Ådne Garpestad Tønnessen

Climate change's effect on transportation of PCBs

Bachelorprosjekt KJ2900

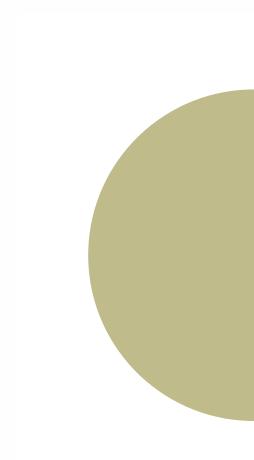
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NTNU

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Abstract

The change in climate affects the global temperature and weather systems, which are key factors in the transportation of PCBs. Using theories and studies to investigate the transportation and volatility of PCBs in the environment. The focus will primarily be on the atmospheric transportation but also bioaccumulation. This thesis will look into how the climate change can affect these processes.

Summary

Studies indicate that climate change and higher temperature stimulates the transportation, partitioning and volatility of polychlorinated biphenyls (PCBs). PCBs are a persistent organic pollutant, which is toxic for the environment. It gets released into the environment, through old electrical equipment, paint and other anthropogenic sources, and will then be transported

via the air, ocean or bioaccumulation through living organisms. Long-range atmospheric transportation (LRAT) is the main source of transport and the fastest, and is stimulated by the increase in temperature due to the climate change. The LRAT is also affected by the weather systems, and higher wind speeds. The increase in ocean temperature and PCB exposure, affects the food web and uptake of PCBs in bioaccumulation. Increase in temperature also increases the atmospheric water vapor, which produces more OH radicals. The OH radicals react with the PCBs, and are the main source to its degradation in the atmosphere. A possible increase in the total amount of PCBs in circulation in the environment, will also lead to more reactions with ozone, which could potentially damage the ozone layer.

Introduction

In this paper we will focus on how PCBs (polychlorinated biphenyls) get transported and how the climate changes can have an effect on this transport. When discussing the transportation of PCBs, it is important to have an understanding about what PCBs are, where PCBs come from, and why PCBs are an environmental problem. EPA (United States Environmental Protection Agency), is a federal government agency made to protect humans and the environment by regulating chemicals and pollutants. One of the pollutants they have regulated are the anthropogenic PCBs, which were domestically manufactured from 1929 until its ban in 1979. They have a variety in toxicity and toxic effects, and was used in many industrial and commercial application due to their chemical stability, high boiling point, non-flammability and their electrical insulating ability. Even though production is banned in many countries, there are still many products before the ban that are still in use, which can contain PCBs. A few examples of products are as follows:

- Cable insulation
- Transformers and capacitors
- Electrical equipment
- Oil-based paint
- Thermal insulating materials

PCBs have been released into the environment from the early 20th century up to its ban in 1979, and is still being released from countries not following the ban, and also from

equipment containing PCBs made before the ban. Improper disposal of older equipment is a common source to the exposure of PCBs. The abilities that have made PCBs useful in for example paint, is that it doesn't break down easily. The health effects that PCBs can cause are cancer, immune effects, reproductive effects, neurological effects, endocrine effects and more (EPA, 2022). This is one of the major problems with PCBs, and one of the reasons it is categorized as a POP (Persistent Organic Pollutant) by the Stockholm Convention (EPA, 2009).

One recent case of PCB pollution, that has caught the media's attention, is the case about Monsanto and their contamination of the city of Anniston. According to The Washington Post and CNN from articles back in 2002, Monsanto (a former producer of PCB mixture aroclor) engaged in "outrageous" behavior, when they released tons of PCBs into the city of Anniston Alabama, and then covered it up for decades. At the time yards and creeks had the highest levels of PCBs ever recorded in a residential community. Documents showed that Monsanto knew about the pollution for decades and the managers discovered that fish dunked in a local creek turned belly up within 10 seconds, spurting blood and shedding skin. Still, they didn't act on it, and has been left with paying millions of dollars to the offended parties (Grunwald, 2002) (CNN, 2002). From a New York Times article a year later, the Monsanto Company agreed to pay 700 million dollars to settle the case with more than 20000 residents in the city of Anniston (TheNewYorkTimes, 2003). The huge amount of money paid to settle the case, gives a good indication to what kind of problem PCBs can cause, and why it is important to learn more about it, to help in reducing its environmental problem.

