P3.13 SOURCES OF BIOGENIC PARTICLES OVER THE CENTRAL ARCTIC OCEAN ASSOCIATED WITH OPEN LEAD SURFACE MICROLAYER

Caroline Leck, E. Keith Bigg and Michael Tjernström
Stockholm University, Stockholm, Sweden

1. INTRODUCTION

Clouds have a profound impact on the radiation budget of the Earth and calculations show that small changes in cloud cover or optical thickness may offset – or double – the effects of even a doubling of the greenhouse gas concentrations. In contrast to the mid-latitude oceans, low-level clouds are a warming factor in the central Arctic through most of the year. In winter the effects of low-level clouds are the single most important local factor determining the stability of the lower troposphere. In summer, with frequent low clouds, changes in their microphysics can alter their reflectivity for solar radiation as well as cloud lifetimes. These processes are very poorly described in current climate models. Because of the potential for a large effect, it is essential that we should understand the sources, nature and controls on the supply of cloud droplets.

Formation of clouds requires the presence of small airborne aerosol particles, so called cloud condensation nuclei (CCN). While the amount of condensed water in a cloud is determined by thermodynamic and dynamic properties, the number of droplets is regulated by the abundance of CCN. With many CCN, the condensed water is distributed over many small droplets rather than over a few large. This in turns makes the cloud look "whiter", thus reflecting more solar radiation back to space. This is known as the "indirect effect" of particles on climate.

The well-known hypothesis of Charlson et al. (1987) proposed one biological influence on radiation and climate based on the indirect effect of aerosols. The gas dimethyl sulfide (DMS), produced by marine phytoplankton, is oxidized in the atmosphere to sulfuric acid, nucleating particles that grew to become CCN. It was suggested that climate change would change DMS production to form a negative feedback through its effects on CCN.

Sulfate-containing aerosols are ubiquitous in the atmosphere and usually the most numerous particles capable of acting as CCN, so the theory seems reasonable. But does DMS alone control the *number* of CCN or could there be other biological controls of CCN formation in marine air remote from land sources? The central Arctic Ocean in summer provides an ideal laboratory for studying this question. Excursions of continental and often polluted air into the basin are infrequent in summer, and low cloud and fog at the fringes of the pack ice rapidly remove aerosols. A shallow boundary layer capped by a temperature inversion (Tjernström et al., 2004) limit mixing from above the clouds, where long-range transported aerosols from distant sources may reside.

* ***Corresponding author address:*** **Caroline Leck, Stockholm University, Department of Meteorology, SE-106 91 Stockholm, Sweden. Email:** lina@misu.su.se

2. CLOUD FORMING PARTICLES OVER THE PACK ICE

An expedition in 1991 (Leck et al., 1996) revealed strong summer sources of DMS near the ice edge and adjacent waters and the dominant sulfate and methane sulfonate ions in the accumulation mode (diameters 100 to 1000 nm) aerosol (Leck and Persson, 1996a,b). However, as these particles become CCN, while traveling in over the pack ice, they become parts of clouds droplets that eventually deposits at the surface and are lost forever. The number of CCN with a source at the ice edge thus decreases with time of transport away from the ice edge. This has a profound impact on the properties of Arctic clouds, making them appear "grayer" than their mid-latitude counterparts.

Can climate change alter the Arctic system such that more biogenic particles are produced locally by generating larger areas of open water in the pack ice? Are there already other processes that produce biogenic aerosols in the pack ice? Will an enhanced production of CCN in the central Arctic Ocean act as a negative feedback, by producing brighter clouds reflecting more solar radiation back to space?

To help answering these questions, Arctic Ocean summer Experiments were launched the same area north of 80°N, in 1996 (Leck et al., 2001) and 2001 (Leck et al., 2004) on the Swedish icebreaker *Oden*. We found clear evidence that local aerosol production at the ocean surface occurred even when the fraction of ice was large (~ 95%). These novel conclusions were based on *in situ* measurements of atmospheric aerosols, boundary-layer structure, and of the film on the surface of the open leads, the "surface microlayer of the open leads" (SMOL).

A radio-controlled miniature boat was used to collect the <100 µm thick surface film of the open water between ice floes (Knulst et al., 2003), and the water from the collected film was examined. Aerosol particles were simultaneously collected from the atmosphere. Similarity in morphology, chemical and physical properties of the numerous aggregates and their building blocks, and of bacteria and other microorganisms was found in both the air and water. This strongly suggests that the airborne particles were ejected from the water by bursting bubbles (Bigg et al., 2004; Leck and Bigg, 2004).

On average during the five weeks spent in the pack ice region during 2001, SMOL-derived particles represented more than one-third of the collected airborne particles, more than two-thirds on sunny days. Instead of being liquid sulfuric acid, these particles were water insoluble, often having a crystalline appearance, either as aggregates or individuals (*Leck and Bigg,* 1999), Figure 1. This invalidates the hypothesis by Charlson that DMS oxidation products alone produces particles of this size.

