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CIRCULAR ECONOMY FOR RESOURCE CONSERVATION, GHG EMISSION REDUCTION AND CARBON SEQUESTRATION

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Abstract:

Increasing population and quality of life put more pressure on finite natural resources, both organic & inorganic and also disposal of waste generated during manufacturing, use phase and after use. This is becoming growing challenge today due to Linear Economy model currently practiced in the majority of the areas. On the other hand Circular Economy model offer suitable and sustainable solution for the issues faced by the humanity today such as resource scarcity, waste disposal, green house gas (GHG) emissions and associated climate change. The present study is focused on manufacturing process adapted in Pulp & Paper, Filler and Cement manufacturing in TNPL based on Circular Economy model for waste & by-product utilization, resource conservation, GHG emission reduction and carbon sequestration. The study evinced that for the five year period, from 2017-18 to 2021-22, total 21,679 KL fuel oil saving by utilizing biogas generated from organic waste from wastewater, 1.33 million MT Coal by the use of biogenic waste put together replacing 30,631 TJ of fossil fuel energy with renewable energy, 3.09 lakh MT Lime stone saving by using inorganic waste. The foresaid process contributed 3.47 million MT CO_2e GHG emission reductions. Finally, the carbon sequestration pathway, from atmospheric CO_2 to Paper filler through sugarcane, Bagasse pulping, Chemical recovery, Cement and PCC manufacturing, is established and explained using data collected from above process with published scientific evidence. The study indicates that 31,468 MT CO_2 is sequestered by adaption of foresaid manufacturing process as per the principles of Circular economy model.

Keywords: Circular economy, Renewable energy, Resource conservation, GHG emission reduction, CO_2 sequestration.

Introduction:

Resources from natural environment viz. land, water, air are continuously exploited and used for economic activities to produce goods and services. These include bio-based (crops for food, energy, medicines and other industrial uses), fossil fuels (coal, gas & oil for energy and goods), metals, and non-metallic minerals, notably sand, gravel and limestone[1]. Global resource extraction and material utilization has tripled, from 27 billion tonnes in 1970 to 92 billion tonnes in 2017 and growing rapidly with minor decline during the Covid 19 period and started resuming back again. Reducing impact of energy and organic and inorganic material extractives systems on the natural ecosystem requires rapid switch over to circular and resource efficient models. This will reduce the amount of new resources required to support growing population for sustainable living and to maintain the ecosystem function without much damage. Also Identified oil, gas, metal and mineral reserves are increasingly difficult to extract and become expensive and environmentally unsustainable[2,3].

The, International Resource Panel forecasts that by 2050 material use will reach between 170 and 184 billion tonnes[1,2]. Majority of these are non-regenerative and non circular for example, fossil fuels used for energy and combustion engines of vehicles referred as short lived. Secondly, the build-up of physical assets in utility infrastructure is on the increase due to raising provision of services such as energy, water, sanitation, housing, communication, mobility and capital equipments and are unavailable as secondary feedstock for as long as they remain stored and in use and referred to as "Products that Last and require new thinking on design". On a positive note, many parts of the world, recovery rates are on the increase. This is steered by technical innovation and investment directed at increasing material efficiency, extending and intensifying use and enabling end-of-life recovery. Recycling rates have also been improving over the years and reduce solid waste generation and disposal. But ever-increasing rate of material extraction is overtaken waste recycling to fuel our growth. The yield of secondary materials is, therefore, simply not sufficient to feed our hungry economy on its own[2]. Only with a radical reset of extraction, production and consumption processes can decouple natural resource use and environmental impacts from economic progress and human well-being. Adopting resource-efficient, nature-positive strategies based on circular model and improving societal resilience can accelerate this shift[4].

The present study focused on utilization of both solid and liquid organic wastes generated in TNPL as renewable energy or converting into renewable energy for in-house use and associated GHG reduction. Similarly utilization of inorganic wastes such as, first stage lime sludge from causticizing process, lime grids and Power boiler fly ash as raw material to make Cement. Also to workout circularity carbon path way from bagasse or wood, Black liquor, Green liquor, fist stage lime sludge, cement production, PCC production and then papermaking.