In this paper we will not take a deeper look into lethal dose, sources of PCBs or toxic effects. The focus will be primarily towards atmospheric transportation of PCB and bioaccumulation, and the climate change's possible effect on these transportations.

Theory

PCB

The structure and nomenclature of a general polychlorinated biphenyl or PCB is shown in the figure below. As the name suggests, a PCB consists of two benzene rings (phenyls) connected by a covalent carbon bond. A number of chlorine atoms can be substituted to the 5 free carbon atoms on each ring on either the *ortho*, *meta* and or, *para* carbons. This makes it possible to

have many different congeners of PCB and it is theoretically possible to have a total of 209 different congeners. The rings are free to rotate about the carbon bond, with exception to orthochlorine substitution on 2,2' or 6,6', which will make the PCB non planar. The toxicity of the PCB is affected by the position of the chlorine substitutions, the possible rotation about the bridging carbon bond, and the reactivity of the chlorine atoms. Meta and para substituted PCBs can resemble dioxin, when in a coplanar confirmation. 2,2' or 6,6' ortho substituted PCBs which cannot be coplanar due to steric hindrance from the chlorine atoms, can resemble estrogen. (G. Landis, et al., 2018, pp. 182-186) (Kraft, et al., 2016)

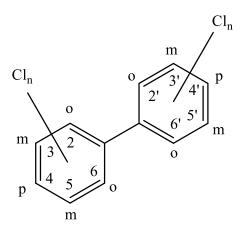


Figure 1: Structure and nomenclature for PCBs. The PCB compound exist with the same general structure, with a different amount of chlorine atoms, located on the different carbon atoms of the two aromatic rings. The number on each carbon denotes the position of the substituted chlorine atoms. The relative positions are denoted by, o, m and p, for ortho, meta and para. (G. Landis, et al., 2018, pp. 182-186)

Transportation

Most POPs (persistent organic pollutants) are volatile enough to evaporate and deposit from air, water and soil at ordinary temperature. The warm temperature in tropical and subtropical regions favors evaporation of POPs. Cooler temperature at higher latitude and in polar regions favor deposition from the atmosphere to water and soil. This makes it so that POPs accumulate in cooler regions, such as the Arctic and Antarctic. The cooler temperature favor adsorption of POPs to atmospheric particulate matter, which deposits on the surface. Cooler climate also slows down the natural decomposition reactions, making POPs even more persistent than they already are. The cooler temperature also slows down evaporation of POPs

from for example water and favors the condensation or "partitioning" of POPs from the atmosphere to the water. The POPs don't condense in the sense that the atmosphere get saturated or supersaturated, since the conditions are always below the saturation limits. The POPs only partition from a gaseous phase to a non-gaseous phase (Wania & Mackay, 1996) (Kawai, et al., 2014).

Global distillation: Grasshopper effect and global fractionation

Long-range atmospheric transport (LRAT) is when POPs migrate on a global scale. To understand why POPs migrate, we must take a look at the mechanics behind LRAT. PCBs are a class of semi-volatile organic compounds (SVOCs). SVOCs are organic chemicals like the PCBs that have a low or moderate vapor pressure, which can make them volatilize under some of the Earth's environmental conditions (Qiao, et al., 2021) (G. Landis, et al., 2018). Volatilization is temperature dependent, and a higher temperature will increase the volatility of chemicals. This means that the PCBs or others POPs that gets emitted/released in warm tropic environments, will be more volatile than in other regions of the Earth. The volatilization of organic chemicals is also affected by the weight of the chemical, which means the lighter congeners of PCBs are more volatile than the heavier ones containing more chlorine. Since higher temperature will make POPs more volatile, the contaminating POPs in tropic environments are more likely to evaporate than in cooler conditions. Most POPs are volatile enough to evaporate and deposit from air, water and soil at ordinary temperature. The warm temperature in tropical and subtropical regions favors evaporation of POPs. Cooler temperature at higher latitude and in polar regions favor deposition from the atmosphere to water and soil. This makes it so that POPs accumulate in cooler regions, such as the high latitude regions of Arctic and Antarctic. The cooler temperature favor adsorption of POPs to atmospheric particulate matter, which deposits on the surface. Cooler climate also slows down the natural decomposition reactions, making POPs even more persistent than they already are. The cooler temperature also slows down evaporation of POPs from for example water, and favors the condensation or "partitioning" of POPs, from the atmosphere to the water. The POPs don't condense in the sense that the atmosphere get saturated or supersaturated, since the conditions are always below the saturation limits. The POPs only partition from a gaseous phase to a non-gaseous phase. The process of volatilization and condensation of POPs, which results in LRAT is called "Global distillation". Global distillation is one of the reasons why

