DEPOSITION FLUX AND ATMOSPHERIC BEHAVIOR OF OIL SHALE COMBUSTION AEROSOLS

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The atmospheric behavior of oil shale combustion fly ash aerosol was studied under simulated conditions. The fine (respirable) fraction of fly ash particles, which contributes most to the health effects of the aerosol, significantly increased (from 25 to 65 % by mass) in time scale. The toxic elements and carcinogenic PAH adsorbed on the fine particles of fly ash could represent a health hazard for human. The bulk and trace elements in emissions (flue gases) and integrated deposition samples were also investigated based on original measurements and literature data.

Introduction

Oil shale, which is a low-grade fossil fuel, is found in many countries over the world [1, 2]. Estonia's oil shale has long been commercially used to produce raw material for industry and for electric power generation [3]. The main technology of processing oil shale in power stations is the combustion of the pulverized fuel. The atmospheric emissions from the power plants have been studied in detail by Aunela *et al*. [4] and Häsänen *et al*. [5].

This process provides, in addition to gaseous products (SO_2, NO_x, CO_2), a high amount of the airborne particulate matter. The particulate fraction is referred to as fly ash and includes particles of different types (carbonaceous, inorganic ones) and different sizes. It is estimated that two large power plants in Narva (Northeastern Estonia, Fig. 1), at full capacity of 3000 MW, have produced more than 300 million tons of ash including about 5 million

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tons of fly ash, which is emitted directly to the atmosphere [6]. Deposited fly ash can cause environmental problems not only at a local level but also for other European countries, particularly Finland and Russia [7].

In the present study the atmospheric behavior of emission from oil shale thermoelectric power plants is investigated. The aim is to estimate the pollution levels and the distribution as well as the atmospheric fate of emitted fly ash aerosols under simulated conditions.
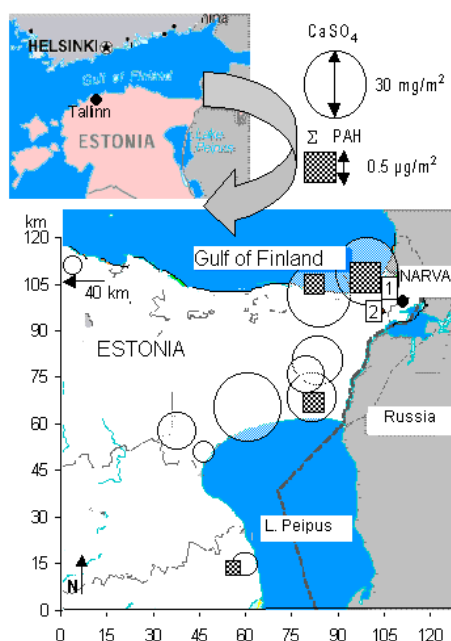
Fig. 1. Map of oil shale area in Northeast of Estonia and pollution load. The oil shale power plants (*1* – Baltic and *2* – Estonian) are located close to the town of Narva. The integrated flux (mg/m^2 per day) of typical mineral components (CaSO$_4$) and organic trace compounds (total identified PAH) is indicated

1. Simulation of Fly Ash Aerosols

The atmospheric discharge of any pollution source could be directly estimated by impactor measurement of stack emissions. The distribution, transport and transformation as well as deposition of airborne pollutants could be investigated mainly by indirect way, by sampling and analyzing of wet precipitation or dry deposition matter. An alternative way is to model the atmospheric processes in a big enough chamber to minimize wall effects, in conditions as similar as possible to real situation. At present two big so-called smog chambers are available for such a modeling study, one located in Europe (Valencia, Spain) and the other one in North America (North Carolina, USA). Our experiments were performed in North Carolina.

1.1. Experimental

Atmospheric behavior of oil shale fly ash was studied at the University of North Carolina 190-m^3 outdoor smog chamber [8–10]. This chamber was a wooden A-frame covered with Teflon film. The chamber leak rate, including sampling, was between 1 and 5 % per hour, as measured by a sulphur hexafluoride tracer. After venting (24 h with rural North Carolina air) and drying (24 h) prior to an experiment, the chamber background particle concentration ranged from 87.5 to 111.9 μg/m^3. Two sets of experiments were conducted, one at night to avoid photochemical reactions and one in the daytime.

