently there are two experimental techniques for studying aerosol hygroscopic properties, hygroscopic tandem differential mobility analyzer (HTDMA) and electro-dynamic balance (EDB), the latter being a laboratory technique. These experimental techniques coupled with model calculations should be tested against remote sensing methods to unravel a mixing state of aerosol particles, the parameter which is extremely difficult to measure directly. However, further development and application of single particle mass spectrometric methods, which became recently available, may one day give an unambiguous answer to the particle mixing state.

## 5. Conclusions

Our understanding of the organic aerosols in the atmosphere has significantly improved during the last decade, although significant gaps remain. The involvement of organics in the new particle formation deserves further studies to determine its importance on regional and global scales. Better measuring techniques are needed for carbonaceous aerosols especially with regard to the understanding of the chemical composition and the ageing processes of primary aerosol. Particular attention is needed to avoid or quantify artefacts in sampling and analysis of OA compounds. Feedback-climatic mechanisms involving organics have to be investigated since SOA from biogenic volatile organics is estimated to be the major contributor to the SOA mass. Quantification of primary production of OA needs further efforts as well. Thus significant biosphere–climate feedbacks are expected to be involved with SOA and POA existence in the atmosphere.

## References

[1] IPCC, 2001, *Climate Change 2001: The Scientific Basis, Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change*, eds. J.T. Houghton, Y. Ding, D.J. Griggs, M. Noguer, P.M. van der Linden, X. Dai, K. Maskell, C.A. Johnson (Cambridge University Press, Cambridge, UK and New York, NY, USA, 2001), 881pp.

[2] V. Ramanathan, P.J. Crutzen, J.T. Kiehl, and D. Rosenfeld, Atmosphere – aerosols, climate, and the hydrological cycle, Science **294**, 2119–2124 (2001).

[3] P. Leahy, G. Kiely, and T.M. Scanlon, Managed grasslands: A greenhouse gas sink or source? Geophys. Res. Lett. **31**, L20507 (2004).

[4] S.A. Twomey, M. Piepgrass, and T.L. Wolfe, An assessment of the impact of pollution on the global albedo, Tellus **36B**, 356–366 (1984).

[5] R.J. Charlson, S.E. Schwartz, J.M. Hales, R.D. Cess, J.A. Coakley, J.E. Hansen, and D.J. Hofmann, Climate forcing by anthropogenic aerosols, Science **255**, 423–430 (1992).

[6] Y.J. Kaufman and R.S. Fraser, Confirmation of the smoke particles effects on clouds and climate, Science **277**, 1636–1639 (1997).

[7] Y.J. Kaufman, D. Tanre, and O. Boucher, A satellite view of aerosols in the climate system, Nature **419**, 215–223 (2002).

[8] J.P. Putaud, F. Raes, R. Van Dingenen, E. Bruggemann, M.C. Facchini, S. Decesari, S. Fuzzi, R. Gehrig, C. Huglin, P. Laj, G. Lorbeer, W. Maenhaut, N. Mihalopoulos, K. Muller, X. Querol, S. Rodriguez, J. Schneider, G. Spindler, H. ten Brink, K. Torseth, and A. Wiedensohler, A European aerosol phenomenology-2: Chemical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe, Atmos. Environ. **38**, 2579–2595 (2004).

[9] G.C. Roberts, M.O. Andreae, J. Zhou, and P. Artaxo, Cloud condensation nuclei in the Amazon basin: "Marine" conditions over a continent? Geophys. Res. Lett. **28**, 2807–2810 (2001).

[10] M. Kanakidou, J.H. Seinfeld, S.N. Pandis, I. Barnes, F.J. Dentener, M.C. Facchini, R. Van Dingenen, B. Ervens, A. Nenes, C.J. Nielsen, E. Swietlicki, J.P. Putaud, Y. Balkanski, S. Fuzzi, J. Horth, G.K. Moortgat, R. Winterhalter, C.E.L. Myhre, K. Tsigaridis, E. Vignati, E.G. Stephanou, and J. Wilson, Organic aerosol and global climate modelling: A review, Atmos. Chem. Phys. **5**, 1053–1123 (2005).

