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Incorporation, transport, and release

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GLACIERS AS A TEMPORARY RESERVOIR OF PERSISTENT ORGANIC POLLUTANTS: INCORPORATION, TRANSPORT, AND RELEASE

A thesis submitted to attain the degree of
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presented by
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Summary

Persistent organic pollutants (POPs) are a group of poorly degradable, bioaccumulative, and toxic chemicals. After a period of widespread use in the 20th century, certain POPs were gradually phased out and banned at the end of the 20th century. Due to their persistence and semivolatile character, POPs are globally distributed and can reach remote areas where these pollutants have never been used and emitted.

In Alpine areas, POPs are transferred from the atmosphere to glaciers by dry deposition, or by deposition initiated by snow- or rainfall. As the snow ages, it transforms to firn, and subsequently to glacier ice. The pollutants can be incorporated into the glacier ice, revolatilize to the atmosphere, degrade, or run off with meltwater or particles. Impurities incorporated into the glacier ice are transported from the accumulation area (where new glacier ice is formed) to the ablation area (where the ice melts), a transport process which can last decades to centuries in Alpine glaciers. When the ice melts, the chemicals contained in the ice are released with meltwater and can reach the aquatic freshwater system. The fate of semivolatile organic pollutants in a glacier system has not been investigated in detail before.

This thesis studies the fate and dynamics of polychlorinated biphenyls (PCBs) in glaciers. The thesis aims at investigating the fate of the six indicator PCB congeners (iPCBs) in two types of glaciers. The PCB congeners 28, 52, 101, 138, 153, and 180 are used as representative model chemicals, as they cover a wide range of properties, and measured environmental concentrations of these congeners are available.

In the first study of this thesis, the fate of PCBs in the accumulation area of a cold glacier is investigated. In a cold glacier, the ice temperature is below 0°C, and melting is negligible in the accumulation area. Chemical fate is studied by developing a multimedia chemical fate model describing deposition, postdepositional processes, and incorporation of chemicals into the Alpine Fiescherhorn glacier (Switzerland) and the Arctic Lomonosovfonna glacier (Svalbard, Norway). The model is based on local snow accumulation data, and is validated by comparing modeled and measured PCB concentrations in ice cores. The ice core of the Fiescherhorn glacier was analyzed for PCBs in the doctoral study of Pavlina Pavlova, which was conducted parallel to this thesis. The modeled and measured PCB concentrations peak in the 1970s, the period of
highest PCB emissions. For the individual congeners, average modeled concentrations are between 50 and 400 pg L\(^{-1}\) in the ice core of the Fiescherhorn glacier, and between 8 and 50 pg L\(^{-1}\) in the Lomonosovfonna glacier. The model is able to reproduce and explain measured PCB levels in the ice cores. The model shows that the majority of the lower-chlorinated congeners is lost from the glacier by revolatilization (86% of PCB 28). In contrast, the higher-chlorinated congeners are stored in the glacier ice (98% of PCB 180).

In the second study, the incorporation model is applied to the accumulation area of a temperate glacier, where the ice temperature is around 0\(^\circ\)C, and melt processes are important. The multimedia chemical fate model is extended by including meltwater and particle percolation and runoff. The model is validated by comparing modeled and measured PCB levels in an ice core from Silvretta glacier (Switzerland), which was also analyzed for PCBs by Pavlina Pavlova in the parallel doctoral study. Average modeled concentrations are between 20 and 200 pg L\(^{-1}\) for the individual congeners, which is lower than the levels in the cold glacier. The lower concentrations are caused by higher losses attributed to revolatilization and particle runoff in the temperate glacier. Again, the PCB concentrations peak in the 1970s, but the concentrations also reach high levels in the 1950s and the 1990s, which were periods of strong melt. Strong melt induces an enrichment of particles and associated chemicals in layers located at the glacier surface during the melt period. As a consequence, the interpretation of temperate ice cores as environmental archives of past atmospheric conditions should include postdepositional processes.