The fly ash samples injected into the chamber was taken from the exit gas flow duct from the Baltic Power Plant (location see in Fig. 1). Combustion fly ash was injected into the UNC chamber *via* a high-pressure venture aerosol generator located outside the chamber. The generator had a 0.3 m × 0.1 m hopper, which gravimetrically fed the dust to one of four circular grooves on a horizontal variable-speed 0.3 m aluminum platter. A venture nozzle pulled the dust from the groove of the rotating platter and discharged it into an 8 cm × 3 m duct connected to the chamber. Typically, about 10 g of dust was injected in about 20 min. The fans inside the chamber were turned off before the injection of fly ash in order to reduce deposition and diffusion of particles onto the floor and walls of the chamber.

Particle size and number were monitored during the experiment using different aerosol particle counters:
1. Laser Optical Aerosol Counter and PMS Model LAS-X CRT
   (90 nm – 3.0 mm)
2. Electrical Aerosol Size Analyzer, Model 3030 (3.2 nm – 1 μm)
3. Laser Optical Aerosol Analyzer, CSASP-100-SP (0.3–20 μm)
4. TSI Electrostatic Classifier (for size 14.3–697 nm)
   The total range of recorded particles was from 3.2 nm to 20 μm.

Temperature and humidity were measured periodically using a thermistor and light-scattered dewpoint meter (EG&G Model 800, Waltham, MA). The duration of each experiment was between 4 and 6 h.

Particles were also collected on filters and gaseous components by denuders. The nucleopore filters were used for collection the particles for SEM imaging. Mass concentration was calculated from the mass of the particles and the sample gas volume. The filters were Soxhlet extracted for 6–8 h with a 20 : 30 : 50 mixture of dichloromethane : acetone : hexane. The organic compounds in gas and particle phases were analyzed by gas chromatography-mass spectrometry (GC/MS) using a Hewlett-Packard 5890 Series II gas chromatography with a J&W 30m DB5 column and a Hewlett-Packard 5971A MSD mass spectrometer. For details on analysis and experimental techniques, see [9, 10].

1.2. Distribution and Deposition of Particles in Air

The initial fly ash particle concentration in the smog chamber ranged from 15 to 20 mg/m^3. After the injection of fly ash into the chamber, the aerosolized particles deposited quickly, whereas the change in particle number concentration with time proceeded slowly (Table 1).

Besides the mass and number concentration the information about the size distribution of fly ash particles is needed for the calculation of dry deposition characteristics. On the basis of the dry deposition rate, the susceptibility of aerosol to long-range transport and environmental impact can be evaluated. At the beginning of the experiment, the coarse fraction (around 6.9 μm) accounted for most of the particle mass. It is important to note that the particle size distribution injected into the chamber [10] was similar to the distribution of fly ash particles in flue gases of Estonian Power Plant as measured with a modified Berner low-pressure impactor [4]. The size distribution of fly ash particles ten minutes after the injection (Table 2, and [10]) showed a bimodal composition with the first maximum at 0.1 μm (fine particles) and second maximum around 3.5 μm (coarse particles). After one hour aging of the aerosol in simulated conditions the mass peak was estimated around 2.5 μm, and after 4–6 hours it decreased to 1.7 μm due to deposition of the bigger particles.

Table 1. **Dynamics of Mass and Number (Count) of Fly Ash Aerosol Particles in Chamber Experiments**

Time, min	Mass, %	Count, %
0–10	100	100
37–42	51	69
78–81	24	50
125–127	11	43
182–193	7	36
232–246	4	31

Table 2. **Dynamics of Prevailing Fine and Coarse Fractions of Ash Particles during Aging of Fly Ash Aerosol**

Time from injection, min	Mass median diameter, μm	
	Fine	Coarse
0	0.35	4.47
42	0.31	3.63
81	0.32	3.09
125	0.37	2.39
217	0.35	1.82

In general, the potential hazard of solid particles present in inhaled air depends on particle size and number concentration [11, 12]. Cecinato *et al.* [13] demonstrated that only the <10 μm (PM10) atmospheric particulate range may be inhaled into the deeper respiratory tract and cause severe health effects. Particles with diameters greater than 10 μm do not reach the thorax; particles ranging from 2.1 to 10 μm are preferentially retained by the

pharynx, trachea and bronchi, and particles smaller than 2.1 μm can reach the terminal bronchi and alveoli and have the strongest impact on human health.