Review of Literature

Current consumption trends of natural resources for modern lifestyle, increasing population and quality of life put more and more pressure on environment and climate and it is unsustainable. For example at the current consumption rate by 2050 India may require 2.7X India[5]. Similarly influence of unsustainable human activity is increasingly visible day by day and left with little time to change manufacturing practices and life style from Linear to Circular model[6,7]. Sustainable production and consumption address the entire life cycle of economic activities from the extraction of natural resources, production, use, and finally disposal of resources. It is necessary to decouple economic development and its negative impacts on the environment and mankind. This can achieve only when resource use and pressure on the environment or human well-being grows at slower rate when compared to activity which causing it[3].

Circular and resource-efficient models are based on principles which differ from the current "take make, use and waste" models. Circular model improve Ecosystem and economic efficiency and reduce environment impact by efficient use of resources, reuse, recycle of resources, waste reduction & utilization, redesign, refurbish and method of product disposal after use such as energy, raw material for other process or product and service[4,6]. Pulp and paper manufacturing is more sustainable when compared to other manufacturing industry. Pulp and paper industry is pioneer in adopting resource efficient circular model within the various process boundaries and beyond. For example, paper recycling is a relatively mature practice and recycling rate is crossed 60% and expected to increase significantly over the next few years. In addition to resource recovery this would save 40-50 billion tonnes of CO_2 emissions between 2021 and 20406. Carbonate sludge, a waste from chemical recovery is partially fed into the lime kiln to produce burnt lime and then, to white liquor. The surplus carbonate sludge is used as a raw material for production of onsite Precipitated Calcium Carbonate to reduce virgin raw materials. Similarly, Kraft lignin is used to manufacture graphite for consumer electronics and the automotive industry[8 9]. Using biocarbon from wood as an alternative to fossil carbon sources not only helps to substitute fossil-based materials with renewable, but also enables carbon capture from the atmosphere, helping to create a low net-carbon emission economy[8].

The possibilities of Carbon Capture and on-site utilization in pulp mills as has been recognized as one of the important strategies on climate change mitigation. Main CO_2 sources in the integrated pulp and paper mill are Lime kiln, chemical recovery and power boiler. Large part of CO_2 formed in the pulping process is biogenic, and the primary source of fossil-based CO_2 is the lime kiln and coal based power boiler. Scope exists to capture biogenic CO_2 from Kraft process and permanently remove from the atmosphere by utilizing it as raw material for other products[10, 12, 13]. Similarly potential for Capturing biogenic CO_2 and using as raw material for bio-products include tall oil manufacturing, lignin extraction and use as renewable energy in Lime kiln and production of precipitated calcium carbonate was reported elsewhere [10, 11,12]. Application of inorganic sludge such as green liquor dregs, slaker grits, lime mud, and fly ash from the pulp and paper mills, based on chemical and physical properties, are reviewed to identify the suitable applications in other industrial process. The applications are grouped into construction materials (Cement, Clinker production, Ceramic wall tiles, Ceramic building materials), Environmental (Acid mine drainage, Landfill cover), Agricultural (Soil amendment and fertilizer) and others to avoid landfill[14]. From the literature it is understood that concept of circular economy in pulp and paper industry for resource conservation, CO_2 emission reduction & sequestration is broadly classified into two, one is utilization organic for renewable energy within the plant and in-organics as raw material within or outside the plant.

Materials and methods

All qualitative analysis are made as per the standard procedure documented in the TNPL ISO:IMS Laboratory manual (IMS/R&D-LAB). The calculation of the GHG emission reduction and CO_2 sequestration is made as per the published literature for pulp and paper industry[15].

Results and Discussions.