PCB contamination is found in the Arctic and Antarctic, far away from their sources (Wania & Mackay, 1996) (G. Landis, et al., 2018).

The process of global distillation can be separated into two different phenomena, called the grasshopper effect and global fractionation. The grasshopper effect is when chemicals travel to higher latitudes in a series of short jumps. The chemical's properties and the environment (temperature, wind speed and wind direction) affect if the chemical volatilize and condense once or more. The chemical migrates and rests in succession with the change of the seasonal temperature. This makes it so the chemical migrates in the warmer seasons, and accumulates in cooler seasons. The less volatile chemicals will have less of these jumps, but the more volatile chemicals may perform these jumps to they reach high latitude areas (Wania & Mackay, 1996) (Gouin, et al., 2003) (G. Landis, et al., 2018).

Global fractionation is when chemicals condense at different temperatures, due to their different volatility. This makes it so more volatile chemicals travel further than the less volatile chemicals. More chlorinated PCBs will for example travel a shorter distance than less chlorinated PCB congeners, since the chlorine atoms are heavier than both the carbon and hydrogen atoms by a large margin, and the different congeners of PCB may therefore have a different volatility. This will result in a disperse redistribution of PCB congeners. The "hops" the chemicals goes through in LRAT will differ in distance and height, depending on the chemical properties and the ambient temperature (Wania & Mackay, 1996) (G. Landis, et al., 2018) (Meijer, et al., 2002) (Gouin, et al., 2003).

Bioaccumulation: Bioconcentration and Biomagnification

Bioaccumulation is the total accumulation of a chemical in and on an organism from all of the sources in the specific environment. It usually results in a greater concentration of the chemical in the organism than with the respiratory environmental medium or the consumed food. Bioconcentration and biomagnification are parts of bioaccumulation (G. Landis, et al., 2018). (Masset, et al., 2019)

Bioconcentration is the accumulation of a chemical in and on an organism, where the source of the chemical is solely from water. Biomagnification is the accumulation of a chemical in an organism, where the source of the chemical comes via the food. In biomagnification there is usually an increase in concentration of the chemical in the organisms up the trophic levels.

PCBs for example, which are lipophilic and chlorinated are poorly metabolized and will therefore biomagnify easily (G. Landis, et al., 2018). (Masset, et al., 2019)

Partition coefficients

The partition coefficients relate to the affinity of a compound in a mixture of two different substances. For K_{OW} those two substances are octanol, which means organic, and water. For K_{OA} those two substances are organic and air. The octanol-air partition coefficient is the ratio of a chemical's concentration in air at equilibrium, and the octanol-water partition coefficient is the ratio of a chemical's concentration in water at equilibrium. The partition coefficients are used to predict the partitioning behavior between air and water, and environmental matrices like, organisms, vegetation, soil and aerosol particles. The formula for the correlation between the octanol-air and the octanol-water partition coefficient is shown below. K_{OA} is the octanolair partitioning coefficient, K_{OW} is the octanol-water partitioning coefficient, R is the ideal gas constant, R is the absolute temperature and R is the henry constant.

$$K_{OA} = K_{OW}(RT)/H$$

(Meylan & Howard, 2005) (Li, et al., 2019)

Atmospheric reactions

There is a cycle of reactions happening in the atmosphere at all time, shown in figure 2 below.

