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### ACCUMULATION OF ADSORBED ORGANIC CHLORINE FROM ACTIVE SLUDGE USED AS FERTILIZER IN PLANTS

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Pressing problem. The pulp and paper mills are a source of polychlorinated condensed substances resulted from bleaching with elementary chlorine, hypochlorite and chlorine dioxide. The bleaching effluents come in biological treatment plant (BTP), where chlorine is adsorbed with active sludge (AS).



Surplus AS in some mills is transported to storing. AS supply is vast (about 405 million tons), enriched with nutrients (C, N, P, K) and is a potential source of organic fertilizers.

The widespread utilization of AS in agriculture, forestry and landscape gardening is restrained by its contamination with heavy metals and chloroorganic substances. In the early 1990-s the studies for evaluating surplus AS impact on physical-chemical properties and fertility of Arkhangelsk regional land were performed. However, recognizing the importance of the above researches it should be noted that the single results of the field and model tests in phytotron conditions can not explain the mechanisms of xenobiotic accumulation in plants after a long period of its introducing in the soil. These tests also ignore the process of self-purification taken place in ecosystems.

Targets. Our work was aimed at investigating biodegradation processes of organically fixed chlorine from OAO Kotlass pulp & paper mill's surplus AS. AS samples of different age and bioaccumulation of organically fixed chlorine in the four plant species, growing at the soils with high dosage of active sludge introducing as well as the post-effects of long AS application were studied.

Methods. AS was produced by filtering wastewater through polycarbonate filter; a part of adsorbed non-organically bound chlorine was determined by 5-fold washing the residue with acid-nitrate solution before its burning. AS of 3-year age, stored in piles on the OAO Kotlass pulp & paper mill's ancillary agricultural lands, was used in agrocoenosis. AS was introduced in period from 1983 till 1995 at 150-200 t/ha dosage by the tractors T-150K at 0-25 cm beneath the surface of topsoil. The total dose was 300, 600, 1000 and 1500 t/ha. 2000 t/ha single dose was introduced by the C-130 tractors in autumn 2005 on the stubble-fields. The soils of the fields were sandy and sandy loam.

Four species of 1-4, 9-12 and 17-year age were used to examine chlorine bioaccumulation. The plants had various roots and responses for high dose of AS introduction. The plants were sampled in 2006: the whole aerial part (shoots), leaves, buds, stems and perennial part of roots. The samples were disintegrated up to 5-7 cm, averaged, dried at room temperature and stored in polyethylene bags for constant moisture content.

For measuring total chlorine content the samples were beaten and burnt. Organically bound chlorine was determined through the following procedures: first, the samples disintegrated were placed in acid-nitrate solution, where active carbon and water were added. The mixture was shaken in 1 hour and filtered with the membrane filter to remove a part of non-organically bound chlorine.



Burning was done out in oxygen flow at 950°C. At burning organically bound chlorine and total chlorine transformed to hydrogen chloride and chlorides produced were measured by microcoulometric method using AOX analyzer MT-20 (Haberkorn + Braun Messtechnik Software Umweltanalytik, Germany).

Results. AS from BTP consists of zoogleal saprophitic bacteria community utilizing the pollutants from wastewater. It was found that 91,3 % of chlorine absorbed by the zooglea were organically bound and the residual 8,7 % had non-organically fixed form. Organically bound chlorine content in AS was 2400 times more than its amount in wastewater and was 6,4 kg/t. Organically bound chlorine value in AS samples for 2005 was one-fourth of its previous content – 1,5 kg/t due to the mill's shift to ECF bleaching including oxygen stage.

Total chlorine content in recycle sludge suspension during its passing over the channel – from the aerotanks through the secondary settling tanks and back to the inlet proved to be lower by 6,3%. At the same time a share of organically bound chlorine increased from 91% to 97%. Total chlorine content and organically bound chlorine share in the AS sample taken in 2005 after its drainage through the press-filters up to 70% moisture content and 1 month exposition in the open air reduced by 35% and 80%, respectively.

#### Results of plant testing.

- 1.Common orach (Atriplex patula) an annual plant is rather sensitive to high organics dosages. The reference plants are undersized (20-25 cm) and have no organically bound chlorine. With introducing 2000 t/ha of AS the plant roots grew in AS layer, its shoots being 3 times as high as common ones without AS addition (75-83 cm); total chlorine content increased by 6,4 times (up to 2 237 mg/kg), organically bound chlorine was not detected.
- 2. Couch-grass (*Elytrigia repens*) is a perennial long-root plant with 2-3 year shoots; the roots are in the top soil layer. It has a good growth response to organics. Therefore, at the addition of a single dose 2000 t/ha its shoots became higher by 25%. Total chlorine content in the growing samples was twice as much as that of the blossoming ones. Organically bound chlorine was detected only in the beginning of growing phase, the tested and reference plants having a slight difference in its concentration 6,7 and 5,6 mg/kg or 0,09% share.
- 3. Yellow alfalfa (*Medicago falcata*) a perennial plant having the micorhiza; the roots deeply penetrate into the subsurface (up to 1-2 cm and more). Growing of the species slightly depends on organics level because these plants are able to take water and nutrients from the bottom clay layers of

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sandy loam. So, the species are an indicator of the part of organically bound chlorine that has been introduced with AS into the soil. Organically bound chlorine was not found in the samples, taken from the soil with the sum dose 2000 t/ha introduced in the period 1983-1995. The plants growing in the lowland contained only traces of organically bound chlorine: 1,1 mg/kg; total chlorine amount was lower (638 against 1100 mg/kg). With introducing of extra single dosage 2000 t/ha the plants had a stress (shoots height was 50-55 cm against 70-88); organically bound chlorine content rose to 6,3 mg/kg (organically bond chlorine share – 0,17%).

4. Rhaponticum carthamoides – a long-live plant, the roots are at a depth of 0-30 cm; its growing is independent of organics level in the soil. The leaves have higher chlorine content as compared with other species that may be explained by its more developed rootage and perennial shooting. A tendency toward increasing of organically bound chlorine content in ontogenesis was observed: the four-year reference plants growing in the clay loam comprised 21 mg/kg, ten-year ones - 35 mg/kg. The lowest levels of organically bound chlorine were in the plant stems and seeds, the perennial parts -roots and radical buds formed on the roots – had the biggest content. 9-year samples growing at the sandy soils with AS dosage 300 t/ha (1994-1996) accumulated 24 mg/kg of organically bound chlorine (total chlorine content was 2700 mg/kg). Organically bound chlorine level in the plants growing on loamy sand with clay bottom layer with AS background level 1500 t/ha (1983-95) was 57,77 mg/kg (total chlorine 2700 mg/kg). Single AS dose 2000 t/ha did not result in changing of growth rates and increasing of total and organically bound chlorine contents: 9890 mg/kg (total chlorine) and 46,9 (organically bound chlorine). After reaching the highest productivity in blossoming phase the levels of organically bound chlorine reduced 2,4-3,4 mg/kg (0,10-0,18%).

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