

Greenhouse gas emissions from liquid fuels produced from Estonian oil shale

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Adam R. Brandt

Department of Energy Resources Engineering
Stanford University
Green Earth Sciences 065, 367 Panama St.
Stanford, CA 94305-2220

Email: abrandt@stanford.edu
Phone: +1-650-724-8251

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1 Estonian oil shale

Oil shale is broad classification for sedimentary rocks that contains fossil organic matter called kerogen. Kerogen is believed to be the source material from which naturally occurring oil and gas were formed over geologic time [1, 2]. Kerogen is made of organic macromolecules, and can be burned directly to release heat. Kerogen can also be converted to liquid and gaseous fuels through retorting processes. Commonly, retorting involves heating in an oxygen-free environment, but other chemical reaction schemes can be utilized (e.g., heating with supplemental hydrogen) [2].

There are two varieties of Estonian oil shale, Argillite and Kukersite [3]. Kukersite is the primary type of oil shale mined in Estonia. Oil shale properties vary significantly across formations, as well as within formations, with some layers being sub-economic due to low energy density (i.e., low kerogen content) [3]. Oil shale consumption is the largest source of CO₂ emissions in Estonia, representing some 72% of combustion CO₂ emissions [4]. Total consumption of oil shale in Estonia in 2002 was 116.5 PJ, of which 88.3 PJ were used for electricity and heat generation and 25.9 PJ were used for oil production [5], with the remainder used for cement production feedstock.

Liquid fuels are produced in Estonia using two different retorts: the Kiviter and the Galoter retorts. The Kiviter process is an internal combustion process fueled by produced gases and residual kerogen char (see below). The Galoter process relies on recycling of hot solids (hot ash from the combustion of shale char) from an external combustion chamber. Operations using the Kiviter retort processed approximately 3 times as much shale as the operation using the Galoter retort in 2002 [5, Figure 5].

Liquid hydrocarbons produced from Estonian oil shale are of relatively low quality.

Approximately 11.9 PJ of shale oil was produced in 2002 (≈ 2 Mbbbl/y), of which 53% was exported and the rest was consumed domestically for heat and electricity generation [5]. The oil shale liquids industry in Estonia is somewhat subsidized by the oil shale electricity industry: liquid fuel producers have access to low-cost mining residues (lump shale) that would not be generated without primary demand for shale from the electricity industry [3].

2 Oil shale CO₂ emissions

Oil shale emissions in Estonia arise both from combustion for electricity generation and conversion of oil shale to liquid fuels. This review will focus on CO₂ emissions from converting oil shale to liquid fuels, such as synthetic gasoline or diesel.

Conversion of oil shale to gaseous and liquid hydrocarbons can occur in place in the geologic formation, called *in situ* retorting, or in above-ground retorts, called *ex situ* retorting. Nearly all oil shale retorting is currently *ex situ* retorting, including retorting of Estonian oil shales [3]. *In situ* retorting is under development, but not practiced at commercial scale.

Carbon dioxide is directly emitted in the three primary stages of producing, refining, and consuming liquid fuels from oil shale [6]. First, emissions result from the retorting of oil shales to generate unrefined hydrocarbons (HCs), including crude shale oil and HC gases. Second emissions result from the upgrading and refining of crude shale oil to refined fuels (e.g., gasoline or diesel). Third, emissions result from combustion of the refined fuel by the consumer. Additionally, there are minor indirect emissions from the consumption of materials such as steel or cement used in oil shale extraction.

Emissions from oil shale retorting process can be divided into three components: CO₂ emitted due to the thermal energy requirements of retorting, CO₂ emitted from other energy uses associated with retorting, and CO₂ emitted from the shale itself [6]. These emissions sources will be discussed in order.

The thermal requirements for retorting require the addition of thermal energy to the raw oil shale. This is often done by burning produced shale gas, shale “char,” or coke generated during oil shale retorting, or using an auxiliary heat source (e.g., imported natural gas or electricity). These thermal requirements for retorting result from having to heat the inert mineral matter surrounding the kerogen, having to heat the kerogen, and having to fuel the kerogen decomposition reaction as well as other mineral phase reactions [6]. In addition to the thermal requirements for retorting, emissions result from other process energy consumption, including device operation (e.g., pumping, materials handling, or rotating of kiln), product separation (e.g., oil, gas, water separation) or primary upgrading of produced products. These requirements are typically small in comparison to the thermal demands of retorting.

In addition to emissions from energy consumption, CO₂ is emitted from the shale itself during retorting. These emissions result from a multiple processes. First, kerogen contains some oxygen in its native state, some of which reacts with carbon during retorting to form CO₂ (oxygen also forms water vapor through reaction with hydrogen). More importantly, high temperatures can drive decomposition of carbonate minerals (e.g., CaCO₃) to mineral oxides and CO₂ (e.g., CaO + CO₂). This is especially important during combustion of oil

shale, as in Estonian oil shale-powered electricity generation [7, 8, 4, 9], or in cases where the retorting system is fueled with spent shale residues.