Based on chamber experiment data, we calculated the role of inhalable (10 μm), thoracic (<10 μm, includes also the smaller particles), and respirable (<2.1 μm) fractions of fly ash aerosol during aging (long-range transport). The results illustrated in Fig. 2 clearly demonstrate that the role of the respirable fraction of particles, which contributes most to the health effects of the aerosol, significantly increased in time. Here, 25 % were in the respirable fraction immediately after the injection of fly ash into the chamber and this increased to 65 % by the end of the experiment [10].
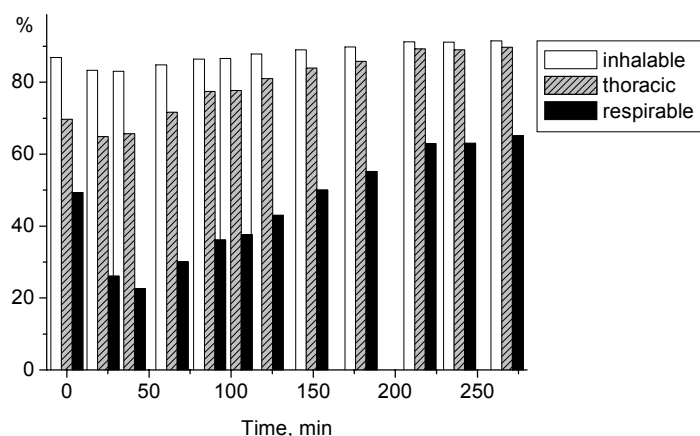

Fig. 2. Distribution of inhalable, thoracic, and respirable fractions of fly ash particles during aging of fly ash aerosol

2. Chemical Composition of Emission

From the model experiment (section 1.2) is evident that during the aging, e.g. long-range transport of fly ash particles, significant changes between the size fractions occurred. From the aspect of environmental health not only the size of particles but also chemical composition of aerosol is relevant.

Alkaline oil shale itself is characterized by high concentrations of several heavy metals, carbonates, sandy-clay minerals and also harmful organic substances [6, 14–16]. Continuous monitoring was used for the analysis of SO_2 and NO_x in flue gases of both Narva power plants by Aunela *et al.* [4]. The high average emission factors of SO_2 per fuel energy unit (mg/MJ) were stated in the range of 820–1360 for different boilers. Corresponding numbers for NO_x were found in the range of 90–100.

Particle average concentrations in flue gases was estimated at 2250 mg/m^3, but the actual range was 1730 to 3320 mg/m^3 for emissions of the Baltic Power Plant [4]. The oil shale fly ash consisted mainly of mineral

oxides with a high content (30 %) of CaO [14, 16]. At elevated temperatures, the reaction between CaO and SO_2 is rapid and $CaSO_4$ is produced. Häsänen *et al*. [5] provided results of chemical analysis of different size fractions of oil shale fly ash. Al, Ca, Fe, K and Mg are major constituents in emissions from oil shale fueled power plants.

As can be seen from the elemental composition of two fractions, above and below 6 μm (Fig. 3), As, Tl, V and Cd were essentially enriched, and Sb, U, Cr and Zn were enriched in the fine fraction of fly ash particles of both power plants studied [5]. At the same time, the concentration of Co, Ni and Sr in both fractions was very close. Thus, the chemical composition of various size fractions of fly ash is different. Hence, the concentration of toxic trace elements in fly ash usually increases with decreasing grain size. High enrichment levels of oil shale fly ash in the fine particle fractions have been reported by several authors [3, 6, 14, 16, 17]. Thus, exposure to toxic trace elements would appear to be related to their associated particle sizes.
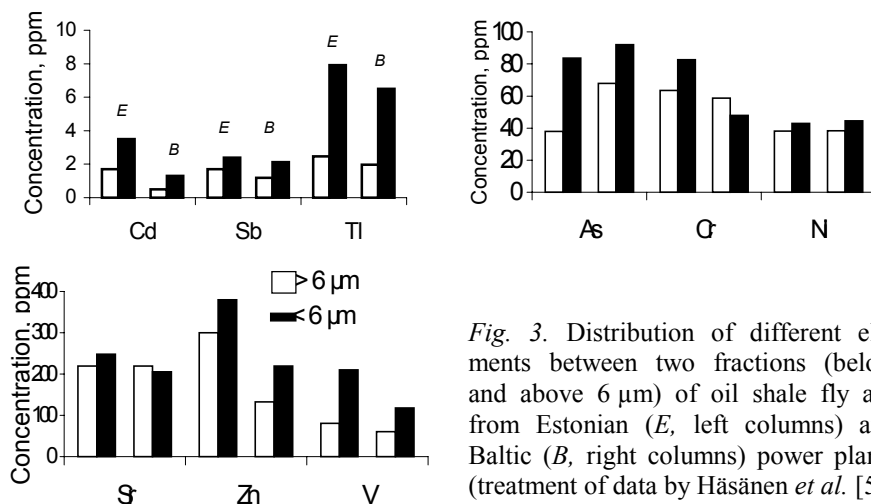

