A Comparative Life Cycle Assessment (LCA) of Gasoline Blending with Different Oxygenates in India

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ABSTRACT

This paper includes a cradle-to-gate life cycle impact evaluation of gasoline blends in India. The potential environmental impacts of gasoline blends with three major components, i.e., methanol, ethanol, and n-butanol are assessed. The production of methanol from the natural gas reforming process, ethanol from hydrogenation with nitric acid, and n-butanol from the oxo process are considered in the current study. The results show that the gasoline blending with methanol has the lowest impact (11 categories) and is nearly constant from 5 to 15%. For gasoline with ethanol as an additive, the global warming potential, ozone depletion potential, and abiotic depletion potential rise with increasing ethanol addition. Meanwhile, increasing ethanol addition reduces the acidification potential and terrestrial ecotoxicity potential impact of gasoline blends. Similarly, gasoline with n-butanol as an additive has higher acidification potential, eutrophication potential, human toxicity potential, terrestrial ecotoxicity potential, marine aquatic ecotoxicity potential, and photochemical ozone creation potential compared to methanol and ethanol.

INTRODUCTION

India's energy security has become a critical issue with major concerns about oil and other fossil fuel depletion, environmental issues (in particular climate change), reliance on foreign sources, etc. Pollution is a major contributor to climate change. Many national and international policymakers are making reforms continuously to curtail pollution. Alcohol usage as an oxygenate fuel has the potential to reduce current emissions pollution occurring due to the properties of gasoline and its content (Yusri et al. 2017, Surisetty et al. 2011). Mainly, methanol (CH_3OH), ethanol (C_2H_5OH), n-butanol (C_4H_9OH), and dimethyl ether (C_2H_6O) were commonly used as potential fuels (Yusri et al. 2017).

However, alcohol is a clean-burning fuel that has been blended into gasoline since 1980 (Chen et al. 2018). Because of its higher octane rating and high intramolecular oxygen concentration, it can be used as a fuel in machines that have a greater compression ratio and higher thermal efficiency. However, due to the hydrophilic property of alcohol, it leads to phase separation, which is a major difficulty in alcohol blended fuels, causing operational problems and engine damage. Different blending agents have been reported by researchers to avoid methanol-gasoline phase separation (Karaosmanoglu et al. 2000). It has proven scientific records for methanol to blends of M5 to M100 (Sheehy et al. 2010,

Yuen et al. 2010) and ethanol to blends of E5, E10, and E85 (Shirvani et al. 2020) where M and E represent the percentage of methanol and ethanol in the blend and remaining is gasoline.

Multiple researchers have reported that the blending of alcohol with gasoline can minimize air pollution. Alcohol emits lesser pollutants such as nitrogen oxides (NOx), SOx, and particulate matter compared to gasoline (Canakci et al. 2013, Yanju et al. 2008).

The environmental impact of any fuel was observed in two conditions, one is during production and the second during vehicular emission or combustion. In India, similar to China, the majority of coal to methanol process is feasible due to the abundance of coal (Saraswat & Bansal 2017). Hence, the scientific community and government authorities considered oxidative additives as oxygen for the energy sector, with the goal of reducing foreign dependency in the future. Out of multiple oxygenates, methanol and ethanol prove low-cost sustainable options for gasoline blending (Shirvani et al. 2020, Saraswat & Bansal 2017).

There are mainly two sources reported for the production of methanol, i.e., natural gas, and coal. Natural gas (NG) to methanol emits much carbon dioxide per unit of energy used as gasoline. Whereas, methanol produced from coal produces double carbon dioxide, even if emission remains

the same. Butanol has better fuel properties as compared to ethanol such as higher heating value, lower vapor pressure, lower heat of vaporization, etc. That is why butanol can be used 100% as fuel in a spark-ignition (SI) engine. The only major issue is that biological and chemical pathways to butanol synthesis are both expensive (Ndaba et al. 2015, Popuri & Bata 1993). Henceforth, it is necessary to understand how to oxygenate blended gasoline and its production route may impact the transportation network, storage, and environmental impacts, among other things (Chen et al. 2018, Karaosmanoglu et al. 2000).