(Prinn, 1994)

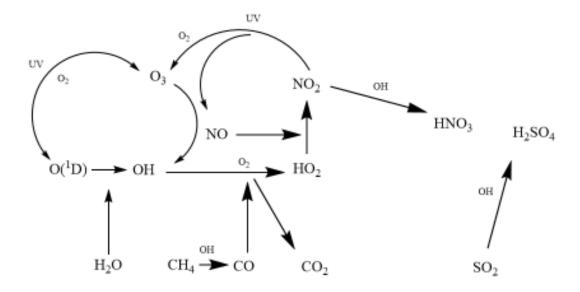


Figure 2: A simplified schematic of some important reactions involved in oxidation in the stratosphere. (Prinn, 1994)

Discussion

Weather systems and LRAT

The climate change has already started with visible indicators as the rise in global temperature, melting of polar ice, increasing ocean levels and more radical weather patterns (EPA, 2021). The atmospheric weather systems and its patterns are directly affected by the climate change. LRAT of POPs is directly affected by the different weather systems around the globe, and the changes in weather patterns from climate change will have a major effect in the transport of POPs such as PCBs. The climate change and climate variability can change the exposure to chemicals, by alteration of pathways or through alteration of environmental concentration mechanics. (Macdonald, et al., 2003) (Beyer, et al., 2000)

Once a contaminant is released in the environment, it undergoes a transportation by air or water. Both water and air are important contributions to the transportation process of contaminants, but out of the two air is a much faster process. The air can move contaminants from where they were released to remote locations within days or weeks with a velocity of meters per second, while the ocean takes years with a velocity of cm per second. The atmospheric transport is often not direct and may involve a number of transfer and degradation processes. In this way POPs can be removed from the atmospheric transportation

via exchange with surfaces (soil, vegetation, water), by partitioning into rain or onto snow/hail, or partitioning onto particulate matter, which can be removed by precipitation. The removal of the POPs from the atmosphere can be temporary, by chemical revolatilizing in the seasonal or daily thermal cycles, or different processes that enhances the chemical vapor pressure of the POPs. The semi-volatile chemicals travel through the atmosphere trough several hops, before they settle at their final destination. Since the partitioning depends on the chemical properties, described with the partitioning coefficients, the exchange during the transport results in what can resemble a chromatographic process which favor a quicker transport of the chemicals that exchange poorly and are more volatile. (Macdonald, et al., 2003)

If one looks at the Earth as a chromatograph, one can picture the air as the mobile phase and the soil as the stationary phase. The chemicals that favor the stationary phase, will be transported more slowly, and the chemicals that favor the mobile phase will travel further and faster. Within chromatography the elution of different compounds of different concentration will vary with time. For the global chromatograph, this time can be anything between weeks to decades. POPs released to the environment within the last decades have been undergoing redistribution in the global environment, in soils, permafrost, water, vegetation etc. Since the relatively "light" composition of chemicals is reaching remote locations, one can predict that heavier compounds over time will reach more and more remote locations, provided that the compounds of chemicals are persistent enough. This point is important since it shows that the global chromatograph isn't perfect, with internal processes that may degrade, transform, delay or permanently sequester compounds during their travels, and these processes are affected by the climate. Climate change scenarios suggest that the average global temperature can rise with several degrees (IPCC) during the next century, which means that one can predict an enhancement in the rate of cycling of semi-volatile chemicals, only through revolitalization. The global chromatograph can then run on a progressively higher temperature, but probably with a slightly decreased thermal contrast, since the polar region are suspected to warm faster. (Macdonald, et al., 2003) (Beyer, et al., 2000) (Ma & Cao, 2010) (Lamon, et al., 2009)

Rainfall patterns are highly sensitive to global change, but it is hard to predict how they will change with global warming. What we can project however, is that the hydrological cycle (continues circulation of water in the Earth-atmosphere system) will become more vigorous in the future. It can therefore become, a more important component of contaminant pathways than it is now. (Macdonald, et al., 2003)