Tamilnadu Newsprint and Papers Limited (TNPL) initially started with the motto of utilizing bagasse, sugarcane waste from the sugar mill, as raw material to manufacture writing and printing paper to conserve the wood which is used predominantly to manufacture paper. TNPL initiated and successfully implemented and reported many project towards conservation of the natural resources, such as, water: through conservation, reuse and recycle, wood fibre: with bagasse and wastepaper, Fossil fuel: with bagasse pith, wood dust, black liquor solids and biogas[16 -20]. Also the inorganic solid wastes such as lime sludge, lime grids and fly ash are utilized for manufacturing cement through Lime Sludge and Fly ash Management system (LSFM). In addition to the above, CO_2 emission reduction by utilizing renewable energy to replace fuel oil, replacing anaerobic lagoon with closed high rate bio-methanation plant to avoid methane emission and to reduce the impact of climate change[21,22].

Utilisation biogenic liquid waste

The high rate bio-methanation or biogas plant 1 with two Upflow Anaerobic Sludge Blanket (UASB) reactors and other accessories are commissioned in the year 200322. Considering the increased wastewater generation due to backward integration of CBP fibre line plus long stoppage for replacement of three phase separators, biogas plant was expanded by installing biogas plant 2 in the year 2008 and then plant 3 in 2017 **(Table 1)**.

Wastewater from the Bagasse yard and bagasse washing are containing high COD due to the presence of residual sugar from bagasse which is not extracted from the sugar mill and

Table 1: Biogas plant data

Design	Units	Plant 1	Plant 2	Plant 3
Flow	M^3/day	12,000	9,600	6,000
Total Reactor volume	M^3	10,000	5,000	6,580
Inlet COD	ppm	6000	5120	5,000
COD redun. In reactor	%	85	80	80

also the sugar acids due to the conversion of sugars into sugar acids by fermentation in the wet pile storage[23]. This high COD wastewater was treated in anaerobic lagoon before implementation of biogas plant and the biogas generated in anaerobic lagoon due to the anaerobic digestion is not captured. Implementation of the closed anaerobic digester or UASB, the generated gas is captured and used as fuel in the lime kiln to replace fossil fuel and also avoid methane emission to atmosphere which is second largest GHG contributes to climate change[6,8,22]. In addition to biogas generation, efficient degradation COD in UASB reactors reduce the organic load to forward treatment system viz. activated sludge process and reduce energy consumption. The data on, quantity of wastewater treated, COD reduced, biogas, an in-house renewable energy generation, GHG emission reduction with respect to methane avoidance and fuel oil saving from the UASB reactors from 2017-18 to 2021-22 is presented in the **Figure 1,2 and 3.** The data indicate that the implementation of UASB reactors resulted in environmental benefits such as 78.0 thousand MT COD reduction, 4.83 lakh MT CO_2e GHG Emission reduction due to CH_4 avoidance, 70.4 thousand MT CO_2e GHG Emission from fossil fuel saving, together total 5.53 lakh MT CO_2e GHG Emission reduction in five year. Also 36,127 Mega Lit. of biogas generation resulting 21,676 kilo litters of furnace oil saving as economic benefits to company.

Figure:1 Biomethanation Environmental Benefits: Wastewater Treated and COD Reduced