In India, according to Economics Times, the Government transport ministry is looking to push legislation to increase the ethanol and methanol blending in gasoline to reduce the import of fossil-based gasoline. As per Indian Government policy, the Indian Oil Corporation produces a 10% ethanol and 15% methanol (depends on availability) blend. Methanol production costs are less than half of what ethanol costs to produce, which is meant to be blended at 10%, but producers are struggling to supply sufficient ethanol to meet the mandate.

Life cycle assessment (LCA) of gasoline and diesel blending options is a well-defined track for crude oil (Mata et al. 2003). Many researchers studied the life cycle assessment of methanol, ethanol, and butanol blended with gasoline and compared environmental impacts and leak emissions only during transportation. However, depending on the type of engine, the research octane number and Reid vapor pressure after blending with gasoline have limitations. Most of the emissions comparison has been shown with different methanol blends. The research on gasoline blending, different additives, and its development technology has high growth in India. However, very few authors have studied the gasoline blending production impacts. It has been noticed that no literature is accessible on the comprehensive LCA of gasoline blending in India. The objective of the present investigation is to examine cradle-to-gate ecological implications of gasoline blending with different oxygenates, namely, (a) methanol, (b) ethanol, and (c) n-butanol in India during the year 2020-2021.

MATERIALS AND METHODS

Alcohol Production

Methanol production

Despite the growing demand for methanol as a transportation fuel component and an alternative fuel in India, conventional processes are still dominant. The most common route for the production of methanol is via syngas, although there are numerous available sources of feedstock for syngas production. The traditional route for methanol production could be summarized as:

Carbon Source + oxygen (or air) $\rightarrow$ Syngas (CO + hydrogen) $\rightarrow$ methanol

Syngas production can be prepared using several methods such as steam reforming of natural gas or naphtha (Heo et al. 2020), partial oxidation of natural gas and other hydrocarbons (Ma et al. 2019), auto thermal reforming (Hu et al. 2020), gasification technologies (Ramalingam et al. 2020), etc. India's present focus is on producing methanol via syngas from low-grade coal and solid waste (fossil or biomass) that would otherwise be burned or incinerated, as well as by-products of other sectors such as steel factories, cement plants, and refineries.

$$CH_4 + H_2O \leftrightarrow CO + 3H_2 \qquad ...(1)$$

$$CO + 2H_2 \rightarrow CH_3OH \qquad ...(2)$$
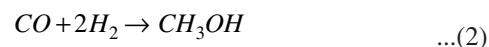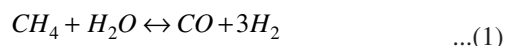
The chemical reactions carried out in the production of methanol are mentioned in Eq. 1 and Eq. 2. Whereas, Eq. (1) and Eq. (2) the overall reaction is endothermal at reactor pressure 5- 30 MPa, and temperature around 300-350°C. The detailed process unit operations used for the production of methanol from NG are shown in Fig. 1. The highest efficiency reported in the manufacturing of methanol from NG is 66% (Kajaste et al. 2018). The coal to methanol production route contributes to higher CO_2 emissions with low energy efficiency (Xiang et al. 2015).

Ethanol production (96 % concentrated)

Before 1947, ethanol was produced by indirect hydration of ethene. However, after industrialization, production was routed via direct hydration of ethene as shown in equation 3 (Weissermel & Arpe 2008, Liu et al. 2019). Ethanol (96%) production from the hydrogenation of the nitric acid process is shown in Fig. 2.

$$H_2C = CH_2 + H_2O \leftrightarrow C_2H_5OH \qquad ...(3)$$
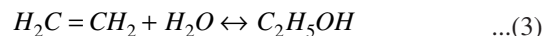
The reaction occurs in a gas phase reactor with acid catalysis. The catalyst is nitric acid. The conversion rate is quite low (only 5.6%), so unreacted ethylene and ether from side reactions are recycled. The raw product is purified by distillation and the addition of sodium hydroxide to remove aldehydes (Falano et al. 2014, Li et al. 2018).

Currently, India produces ethanol from B-heavy molasses and damaged food grains to fulfill the demand for blending. Ethanol production through molasses is a fermentation biological process, in which molasses are converted into cellular energy, ethanol, and carbon dioxide (Soam et al. 2015).