Partitioning coefficients

When determining chemical fate in terrestrial environments, partitioning between gas phase and solid lipophilic compartments is one of the most important phenomena. The octanol/air partitioning coefficients (K_{OA}) have been used with great success to describe SVOC between the gas phase and atmospheric particulate matter, soil, vegetation and other terrestrial surfaces. As the octanol/air partitioning coefficient is defined as $K_{OA}=C_O/C_A$, where C_O (mol/L) is the concentration of the compound in the octanol, and C_A (mol/L) is the concentration of the compound in gas phase in the air at equilibrium. The equilibrium partitioning of a chemical between two compartments can be expressed as:

$$K_{OA} = exp\left[\frac{\Delta G_{OA}}{RT}\right]$$

where K_{OA} , is the octanol/air partition coefficient, ΔG_{OA} (J/mol) is the Gibbs free energy of the transfer from octanol to air, R is the ideal gas constant (J/mol/K), and T is the absolute temperature (K). The Gibbs free energy used in the equation above is given by this equation:

$$\Delta G_{OA} = \Delta H_{OA} - T \Delta S_{OA}$$

Where ΔS_{OA} is the entropy of phase change (J/mol/K). These equations shows that K_{OA} can be a strong function of temperature. Table 1 shows the K_{OA} of the different PCB congeners, and there is a clear correlation between the K_{OA} and number of chlorine atoms. The less chlorinated PCBs have a lower K_{OA} and will therefore have a stronger affinity to the gaseous phase than more chlorinated PCBs.

(Komp & McLachlan, 1997) (Hongxia, et al., 2008) (Li, et al., 2019)

Table 1: Measurement of KoA for different PCB congeners (Komp & McLachlan, 1997)

PCB substitution pattern	PCB IUPAC no.	logK _{OA} (25°C)
(2,2'); (2,6)	4 + 10	7.18
(2,4'); (2,3)	8 + 5	7.40
(2,2',5)	18	7.60
(2,2',3); (2,4',6)	16 + 32	7.72
(2,4',5); (2,4',4)	31 + 28	7.92
(2,2',5,5')	52	8.22
(2,2',3,5')	44	8.36

(2,3,4',6)	64	8.41
(2,2',3,5',6)	95	8.71
(2,2',3,3',6); (2,2',4,5,5')	84 + 101	8.80
(2,3,3',4',6)	110	9.06
(2,2',3,4',5',6)	149	9.27
(2,2',4,4',5,5')	153	9.37
(2,2',3,4,4',5'); (2,3,3',4,4',6)	138 + 158	9.51
(2,2',3,4',5,5',6)	187	9.87
(2,2',3,4,4',5,5')	180	9.88

Atmospheric reactions

As mentioned earlier, PCBs are primarily transported via the atmosphere. The PCBs can undergo several chemical reactions through the atmospheric travels, and particularly with the OH radical, while the PCB is in the vapor phase. From the article (OH Radical Reactions: The Major Removal Pathway for Polychlorinated Biphenyls from the Atmosphere) (Anderson & Hites, 1996), the rate constant of the gas-phase reaction of 14 congeners of PCB with OH radicals were tested to calculate the atmospheric lifetime of the OH initiated PCB reactions. These lifetimes varied from 2 days for biphenyl, to 34 days for a pentachlorobiphenyl. At the time the article was written, the calculated global loss rate of atmospheric PCB due to removal by OH radicals were (8300 t/year). (Anderson & Hites, 1996) (Totten, et al., 2001)

Atmospheric reactions of PCBs with other oxidant like O₃ (Ozone), NO₃ and Cl-atom are slow in comparison to other aromatic compounds. This makes the reaction with the OH radical the main source of PCB degradation in the atmosphere, even though oxidation of PCBs by for example ozone, still happens. Reactions of PCBs with the OH radicals, forms a large fraction of HO-PCB compounds (Awad, et al., 2016) (Dudasova, et al., 2016). Due to the OH group, HO-PCBs are less volatile and more soluble in the aqueous phase than their parent PCBs. HO-PCBs might therefore partition in a liquid phase more likely in the atmosphere or enter an aqueous environment such as the ocean. HO-PCBs in an aqueous environment might degrade via different mechanics and might have another ecological effect than regular PCBs. Several PCB-OH radicals react with O₂ at a low effective bimolecular rate, which allows their reaction with NO₂. PCB-OH reacting with NO₂ forms Nitro-PCBs, which are likely stronger carcinogens than regular PCBs (Liao, et al., 2019) (Kanazawa, et al., 2011).