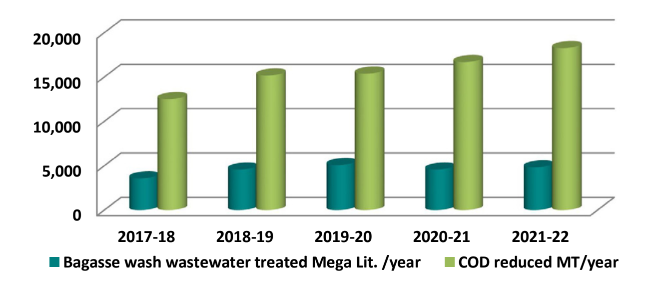


Figure:2 Biomethanation Environmental Benefit: GHG Reduction MT CO_2e/ year

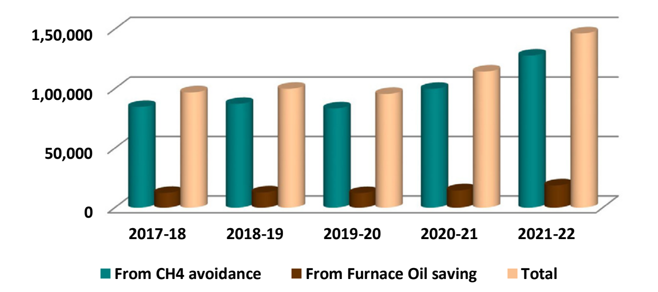
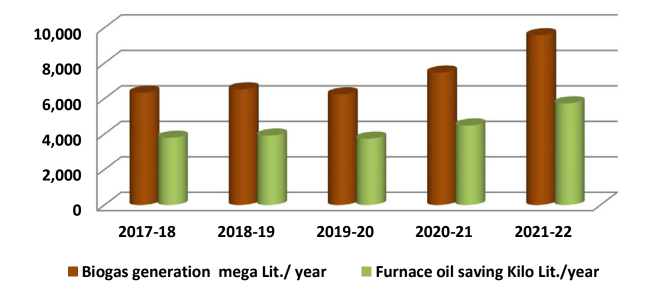


Figure:3 Biomethanation Economic Benefits: Biogas produced and F. oil saving



Utilization of biogenic solid waste

All the bio-waste such as bagasse pith from dry and wet de-pithing, chipper dust from chipper house and black liquor from pulping process generated from the plant are used as renewable energy to replace the fossil fuel and to reduce the GHG emission. The environmental and economical benefits are presented in the **Table 2.** Use of bio-waste as renewable energy fuel for the period from 2017-18 to 2021-22 resulted in total 2.91 million tonnes of CO_2e GHG emission reduction, 1.33 million tonnes of imported coal saving equal to 29,722 TJ of energy.

Utilization of inorganic solid waste

Inorganic waste materials are more detrimental to environment when compared to organic waste, because of its non-biodegradable nature and difficult to treat and dispose. An innovative solution was evolved in TNPL by combining these inorganic solid wastes and converting them into high grade cement namely Lime Sludge and Fly-ash Management (LSFM) plant under the principle of circular economy to conserve the natural resources. The concept was suggested and studied internationally by many others[12,13,14]. But to our knowledge TNPL is first mill to put the concept practically in industrial scale successfully to manufacture cement. Initially 600 tpd plant was put up in the mill premises and the project was commissioned during 2012. Further, the plant has undergone expansion from 600 tpd to 900 tpd in the year 2015. In addition to Lime sludge and fly ash other in-organic solid wastes generated from the mill such as Coating colour kitchen sludge, Lime grits, PCC grits and Bed ash is also used in the LSFM plant. The quantities of the inorganic waste generated from 2017-18 to 2021-22 are presented in the **Table 3.** Lime stone, one of the major natural resource used for making cement, is replaced with Lime sludge, Lime grits and PCC grits and associated GHG reduction is listed in the **Table 4.** In the five year period 3.09 lakh MT of lime stone is conserved and avoided 1.36 lakh MT CO_2e GHG emissions.

Circular economy for Carbon sequestration

Process flow diagram of Circular economy for carbon sequestration adopted in the Pulp & Paper, Cement and PCC/

Table 2: Environmental and economic benefits from Bio-waste used as renewable energy

Particulars	UOM	2017-18	2018-19	2019-20	2020-21	2021-22
Chipper dust	MT	6,891	11,764	11656	2,519	11,815
Thermal energy saved	GJ	47,602	81,214	80,468	17,390	81,566
Bagasse pith	MT	99,483	1,85,048	1,79,462	1,00,147	1,69,267
Thermal energy saved	GJ	5,88,559	11,03,843	11,26,304	6,28,523	10,62,320
Black liquor fired	MT	3,31,393	4,92,115	4,59,247	3,88,143	4,09,184
Thermal energy generated	GJ	37,72,798	58,08,122	53,95,542	48,57,355	50,71,021
Total thermal energy saved	GJ	44,08,958	69,93,179	66,02,314	55,03,268	62,14,906
Coal saved	MT	1,97,847	3,13,810	2,96,271	2,46,952	2,78,886
Total GHG reduction CO_2 e	MT	4,32,122	6,85,401	6,47,093	5,39,375	6,09,123