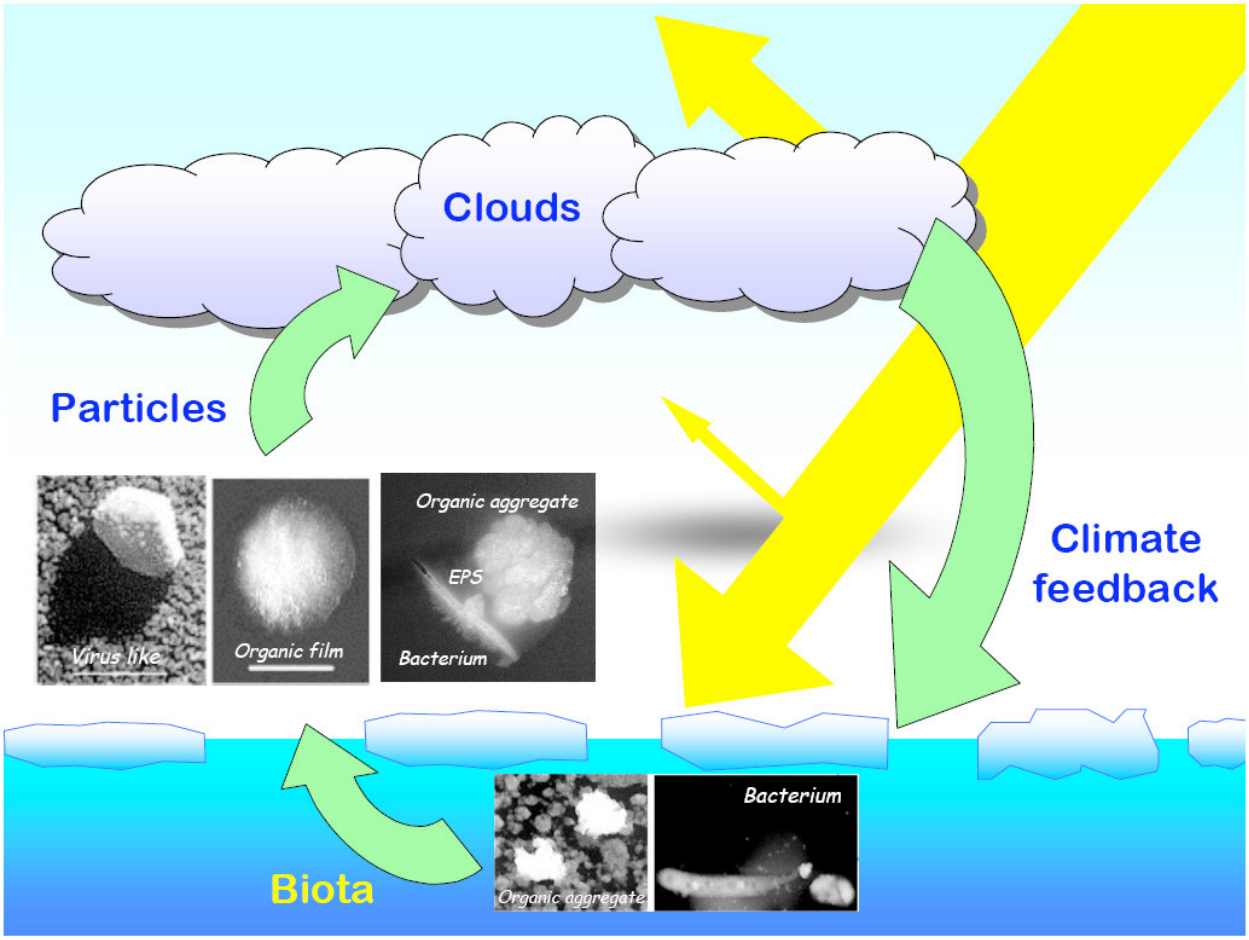

Figure 1. A schematic picture of the relationships between the processes described and how they are connected to cloud–aerosol interactions, along with examples of particles sampled in the boundary layer and in the SMOL.

One feature of SMOL particles was that they were joined together and surrounded by a diffuse electron-transparent material. Close examination of airborne particles revealed its presence on them as well. Examples are shown in Figure 1. The gel-like secretions of microalgae and bacteria known as “exopolymer secretions” (EPS) are well known to marine biologists, but previously not to aerosol scientists. EPS consists mainly of polysaccharides and has a number of properties (Decho, 1990). The molecules are highly surface-active, take up water like a sponge and release it very reluctantly. They capture heavy metal ions and readily bind other molecules, large and small, into their structures and spontaneously assemble into gels. The gels collapse under the influence of ultraviolet light and acidification (Chin et al. 1998). Their lifetime in the atmosphere is therefore limited and the collapse of the structure having such strong water retentive properties explains some of the puzzling features of the aerosols observed over the pack ice. For example the expulsion of water as the gel collapses may explain why airborne aggregates and bacteria very rarely have attached sea salt. The breakup of aggregates when the joining EPS gel collapses is also a suffIcient reason why the airborne aggregate size distribution so closely resembles that of the SMOL aggregates, but is shifted to a smaller size.

Comparison of the size distribution of airborne aggregates and particles with the size distribution of the total aerosol provided by a differential mobility sizing system strongly suggests that broken aggregates provide almost all the particles between 10 and 70 nm diameter, the Aitken mode.

3. IMPLICATIONS OF A LOCAL PACK ICE SOURCE OF CLOUD FORMING PARTICLES

Fresh aggregates with gel on them could act as CCN directly because of the gel’s strong surface active properties. Aqueous oxidation of sulfur dioxide could then produce sulfur-containing particles with aggregates inside. Those that have lost their gel could still act as sites for the condensation of the oxidation products of DMS, and so could lead to production of sulfur-containing aerosols. DMS concentration will determine the mass of sulfate produced but will have only a minor influence on the number of CCN, and thus cloud droplets, which will instead be dictated by the number of airborne particles originating in the SMOL.

Boundary layer clouds are frequent in the summer Arctic, are optically thin and have low concentrations of CCN, compared to boundary-layer clouds at lower latitudes. These are conditions that maximize the effects of changes in CCN and cloud droplets number concentration on short-wave radiation. On a regional scale there is therefore a potential for a biological impact on climate, but the emphasis has now shifted from phytoplankton beyond the ice edge to bacteria and microalgae, and their secretions, within the pack ice. While there can at present be no definite answer to the question in the title, it does look to be a tentative "yes", and the marine biosphere will affect the melting of the ice.

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