Fig. 3. Distribution of different elements between two fractions (below and above 6 μm) of oil shale fly ash from Estonian (*E,* left columns) and Baltic (*B,* right columns) power plants (treatment of data by Häsänen *et al.* [5])

The estimated concentration of organic trace compounds (total polycyclic aromatic hydrocarbons, PAH) in flue gases of oil shale thermal power plants ranged from 0.04 to 3.16 mg/m^3 [4]. The solvent-extractable fraction from fly ash particles collected from the smog chamber included several PAHs (phenanthrene, fluoranthene, pyrene, benz(a)anthracene, chrysene, benzo(b)-fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, which are considered highly mutagenic and/or carcinogenic [18].

3. Monitoring of Integrated Deposition

The more or less permanent snow cover in Nordic countries can deposit different persistent airborne pollutants, including those in water-soluble and non-soluble particulate phases. Accumulation of various compounds in snow

proceeds *via* dry deposition as well as wet precipitation. Thus, by analysis of snow-water samples, an integrated pollution flux for a relatively long period of time can be estimated. We had the opportunity to use this approach in the spring of 1987 after an unusually long period (95 days) of continuous snow cover, and conducted a detailed study on the bulk deposition chemistry in the oil shale combustion area (see Fig. 1) of Estonia [19]. Anions (calcium, potassium, sodium) and cations (chloride, sulfate, nitrate), which are characteristic for the emissions of the oil shale combustion power plant, and additionally PAH, were analyzed in snow-water samples from 21 sampling stations in Estonia.

3.1. Experimental

Precipitation samples were collected according to the methodology developed by the former Institute of Chemistry (Estonian Ac. Sci) using the following procedures [19]: snow samples were collected at several sampling points (each area was 87 cm^2) ranging from the snow surface to the ground by cylindrical sampler ($\varnothing$ 105 mm). The distance between the sampling points was 4–5 m. Snow samples were melted at room temperature and yielded approximately 900–3000 ml snow water depending on the thickness and density of the snow and the number of samples taken.

Content of anions (Cl^-, NO_3^- and SO_4^{2-}) in snow water samples was analyzed using ion chromatography and cations (K^+, Na^+, Ca^{2+}) by atomic absorption spectrophotometry (AAS).

The *n*-hexane extract of snow water was analyzed with respect to PAH by HPLC (*Perkin Elmer* 3B) using UV and fluorescence detectors. Content of the target PAH, benzo(a)pyrene (BaP), was additionally quantified by spectral-luminescence method (Shpolskii).

For collection and analysis of snow-water samples see [19].

3.2. Results of Chemical Analysis

The concentration of mineral and organic compounds in snow-water samples (data not presented in this publication) was found to be high [19]. The typical mineral components of oil shale, i.e. Ca^{2+} and SO_4^{2-} accounted for over 92 % of the cations and 90 % of major anions in snow-water samples. Calcium was the dominant cation in air and in precipitation in terms of mass. Correlation between the major ions in bulk deposition for Ca^{2+}/SO_4^{2-} and $Ca^{2+}/(SO_4^{2-} +2Cl)$ gave correlation coefficients $r = 0.86$, and $r = 0.92$, respectively. The high correlation ($r = 0.83$) between the calcium concentration and the total amount of identified PAH in snow samples taken from different locations around (150 km) thermal power plants indicates the strong impact that these sources can have on local airsheds.