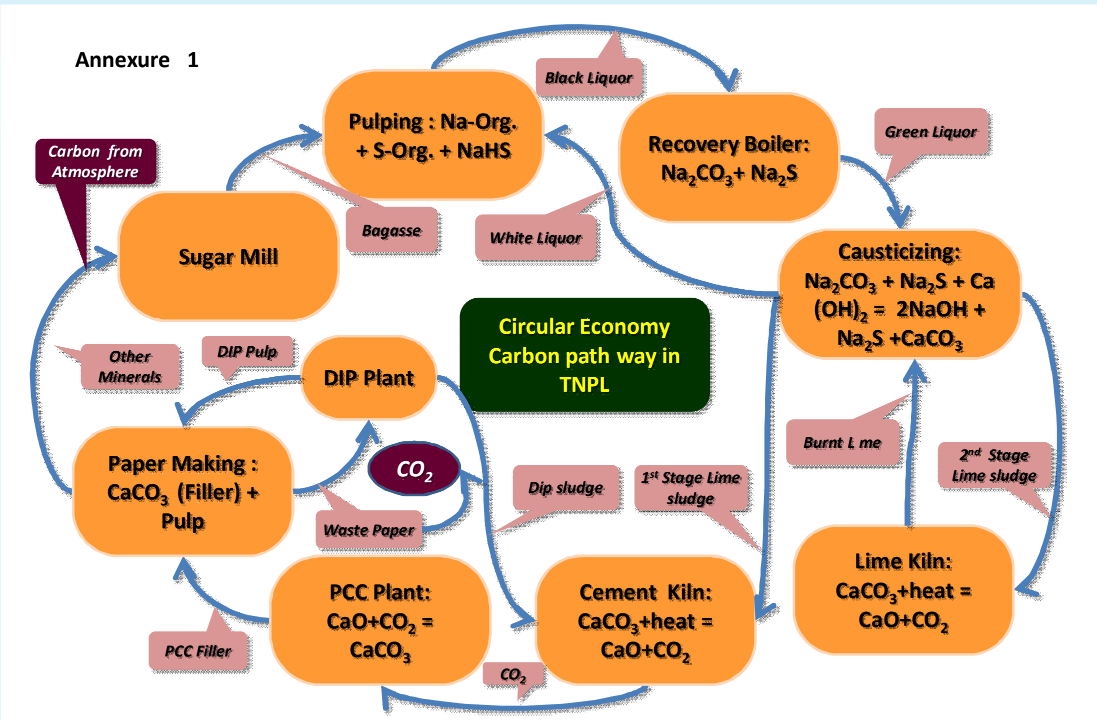
Table 3: Inorganic waste used in LSFM plant

Particulars	UOM	2017-18	2018-19	2019-20	2020-21	2021-22
CCK Sludge	MT	328	446	223	285	60
Lime sludge	MT	20,473	54,702	61,114	56,895	53,225
Lime Grits	MT	3,012	8,418	4,817	3,337	3,987
PCC Grits	MT	1,443	738	198	640	332
Bed Ash	MT	1,922	1,770	2,676	3,243	3,373
Fly ash	MT	65,901	67,008	74,886	93,462	91,618

Table 4: Resource conservation GHG emission reduction from Inorganic waste utilization

Particulars	UOM	2017-18	2018-19	2019-20	2020-21	2021-22
Lime stone conserved	MT	28,208	72,260	74,830	68,881	65,116
GHG reduction	MT	12,404	31,774	32,904	30,288	28,632

Filler complex practiced in TNPL is presented in **Annexure -1.** TNPL being agro based integrated pulp and paper mill uses nearly one million MT of sugarcane bagasse, biogenic waste from the sugar mill, for making chemical bagasse pulp saving almost equal amount of virgin fibre. Similar to any other autotrophic plants, sugar cane (Saccharum officinarum) is one of the C_4 plant and photosynthetically more efficient to produce high content of dry mass by fixing atmospheric carbon in its biomass[24,25] and bagasse is obviously forms the part sugarcane biomass[23]. After extracting the sugar cane juice, the left over bagasse is de-pithed to remove the pith which contains mostly parenchyma cell that do not have the fibre value is used as fuel. The de-pithed bagasse is stored by wet bulk storage for seasoning and washed to remove the residual organics. The washed and de-pithed bagasse is used as fibrous raw material for making chemical pulp[23].