In the third study, the incorporation model is applied to the entire glacier surface of the Silvretta glacier, and connected to a glacier flow model based on fluid dynamics, and a multimedia chemical fate model describing the glacial lake. The amount of chemicals incorporated into the glacier surface is passed on to the flow model, and transported along glacier flowlines. When ice traveling along a flowline reaches the glacier surface, the chemicals contained in the ice are released to the proglacial lake fed by glacier meltwater. In the lake, the chemicals can be deposited to the sediment, or leave the lake with water advection. The models are validated by comparing modeled and measured levels in the ice core, in a sediment core from the glacier lake, and in the stream water. Again, PCB levels in the sediment core and the stream water were determined by Pavlina Pavlova in the parallel doctoral study. The models are able to reproduce the measured concentrations of the higher-chlorinated congeners in the different matrices, with average modeled concentrations in the sediment between 150 and 170 pg g\(^{-1}\) dry weight for the individual congeners. In contrast, the concentrations of the lower-chlorinated congeners in the sediment core are underestimated by the model (average modeled sediment concentrations between 4 and 14 pg g\(^{-1}\)). This deviation might be caused by overestimated revolatilization and missing processes in the ablation
area. The PCB storage in the Silvretta glacier increases until the 1980s, reaching a maximum of 70 g iPCBs. Thereafter, PCB storage decreases, with a strong release of chemicals in the 1990s and the 2000s. After 2010, the models predict a constant but low release of 1 g iPCBs per year, and an iPCB concentration in the glacial stream of 100 pg L$^{-1}$. This concentration is below toxic levels.

This thesis studies the fate of PCBs in glaciers depending on the properties of the PCB congener and the glacier type. Higher-chlorinated congeners tend to be more incorporated into the glacier ice than lower-chlorinated congeners, which are mainly lost from the glacier surface by revolatilization. PCB incorporation increases with the net snow accumulation (glacier mass balance), and is higher in cold glaciers than in temperate ones. In 2015, the models estimate that 200 g in total PCBs are stored in the Silvretta glacier. A simplistic extrapolation of the PCB mass stored in the Silvretta glacier to the glaciers in the European Alps based on the surface area results in 200 kg of PCBs. This is a small amount compared to contemporary sources. However, it leads to a constant, but small release of toxic chemicals to remote areas throughout the 21st century.

Future studies may investigate meltwater, particle, and revolatilization processes, as well as chemical fate in the ablation area. Further, the models could be applied to current-use chemicals, and to different glaciers, providing knowledge about the total storage of chemicals in the cryosphere. This could then be used to quantify potential present and future exposure of sensitive remote ecosystems to various persistent, bioaccumulative, and toxic pollutants.
Zusammenfassung


In der ersten Studie dieser Doktorarbeit wird das Umweltverhalten von PCBs im Akkumulationsgebiet eines kalten Gletschers untersucht. In einem kalten Gletscher liegt die Eistemperatur unter 0°C, so dass Schmelze im Akkumulationsgebiet vernachlässigbar ist. Das Verhalten und die Dynamik der Chemikalien wird in dieser Studie mit einem Kompartiment-Modell untersucht, welches die Ablagerung, postdepositionale Prozesse


In der dritten Studie wird das Chemikalienmodell aus der zweiten Studie auf die gesamte Fläche des Silvrettagletschers angewendet und mit einem auf Fluidodynamik basierenden Gletscherfließmodell und einem Chemikalienmodell für den Gletschersee


Zukünftige Studien könnten die Schmelzwasser-, Partikel- und Diffusionsprozesse sowie das Umweltverhalten der Chemikalien im Ablationsgebiet weiter untersuchen. Ferner könnten die Modelle auf Chemikalien angewendet werden, welche heutzutage genutzt
werden. Auch die Anwendung der Modelle auf andere Gletscher wäre interessant. Dies könnte Erkenntnisse über die totale Menge an Chemikalien liefern, welche in der Kryosphäre gespeichert ist. Damit könnte man auch die mögliche aktuelle und zukünftige Exposition von sensitiven, abgelegenen Ökosystemen durch persistente, bioakkumulierende und toxische Schadstoffe abschätzen.