

ABSTRACT

During the Swedish environmental monitoring of contaminants in off-shore sediments, so far carried out in 2003, 2008 and 2014, the levels of hexachlorobenzene (HCB) have displayed an increasing trend. The emissions of HCB have decreased since the 1970/80s and the production is believed to have ceased in 2004. It is therefore unclear why levels are now increasing. To obtain a better picture of the situation, the Geological Survey of Sweden (SGU), commissioned by the Swedish Environmental Protection Agency, has compiled data on the levels of HCB in samples in SGU databases, encompassing Swedish sediment samples. The aim is to investigate where there are high sediment levels of HCB, how the levels vary along the coast and in the sea, and how the levels have changed over time.

HCB is a persistent organic pollutant that has been intentionally produced, primarily for use as a fungicide in agriculture. It is also unintentionally formed in various combustion and chlorination processes. During the 1970s and 1980s, many countries banned the use of HCB as fungicide, resulting in significantly decreasing emissions and declining levels in the environment. As the intentional production has ceased, the unintentional formation is now responsible for the primary emissions. In Sweden, the largest part of HCB emissions (in 2016) derive from electricity and district heating production, mining, manufacturing and construction, and the waste sector, but air emissions have fallen sharply since 1990. In addition to primary sources, HCB can also be mobilized from secondary sources, i.e. sites polluted from previous emissions and from where HCB can now disperse. Secondary sources are, for example, contaminated land, contaminated sediments and landfills.

HCB data in this study comes from two SGU databases of sediment pollution, one containing data from samples taken by SGU in different projects and the other containing data reported to SGU as a data host. Sediment has been sampled between 1986 and 2015 and in total the data comprises 809 samples. At the first stage of the analysis, to investigate where there are high HCB sediment levels, all samples encompassing the sediment surface were included regardless of how deep the samples stretched. The deepest sample integrated the depth 0–50 cm, although most samples covered the 0–1 cm depth. In the second phase, to investigate large-scale geographical and time trends, only surface samples were included, i.e. 0–1 or 0–2 cm deep in the sediment. This represents the particles that sedimented the years just before the sampling. To evaluate trends over time, only data from off-shore samples were used in order to minimize the impact of point sources, as some data was derived from projects focused on contaminated sediments near land.

The results showed that the highest levels were found at the coast or in lakes and streams. Only one off-shore sample had a HCB content that was classified as very high, i.e. in class 5 according to the Swedish Environmental Protection Agency's assessment system. The highest levels were found in Iggesund (up to 450 µg/kg dry weight (DW)), Örnköldsvik/Domsjö (140 µg/kg DW), the Indal river downstream of Hissmofors (26 µg/kg DW), Stockholm (24 µg/kg DW) and at Timrå/Östrand (21 µg/kg DW). Many of these sites are connected to forest-related industries, where for example chlorine bleaching of pulp has taken place but also chlor-alkali processes involving graphite electrodes, which has caused HCB contamination. It should be noted that this study only covers data from SGU databases, including data from projects focused on sediments affected by forest industry-related activities. There are therefore probably other areas with high levels of HCB which have not been included in the study. In many cases, the high HCB levels are linked to high levels of dioxins, but in Hissmofors, an area with a former pulp mill and a sawmill, dioxin levels are surprisingly low compared to the HCB levels. The reason for the HCB pollution here is so far unclear.

In the off-shore areas, the HCB content also varies geographically, with generally lower concentrations on the Swedish west coast (the Skagerrak and Kattegat seas) than in the Baltic Sea. However, the levels on the west coast are elevated at the far north and far south. The lowest levels in the Baltic Sea generally occur in the south-eastern parts of the Baltic Proper while the levels are elevated outside Stockholm as well as in the western parts of the Arkona basin south of Sweden. In the Bothnian Sea and Bay, some samples with elevated levels occur in the region of Kvarken, but if these early samples (all taken 1990) are omitted, the levels are elevated in the southern parts of the Bothnian Sea compared to the northern parts of the Bothnian Sea or the Bothnian Bay, especially if the levels are normalized to the amount of organic carbon in the sediment.

No north–south trend in HCB concentrations could be observed on the west coast or in the Baltic Sea, neither normalized to dry weight or to organic carbon in the sediment. This shows that global long-range air transport does not determine the levels of HCB; instead there seem to be an impact from point sources. HCB levels in off-shore samples were significantly correlated with the levels of pentachlorobenzene, which is expected because HCB is degraded to this substance and they have similar sources. The number of samples for which both HCB and dioxin levels were determined was low and it was therefore not possible to see any correlations between these substances.

Overall, HCB levels seem to have increased in the past decade. This is the case in the Bothnian Bay, north and south Baltic Proper and the south west coast (Kattegat). In the Bothnian Sea, it is more difficult to see a clear trend, either on a dry weight basis or when the levels are normalized to organic carbon. In the north west coast (Skagerrak) there are a few samples with deviating high levels, but apart from this, the trend seems to be increasing. The increasing trend in sediment over time is supported by analyses in deeper sediment layers made in a previous study of a few of the national environmental monitoring stations in the Baltic Sea, where levels of HCB were found to begin to increase after about 2000. Similar time trends have also been observed in biota from the Baltic Sea and the Swedish west coast, with increasing levels of HCB in herring, cod and guillemot eggs from around 2005 at most monitored stations. In addition to these marine matrices, increasing time trends for HCB have also been observed recently in, for example, air in the Arctic and in Europe, and in reindeer and cattle in Sweden.

The reason for the increasing levels has not been established. The possible reasons are increased emissions from primary sources or increased remobilization from secondary sources. Emission inventories point to decreasing rather than increasing emissions from primary sources. There are probably emission sources that are not included in the inventory – in a modelling of HCB levels in air in Europe, the modelled levels, based on reported emission data, are much lower than the measured levels, suggesting that there are HCB sources missing in the inventory. However, it is unclear why HCB emissions from these not inventoried sources would increase. This illustrates that there is a lack of knowledge on HCB primary sources, such as the content of HCB contamination in pesticides containing chlorine, and hence the amount of HCB dispersed via pesticides globally.

The second possibility is increased emissions from secondary sources, that is, increased remobilization from more polluted areas due to, for example, increased evaporation of the substance or increased melting of ice containing the substance due to climate change. However, this should affect more substances than HCB, but similar increasing trends in levels have not been observed for other substances in the monitoring of off-shore sediment. It is possible that HCB due to its physicochemical properties (e.g. relatively high volatility among the persistent organic pollutants) may be affected more quickly by climate changes, but a change should still be observed for other substances. It is thus important to continue to monitor possible increases over time also for other pollutants than HCB.