In the Kraft pulping process white liquor containing sodium hydroxide (NaOH) and sodium sulphide (Na_2S) is used to cook the bagasse at high temperature. During the cooking process about half of the bagasse, mostly lignin and extractives, are dissolved in spent cooking liquor called weak black liquor ($NaOH+Na_2S$+Bagasse = Na-Org. + S-Org. + NaHS). The unbleached pulp fibres containing mostly cellulose and hemicellulose is bleached and used for paper making. The weak black liquor is sent to Kraft recovery system, where after evaporating to 70% solids in the evaporated, the inorganic pulping chemicals are recovered for reuse and the dissolved organics are used as a fuel to generate steam in the recovery boiler after evaporating it to about 70% solids. The combustion of concentrated black liquor gives the recovered inorganic chemicals in the form of smelt and it is dissolved to produce green liquor which contains mostly Sodium sulphite and sodium carbonate (Na_2CO_3+ Na_2S)$_2$6. It is clearly understood that the carbon present in sodium carbonate is biogenic origin[10,11,12] from the sugarcane bagasse in turn from the atmosphere fixed by sugarcane through photosynthesis 24,25. The green liquor is converted again to white liquor for reuse back in the pulping process in the causticizing process (Na_2CO_3 + Na_2S + Ca $(OH)_2$ = 2NaOH + Na_2S +$CaCO_3$). Now the biogenic carbon from the bagasse is shifted to $CaCO_3$. The precipitated $CaCO_3$ is separated from the white liquor as lime sludge and processed in the lime kiln to produce CaO and dissolved in water to produce Ca $(OH)_2$ and again used in causticizing plant. ($CaCO_3$+heat = CaO+CO_2, CaO + H_2O = $Ca(OH)_2$)26.

The Black Liquor generated at TNPL is predominantly from bagasse which posses several challenges in terms of the recovery plant performance27,28. Presence of non-process elements like silica and potassium and poor thermal & rheological properties bagasse black liquors are the major issue faced in every stage of chemical recovery cycle particularly the presence of silica. Also, non-wood black liquors have high viscosity at high solids concentrations, hard scales in the evaporator and hard deposits at various points in the recovery boiler, lower combustion efficiency, formation of colloidal gels in the re-causticizing system, lower the setting rate, reduced Lime mud dewatering, formation of glassy material in lime kilns and reduced slaking rate[14,27,28]. Silica present in bagasse fiber is classified as internal and external. The internal or inherent Silica which form part of the plant structure will vary depending on soil conditions, climate and farming practices whereas, the external Silica enter to system through raw material harvesting, handling, and storage 27,28.

Therefore to overcome the above issues, TNPL adapted two stage causticizing process in the mill. The function of the primary stage slaking is to reduce the amount of Silica present in the green liquor and to remove separately the high Silica Lime mud. The lime mud from second stage slaking with less silica is used in Limekiln for burnt lime production and then white liquor production. In first stage about 35 to 40% burnt lime is added with green liquor depending on silica content. Lime added in green liquor will first react with Silica present in raw green liquor to form Sodium Silicate. The reaction of burnt lime with Sodium Silicate is faster than the reaction with Sodium Carbonate in raw green liquor and thus Calcium Silicate would be produced first and precipitated. This retards, causticizing efficiency resulting in poor causticizing and quality of lime sludge.

Conventional reaction in causticizing :

$$Na_2CO_3 + CaO + H_2O = 2NaOH + CaCO_3$$

1st stage reaction in two stage causticizing :