The flux decreased with distance. The medium Ca^{2+} load in the town of Mustvee (90 km southward from power plants) was 2.7 mg/m^2 per day, which is four times lower than that from close to the power plants – 10.7

mg/m^2 per day (see Fig. 1). The deposition load (μg/m^2 per day) of total PAH in the power plant area was estimated at 0.47, whereas only 0.16 in Varnja, 90 km southwest.

The fact that calcium and other oil shale power plant emission components have been transported over a distance of approximately 100 km southwest is illustrated in Fig. 4, where the deposition of calcium *versus* snow-water pH is depicted. It is clearly seen that the snow-water pH decreased from the distance to oil shale power plants. Thus, the acidic load from Central Europe has been neutralized by the basic cation deposition of power plant discharge.
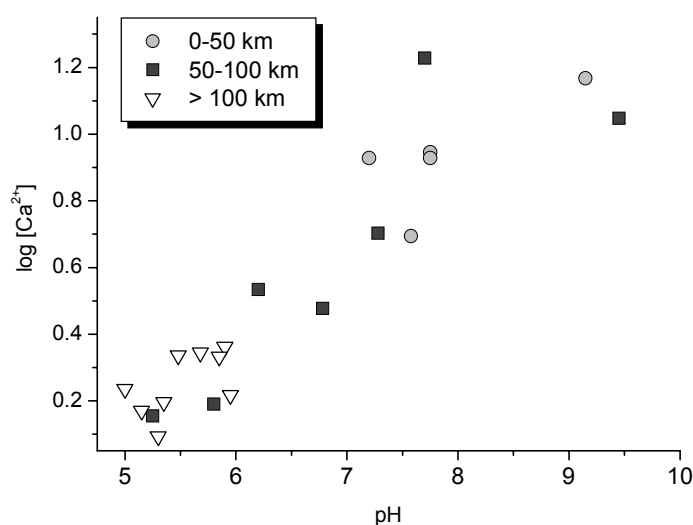

Fig. 4. Concentration of calcium versus pH in snow-water samples taken at different distances from Narva power stations

4. Discussion

4.1. Atmospheric Transport of Fly Ash

It is well known that the atmospheric long-range transport and deposition load of pollution are highly dependent on appropriate meteorological conditions, e.g. wet precipitation intensity and wind direction and speed. Figure 5 illustrates the wind speed in the town of Narva during January and July 1996, calculated from monitoring data [20]. It is possible to evaluate the distribution of emissions between different locations and surrounding countries by considering these average wind directions and wind speeds.

According to this very formal approach, in the wintertime (January) Russia could receive up to 32 % and Finland even 48 % of the total discharge of pollution, whereas Estonia would be left with only 20 %. In the summertime (July) the general distribution of atmospheric pollution would be different,

e.g. 56 % for Russia, 23 % for Finland and 21 % for Estonia. It means that the major elements as well as trace compounds were not evenly distributed around the power stations.
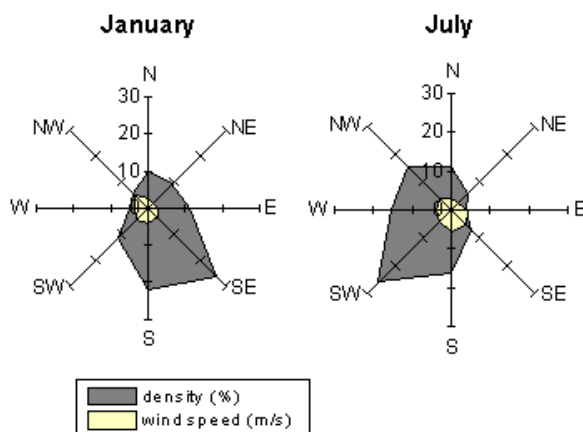

Fig. 5. Average monthly wind directions and densities in Narva
power plants area in 1996 (calculated from the data given in [20])

The emission patterns also showed a clear relationship with the prevailing wind directions in sampling locations in the territory of Finland. In the chamber experiment, we investigated the fly ash particles by Scanning Electron Micrograph (SEM) images of samples taken at different times after injection. The prevailing structures were regular spherical symmetries [10]. Jalkanen *et al*. [7] reported similar shapes of oil shale fly ash particles collected from flue gases as well as from moss-surface samples within different regions of northeastern Estonia and southeast Finland. Their shape and size as well as chemical composition was characteristic of the fly ash particles.