[11] C.D. O'Dowd, Biogenic coastal aerosol production and its influence on aerosol radiative properties, J. Geophys. Res. **106**, 1545–1550 (2001).

[12] Y.F. Luo, D.R. Lu, X.J. Zhou, W.L. Li, and Q. He, Characteristics of the spatial distribution and yearly variation of aerosol optical depth over China in last 30 years, J. Geophys. Res. **106**, 14501–14513 (2001).

[13] O. Krüger and H. Grassl, The indirect aerosol effect over Europe, Geophys. Res. Lett. **29**, doi:10.1029/2001GL 014081 (2002).

[14] M. Wild, A. Ohmura, H. Gilgen, and D. Rosenfeld, On the consistency of trends in radiation and temperature records and implications for the global hydrological cycle, Geophys. Res. Lett. **31**, L11201, doi:10.1029/2003GL019188 (2004).

[15] T.F. Eck, B.N. Holben, D.E. Ward, O. Dubovik, J.S. Reid, A. Smirnov, M.M. Mukelabai, N.C. Hsu, N.T. O'Neill, and I. Slutsker, Characterization of the optical properties of biomass burning aerosols in Zambia during the 1997 ZIBBEE field campaign, J. Geophys. Res. **106**, 3425–3448 (2001).

[16] O. Dubovik, B. Holben, T.F. Eck, A. Smirnov, Y.J. Kaufman, M.D. King, D. Tanre, and I. Slutsker, Variability of absorption and optical properties of key aerosol types observed in worldwide locations, J. Atmos. Sci. **59**, 590–608 (2002).

[17] J.M. Prospero, P. Ginoux, O. Torres, S.E. Nicholson, and T.E. Gill, Environmental characterization of global sources of atmospheric soil dust identified with the Nimbus 7 Total Ozone Mapping Spectrometer (TOMS) absorbing aerosol product, Rev. Geophys. **40**, 1002 (2002).

[18] A.D. Clarke and V.N. Kapustin, Pacific aerosol survey, part 1: A decade of data on particle formation, transport, evolution and mixing in the troposphere, J. Atmos. Sci. **59**, 363–382 (2002).

[19] Y. Gao, Y.J. Kaufman, D. Tanre, D. Kolber, and P.G. Falkowski, Seasonal distribution of Aeolian iron fluxes to the global ocean, Geophys. Res. Lett. **28**, 29-33 (2001).

[20] R.J. Charlson, J.E. Lovelock, M.O. Andreae, and S.G. Warren, Oceanic phytoplankton, atmospheric sulfur, cloud albedo and climate, Nature **326**, 655–661 (1987).

[21] C.D. O'Dowd, M.H. Smith, I.E. Consterdine, and J.A. Lowe, Marine aerosol, sea-salt, and the marine sulphur cycle: A short review, Atmos. Environ. **31**, 73–80 (1997).

[22] C.D. O'Dowd, M.C. Facchini, F. Cavalli, D. Ceburnis, M. Mircea, S. Decesari, S. Fuzzi, Y.J. Yoon, and J.P. Putaud, Biogenically driven organic contribution to marine aerosol, Nature **431**, 676–680 (2004).

[23] S.G. Jennings, Unpublished data, National University of Ireland, Galway, Ireland (2002).

[24] U. Lohmann and J. Feichter, Global indirect aerosol effects: A review, Atmos. Chem. Phys. **5**, 715–737 (2005).

[25] M.R. Allen and W.J. Ingram, Constraints on future changes in climate and the hydrological cycle, Nature **419**, 224–232 (2002).

[26] S.N. Pandis, R.H. Harley, G.R. Cass, and J.H. Seinfeld, Secondary organic aerosol formation and transport, Atmos. Environ. **26**, 2269–2282 (1992).

[27] C.D. O'Dowd, P. Aalto, K. Hameri, M. Kulmala, and T. Hoffmann, First experimental evidence of new particle formation from organic vapours over forests, Nature **416**, 497–498 (2002).

[28] M. Kulmala, How particles nucleate and grow, Science **302**, 1000–1001 (2003).