$$Na_2SiO_3 + CaO + H_2O = 2NaOH + CaSiO_3$$

Hence Silica when removed in the 1st stage would improve the lime mud quality and operation efficiency in second stage. Therefore, the preferential reaction of CaO with the silica in green liquor is first allowed to take place and thereby precipitating the silica as calcium silicate complex27,28. The precipitated silica sludge is removed by settling and the supernatant with remaining lime is taken for recausticizing reaction. Thereby Silica from the recovery loop is reduced and aid to reduce Silica build-up specifically in the non-wood black liquor[27]. Though, the 1st stage is to remove the silica, a considerable quantity of $CaCO_3$ is also removed in the 1st stage. The 1st stage lime sludge contains more than 85% Calcium carbonate as $CaO_2$8 and again the carbon present in the lime mud is biogenic in the form of Na_2CO_3[10,11,12]. By practicing two stage causticizing, a portion of Silica is purged through the first stage lime mud but required environmentally friendly method to dispose first stage lime mud to avoid the solid waste disposal problem. Here in TNPL the 1st stage lime sludge containing biogenic carbonate is utilized as raw material, under the principles of circular economy, to manufacture Cement in LSFM plant and the biogenic carbonate is now shifted to Cement plant. During the Cement manufacturing, the biogenic carbon present in the carbonate is released as CO_2 in the Cement kiln similar to chemical recovery lime kiln. The quantity of 1st stage Lime sludge generated in the Kraft process and used for Cement production for the period from 2017-18 to 2021-22 is presented in the Table 3. Total quantity of Lime sludge with purity of around 88%, used in the LSFM plant for the above five year period is 2,46,409 MT.

During Cement production, the $CaCO_3$ present in the Lime sludge and containing the biogenic carbon is converted into CaO and CO_2. The CaO is used in the Cement production and CO_2 is released through Cement kiln stack. The CO_2 released from the Cement kiln stack is sequestered in PCC at PCC manufacturing process and used as filler in the paper in the papermaking process. After the use phase, the paper is recycled to manufacture deinking pulp again and back to paper making. The quantity of PCC produced and CO_2 sequestered in Cement and PCC manufacturing process is presented in the **Table 5** for the period from 2017-18 to 2021-22. For the above period total 72,997 MT PCC is produced using the biogenic CO_2 from the LSFM plant resulting in 31,456 MT of CO_2 sequestration.

Table 5: Sequestration of CO_2 from Cement stack in PCC

Year	UOM	PCC Production	CO_2 Sequestered
2017-18	MT	9,367	4,036
2018-19	MT	15,768	6,795
2019-20	MT	14,880	6,412
2020-21	MT	15,999	6,894
2021-22	MT	16,983	7,318
Total	MT	72,997	31,456

Now the concept of circular economy is fully exploited for CO_2 sequestration and to conserve the natural resources by linking following manufacturing process such as, Kraft pulping of sugarcane bagasse (pulping, chemical recovery, causticizing), Cement manufacturing, PCC manufacturing and use as filler for paper making (Annexure 1). In addition, the De-inking plant sludge which is also containing paper filler is also used in the Cement plant as alternate energy source and also to replace $CaCO_3$.

Conclusion

Linking of various manufacturing process for effective utilization of by-products and waste material based on Circular Economy concept lead to effective utilisation both organic and inorganic wastes and by-products within papermaking process as well as other manufacturing process in the down stream process such as Cement and PCC, similarly in upstream process like sugar mill. Together, this has resulted in conserving the natural resources, waste elimination & disposal issues and elimination of fossil use by renewable energy. The major benefits from 2017-18 to 2021-22 are listed below,

1. Implementation of UASB reactors resulted in environmental benefits such as 78.0 thousand MT of COD reduction, 4.83 lakh MT of CO_2e GHG Emission reduction due to CH4 avoidance, 70.4 thousand MT of CO_2e GHG Emission from fossil fuel saving, together total 5.53 lakh MT of CO_2e GHG Emission reduction. Similarly, 36,127 Mega Lit. of biogas generation resulting 21,676 kilo litters of furnace oil saving as economic benefits and natural resource saving.
2. Use of bio-waste as renewable energy fuel contributed 2.91 million tonnes of CO_2e GHG emission reduction and 1.33 million tonnes of imported coal saving equal to 29,722 TJ of energy.
3. Utilization wastes such as Lime sludge, Lime grits and PCC grits as raw material source lead to 3.09 lakh MT of lime stone conservation and 1.36 lakh MT CO_2e GHG emissions avoidance.
4. By producing 72,997 MT PCC using the biogenic CO_2 from the LSFM plant stack contributed 31,456 MT of CO_2 sequestration.

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