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Natural formation of trihalomethanes (THM) in soils

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The occurrence of organohalogens in the environment was initially attributed to anthropogenic processes; a natural formation seemed to be limited on a few number of compounds. To date, more than 3800 halocarbons are known and identified to be produced through natural reaction pathways.

Trihalomethanes (THM), with chloroform being the most common, belong to these compounds and play an important role in photochemical processes of the lower atmosphere, but the current knowledge of the known sources and sinks of trichloromethane is still incomplete. The trichloromethane flux through the environment is estimated at ∼660 kt year-1, and 90% of the emissions are of natural origin. Next to offshore seawater contributing ∼360 kt year-1 unknown soil processes are the most prominent source (∼20 kt year-1).This paper describes a new abiotic source of trichloromethane from the terrestrial environment induced through the oxidation of organic matter by iron(III) and hydrogen peroxide in the presence of chloride. Different organic-rich soils and a series of organic substances regarded as monomeric constituents of humus were investigated for their release of trichloromethane. The influence of iron (III), hydrogen peroxide, halide, and pH on its formation was assayed. The optimal reaction turnover for the representative compound catechol was 58.4 ng of $CHCl_3$ from 1.8 mg of carbon applying chloride and 1.55 µg of $CHBr_3$ from 1.8 mg of carbon applying bromide; resorcin and hydroquinone displayed similar numbers. Results presented in this paper pinpoint 1,2,4,5-tetrahydroxybenzene as playing a key role as intermediate in the formation pathway of the trihalomethanes. The highest THM yields were obtained when applying the oxidized form of 1,2,4,5-tetrahydroxybenzene as THM precursor. These findings are consistent with the well-known degradation pathway starting from resorcin-like dihydroxylated compounds proceeding via further hydroxylation and after halogenation finally ending up in trihalomethanes. Additionally, the THM formation was investigated by using different types of iron species - iron (II) as well as iron (III). At this, high THM yields were only obtained in the presence of a 2-Line-Ferrihydrit, a 6-Line-Ferrihydrit or a with iron (III) saturated smectite. Both, the higher oxidation state of iron as well as low crystallinity seem to favour the reaction. In conclusion, Fenton-like reaction conditions (iron (III) and hydrogen peroxide), elevated halide content, and an extended reaction time can be seen as the most important parameters required for an optimal THM formation.