From the book (Global Atmospheric-Biospheric Chemistry) it is said that most of the internal chemical processes occurs where ozone, nitrogen oxide and hydroxyl free radical are the key reactive species. Ozone is an important chemical in the atmosphere, by working as a protective shield against ultraviolet radiation. Hydroxyl free radical (OH) is responsible for most of the gas phase oxidization in the troposphere on a global scale, and its concentration is therefore important for the troposphere's capacity to oxidize trace gases emitted into the atmosphere. The chemistry involved with production and removal of OH is spectacular, since the compounds it involves are playing a central role in determining the radiative forcing of climate, atmospheric ozone concentrations, air pollution levels and acid rain (HNO₃, H₂SO₄) fluxes. Figure 1 in the theory is a simplified example of some of the reaction happening in the atmosphere. (Prinn, 1994) (Nelson, et al., 2021).

If the climate change raises the temperature in the tropical oceans, one can expect an increased amount of water vapor in the tropical lower troposphere. Water vapor is part of the primary source of OH, which means that an increase in water vapor will lead to an increase in OH. A rise in temperature also increases the rate of reaction of OH with organic chlorinated compounds, which will decrease the lifetime of those gases. A decrease in stratospheric ozone, can also increase tropospheric OH, since the singlet oxygen atom that reacts with the water vapor to create OH, is produced by radiation with wavelengths less than 310 nanometer. A decrease in stratospheric ozone will lead to an increase 310 nanometer fluxes, and therefore an increase in OH production. (Prinn, 1994) (Dumka, et al., 2021)

Bioaccumulation

The climate change affects the transport of PCBs, and a rise in temperature will give an increase in the amount of PCBs in circulation. This means that more PCB will enter the oceans, and the marine life will as a result be exposed to more PCB. The effect increasing temperature has on bioaccumulation from climate change is hard to predict. From the article (Gouin, et al., 2012), several tests were performed to check the effects on increased temperature on fish, and how it affected the bioaccumulation. The findings from the tests showed that major impact from climate is more likely to come from indirect effects, like food web changes, as opposed to the direct bioenergetic impact temperatures have on the uptake through the consumption rate (Gouin, et al., 2012). From the article (Alava, et al., 2018), polar bears and sea birds from a warming Artic environment showed increased concentration of some POPs. This was due to a change in their diet composition and food web structure, and an

alteration in the exposure of pollutants and their pathways, driven by the climate change. (Alava, et al., 2018)

Conclusion

It is difficult to be precise about how the climate change will affect the transportation of PCB, but there are indications that can give us a clue. The partition coefficients are directly affected by the temperature, and an increase in temperature will make the PCBs more volatile. The increase in volatility increases the evaporation of PCBs to the atmosphere where they can undergo LRAT (long-range atmospheric transport). An increase in volatility will also make it possible for more chlorinated PCBs to evaporate, and enter the atmosphere. This means that there could be an increase of total PCBs in circulation in the environment. The PCBs will collectively travel further towards higher latitudes, and this process could also increase in velocity, due to more radical weather and wind patterns. Increased volatility and wind speeds will make the "hops" PCB goes through further, and more regular, since the seasonal and daily thermal cycles are warmer. PCBs that have been persistent in soil, snow, permafrost etc., could become active and volatile enough to evaporate and travel further in the environment via the air, oceans or via bioaccumulation in animals. The bioaccumulation of PCBs will also increase with the climate change, by more exposure and food web changes.

Since the PCBs are affected by the OH radicals and ozone in the atmosphere, an increase of PCBs in circulation will have an effect on the atmospheric chemical reactions and might push the equilibrium of the reactions. Increased water vapor from higher temperature, will increase the amount of OH radicals in the atmosphere, which will result in more degradation of PCB. More PCBs in the atmosphere, will also lead to more reactions with ozone, which could potentially damage the ozone layer.

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