In general, calcium and other oil shale power plant emission constituents have been transported over a distance of approximately 140 km to Finland, also in the coarse fraction. The major elements of fly ash (Ca, Al, Fe) emitted from oil shale-fueled power plants close to the town of Narva showed a clear decreasing gradient downwind from the power plants. The annual emissions of many elements, both toxic and non-toxic, from oil shale-fueled power plants are large. The calcium emissions are so large that they have a marked effect both in neutralizing the SO_2 in the flue gases and in neutralizing environmental acidification over large areas of northeastern Estonia and even southeastern Finland [21]. The airborne particulate matter emitted from Narva power plants according to total calcium content represents approximately 5 % of total European emissions [7].

Nevertheless, a significant amount of particle mass rapidly settled close to the discharge site. This was due mainly to coarse particle settling. The deposition of these particles will have an impact on the terrestrial and water environment not far from the pollution sources (see **3.2** and Fig. 1).

4.2. Further Perspectives

Oil shale combustion in Narva power plants was and is based on pulverized fuel technology. However, the renovation of technology to more effective fluidized-bed (FB) combustion is currently under way. How could the results received in smog chamber simulation experiments be applied for modeling the atmospheric behavior and size distribution of fly ash aerosols?

The key issue would be conditions of the combustion process itself. It is known that some redistribution of trace elements between different phases of fly ash occurs during combustion, i.e. during the cooling stage the vaporized semi-volatile trace compounds from the oil shale condense on the fly ash particles [14].

This process was detailed studied for coal combustion [22], where the fly ash also exhibits a bimodal size distribution, mostly in the 1–20 μm range and a small amount in the submicron range. The two size modes are formed by different mechanisms. During the combustion of coal the mineral matter associated with coal particles (clays, carbonates, and pyrites) decompose, fuse, and adhere to the carbonaceous surface. This process yields primarily residual ash particles in the size range of 1–20 μm. In parallel, a small portion of the mineral matter is vaporized and condenses to form submicron particles, which grow to about 0.05 μm [22]. Oxides of Fe, Ca, and Mg, which condense first and form the core of aerosol particles, show no strong size dependence. The outer surface of the aerosol particles is composed primarily of semi-volatile species, and the particle loading of these compounds is proportional to the reciprocal of particle diameter [22].

In FB process the decreasing amount of coarse particles and increasing of fine particles is expected. In aspect of environmental impact, especially human health, it supposed to be not an effective solution of problem (see above, section **1.2**). If coarse particles are removed efficiently from flue gases in power plants, and gaseous pollutants are not removed, this should result in acidification of precipitation in the eastern Gulf of Finland region [7]. More serious problem could be related to the content of PAH in combustion discharges, because the combustion temperatures in FB processes are expected to be lower, e.g. 100–200 ℃, compared with the pulverized fuel treatment [23]. It is known that the content of PAH compounds in flue gases could arise 1.5 to 2.9 times decreasing combustion temperature to 100 ℃ lower [23].

Consequently, at present there is lack of date for the prediction of environmental impact of discharges of FB process of oil shale combustion. Further investigation would be essential for solving the problems like these.

Conclusions

- By using the simulation experiments in big smog chamber it is possible to model the atmospheric transport of fly ash aerosols. The relative part of

fine (respirable) fraction of fly ash particles in the atmosphere significantly increased in time or during the transport due to deposition of bigger particles.

- Massive calcium, sulfate and chloride content were observed in snow-water samples around the oil shale-fueled power stations. The acidic load in emissions was neutralized by the basic cation deposition.

- The content of toxic metals (As, Tl, V and Cd) was essentially enriched in the fine fraction of fly ash particles. Based on the data from literature and experimental results obtained in this study on deposition flux and atmospheric transport of oil shale fly ash aerosol, we can conclude that the use of existing technology for combustion creates serious environmental and health problems. A significant improvement in waste removal and recycling systems is needed for continued commercial use of oil shale deposits in the world.

Acknowledgements

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