GÖTEBORGS UNIVERSITET

Thesis for the degree of Doctor of Philosophy in Natural Science,
Specialization in Chemistry

How ship exhaust particles change cloud activity
Implications for Arctic mixed-phase clouds

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The Arctic is experiencing surface warming rates that exceed those observed at lower latitudes. This is caused by a complex system of feedback mechanisms in the Arctic climate system and is referred to as Arctic Amplification. Persistent mixed-phase clouds cover large areas of the Arctic region and thus, have a substantial impact on the radiative budget in the Arctic. One consequence of the amplified warming is that Arctic sea-ice extent has been decreasing over the past decades. With the rapid decline in sea-ice extent, shipping activity is projected to significantly increase due to easier accessibility and availability of shorter transportation routes. Ships are also a significant source of atmospheric pollutants, which include greenhouse gases, sulfur oxides (SO_x) and particulate matter (PM). Increased local emissions of such pollutants may perturb the natural state of Arctic clouds and thus, can lead to further climatic feedbacks.

Simultaneous with climate change, the shipping sector is undergoing regulatory changes aimed at reducing exhaust emissions of climate- and health-affecting substances. SO_x and PM emissions have been strongly linked to tens of thousands of premature deaths worldwide. As a result, the International Maritime Organization (IMO) implemented regulations that aim to reduce emissions of SO_x and indirectly, of PM to atmosphere. Respective IMO regulations mandate ships to either use fuels with reduced fuel sulfur content (FSC) or to use exhaust aftertreatment systems, such as wet scrubbers, in instances where ships continue to utilize non-compliant high sulfur content fuels. Both, FSC reduction and exhaust wet scrubbing change physicochemical properties of exhaust particles and may therefore, lead to climate feedbacks.

A series of laboratory engine experiments were performed to characterize impacts of FSC reduction and exhaust wet scrubbing on the physicochemical properties of exhaust particles. The secondary impact of compliance choices on exhaust particles' cloud activity, including liquid droplet and ice crystal formation, was a particular focus. Results from laboratory experiments were subsequently implemented into a cloud-resolving large eddy simulation model, in an effort to quantify the impact of additional ship aerosol particles on micro- and macrophysical properties of an Arctic mixed-phase cloud.

Results presented in this study illustrate how international regulations in the shipping sector may affect atmospheric processes. We observed that FSC reduction and wet scrubbing have opposing effects on exhaust particles' ability to form liquid droplets in the atmosphere. These results are supported by observed changes in particles' chemical mixing states. Moreover, we find that the impact of wet scrubbing on exhaust particle properties varies substantially between experiments and between marine engines. Wet scrubbing may lead to particle emission profiles, which counteract the intended aims of the IMO regulation. This may have further consequences for health- and climate-related issues.

Our modelling study suggests that ship aerosol may lead to increased cloud droplet and decreased raindrop number concentrations. Consequently, a reduction in surface precipitation and reduced longwave radiative surface cooling were observed. Nevertheless, changes in cloud properties were mostly observed when ship particle number concentrations were significantly increased and/or strongly depended on the aerosol particle properties.

Increased shipping activity in the Arctic may lead to further climate feedbacks, but as demonstrated in this thesis, the magnitude of the induced changes strongly depends on fuels and engines, and whether exhaust aftertreatment systems are utilized. Given continuous changes in the shipping sector, it becomes challenging to predict how ship exhaust emissions in the Arctic may evolve over the next decades. New regulations are emerging, and the consequences should be studied as the Arctic is one of Earth's most pristine and sensitive regions, where increased shipping activity may lead to substantial environmental impacts.

Keywords: Aerosol particles, mixed-phase clouds, ship emissions, Arctic, maritime regulations