Refining emissions result from upgrading crude shale oil into finished refined products (e.g., gasoline and diesel). These emissions are generally going to be larger than refining emissions from conventional crude oils [6], because shale oil is generally of low quality, especially from *ex situ* retorting. These quality shortfalls include: instability and propensity to form gummy compounds, suspended fine solids from retorting process, high nitrogen content (often $\approx 2\%$, or an order of magnitude larger than conventional crude oils), and high metals content (e.g., arsenic, iron and nickel) [10, 11].

Combustion CO₂ emissions from burning resulting oil-shale-derived liquid fuels will generally be equivalent to those from burning conventionally-derived fuels, because finished products are refined to specifications that fix their CO₂ emissions (e.g., makeup of shale-derived diesel fuel will be virtually identical to crude-oil-derived diesel fuel) [6].

For more information, these CO₂ emissions are outlined in detail in other resources [6, 12, 13, 14, 15, 16]. These sources in general focus on the Green River formation of USA, as that is the preeminent global oil shale resource, and has received the most research attention. In practice, there is a large range of CO₂ emissions intensities from oil-shale-derived fuels. Much of this variability results from process variation, while some results from shale quality variation. The resulting range of CO₂ emissions for producing fuels from oil shale, for Green River oil shales in the United States, are shown in Figure 1. These studies are in general based on process modeling of pilot-scale processes, although some studies attempt to model full-scale operations [12, 13].

3 Previous work on oil shale emissions in Estonia

Previous work on CO₂ emissions from oil shale extraction and conversion in Estonia is limited.

Significant recent research has focused on oil shale CO₂ emissions from electricity generation [9, 5, 4, 8, 7, 17, 18, 19, 20]. This is because oil shale consumption for electricity generation is over 3 times larger than consumption for oil production, and emissions from oil shale combustion from electricity are higher than combustion emissions from using coal for electricity generation. Because of the existent oil shale industry in Estonia, these emissions estimates are based on values for current operations, and are therefore not as speculative as the above estimates of Green River oil shale GHG emissions.

The most comprehensive life cycle assessment of the oil shale industry in Estonia includes significant information on air, water and solid waste impacts [5, 21]. Unfortunately, CO₂ emissions from oil production are not calculated in these studies, in that only CO₂ emissions from electricity production are calculated.

Emissions from Estonian oil shale facilities that generate liquid hydrocarbons are uncertain: no estimates were found of CO₂ emissions from Estonian oil shale operations. Given the similarity of oil shale processing technologies, it is plausible to assume that CO₂ emissions from Estonian oil shale fall within the range of uncertainty outlined in Figure 1 above for the ATP retort (e.g., 1.4-1.6 times those of conventional oil). This similarity is due to the fact that these retorts are fueled using shale char combustion, as is the ATP retort. It is unclear what differences in Estonian shale quality will have on these retorting emissions.

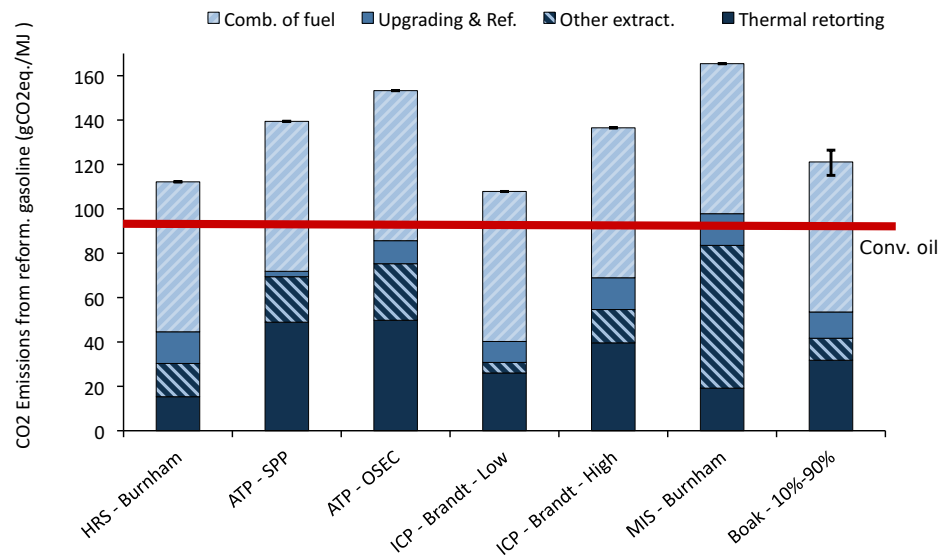


Figure 1: Oil shale life cycle emissions compared to conventional oil (red line represents moderate case assumptions about conventional oil GHG intensity). Data sources: [12, 13, 6, 16]

Without further research into the specifics of the Kiviter and Gaolter technologies, and the characteristics of Estonian shale, more detailed assessments cannot be made.

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