[29] H. Kohler, The nucleus in the growth of hygroscopic droplets, Trans. Faraday Soc. **32**, 1152–1161 (1936).

[30] R.J. Charlson, J.H. Seinfeld, A. Nenes, M. Kulmala, A. Laaksonen, and M.C. Facchini, Reshaping the theory of cloud formation, Science **292**, 2025–2026 (2001).

[31] G. Kiss, E. Tombacz, B. Varga, T. Alsberg, and L. Persson, Estimation of the average molecular weight of humic-like substances isolated from fine atmospheric aerosol, Atmos. Environ. **37**, 3783–3794 (2003).

[32] M.L. Shulman, M.C. Jacobson, R.J. Charlson, R.E. Synovec, and T.E. Young, Dissolution behavior and surface tension effects of organic compounds in nucleating cloud droplets, Geophys. Res. Lett. **23**, 277–280 (1996).

[33] A. Guenther, C. Geron, T. Pierce, B. Lamb, P. Harley, and R. Fall, Natural emissions of non-methane volatile organic compounds, carbon monoxide and oxides of nitrogen from North America, Atmos. Environ. **34**, 2205–2230 (2000).

[34] J. Beardall and J.A. Raven, The potential effects of global climate change on microalgal photosynthesis, growth and ecology, Phycologia **43**, 26–40 (2004).

[35] Y.J. Yoon, D. Ceburnis, F. Cavalli, O. Jourdan, J.P. Putaud, M.C. Facchini, S. Descari, S. Fuzzi, S.G. Jennings, and C.D. O'Dowd, Seasonal characteristics of the physico-chemical properties of North Atlantic marine atmospheric aerosols, Submitted to J. Geophys. Res.

[36] P.A. Raymond, The composition and transport of organic carbon in rainfall: Insights from the natural ($^{13}$C and $^{14}$C) isotopes of carbon, Geophys. Res. Lett. **32**, L14402, doi:10.1029/2005GL022879 (2005).

[37] W.D. Dick, P. Saxena, and P.H. McMurry, Estimation of water uptake by organic compounds in submicron aerosols measured during the Southern Aerosol and Visibility Study, J. Geophys. Res. **105**, 1471–1479 (2000).

# AEROZOLIS, ORGANINIAI JUNGINIAI IR ĮTAKA KLIMATUI

D. Čeburnis [a,b], J. Ovadnevaitė [a], K. Kvietkus [a], V. Remeikis [a], V. Ulevičius [a]

[a] *Fizikos institutas, Vilnius, Lietuva*
[b] *Airijos nacionalinis universitetas, Galway, Airija*

**Santrauka**

Aerozolių ryšys su klimato kaita ir atmosferos hidrologiniu ciklu yra pakankamai painus, nes atspindėdami saulės šviesą jie sąlygoja klimato atšalimą, tačiau sugerdami energiją šildo atmosferą ir keičia temperatūros profilį, o kartu, virsdami debesų kondensacijos branduoliais, veikia hidrologinį atmosferos ciklą, debesuotumą, debesų savybes ir kritulių susidarymą. Aerozolio poveikis labai priklauso nuo jo tipo ir sudėties. Todėl sunku minėtus mechanizmus ir grįžtamuosius ryšius modeliuoti. Be to, aerozoliai yra įvairiausių tipų, pradedant nuo dykumų dulkių ir vulkani-

nių pelenų ir baigiant miestų užterštumu. Aerozolių koncentracijos smarkiai kinta laike ir erdvėje, todėl nuosekliems aerozolių pasiskirstymo, sudėties ir transformacijos tyrimams reikalingas stebėjimo stočių tinklas, Žemės palydovai ir tiksliniai tiesioginiai eksperimentai. Įvertinimą apsunkina savybių kitimas laike, nes industrializacija ir populiacijos augimas sąlygoja aerozolio prigimties kitimą – klimato šiltėjimas veikia biologinį Žemės aktyvumą, kuris savo ruožtu veikia organinę aerozolio sudėtį. Šiame darbe apžvelgtos esamos žinios apie organinio aerozolio reikšmę globaliniame klimato modeliavime ir nustatytos prioritetinės tyrimų sritys.