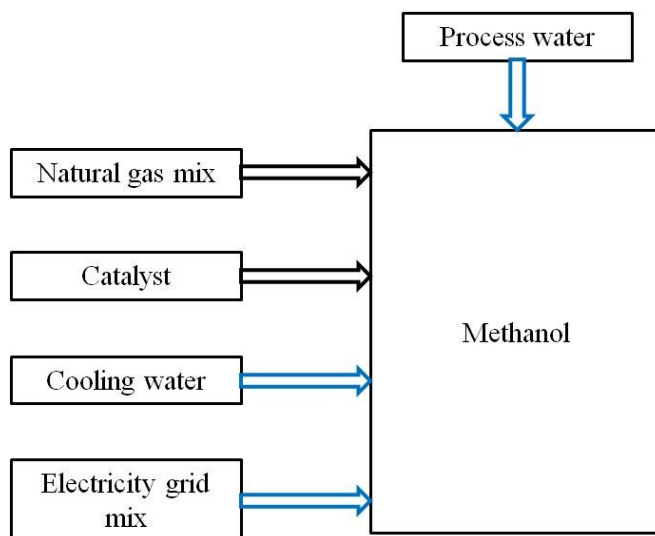Production data for molasses to ethanol mainly for en-

Fig. 1: Methanol production from natural gas.

zyme and yeast were found fluctuating in literature, so we have considered only hydration process production data for further analysis.

n-Butanol Production

Production of n-butanol can be done by two processes, i.e., petrochemical and biobased. The organic manufacture of butanol was one of the greatest commercial fermentation techniques in the early twentieth century, but it lost popularity in 1960 when researchers developed more cost-effective substrates and more efficient petrochemical processes, such as the oxo-synthesis (Patil et al. 2019). Hydroformylation

of propene is also known as an Oxo-synthesis process. Oxo-synthesis process is propene and syngas ($CO + H_2$) in the presence of a catalyst with several reaction conditions (pressure, temperature) used as a feed stream for the production of n-butanol. The detailed reaction is shown in equation 4. Uyttebroek et al. (2015) demonstrated a hydroformylation process using Rh base catalyst at low pressure, producing 95% of n-butanol and 5% 2-methyl-1-propanol (Uyttebroek et al. 2015). A detailed overview of n-butanol production from the Oxo process is shown in Fig. 3.

Production is modeled using the Oxo synthesis process or propylene hydroformylation. This low-pressure liquid-phase
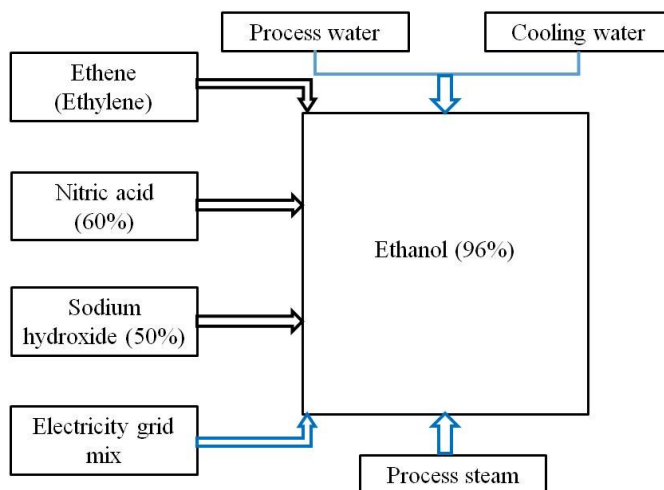

Fig. 2: Ethanol (96%) production from the hydrogenation of nitric acid.

process combines liquid-phase propylene and synthesis gas (a 1:1 mixture of hydrogen and carbon monoxide) in the presence of modified Rhodium catalysts to produce aldehydes, which are further hydrogenated to produce butanol isomers. This process is typically optimized for the production of n-butanol, with yields of up to 98% n-butanol.

$$R - CH = CH_2 + CO + H_2 \rightarrow R - CH_2 - CH_2CHO \qquad ...(4)$$
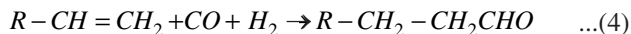
The properties of gasoline and alcohol considered in the present study are mentioned in Table 1.

Life Cycle Assessment (LCA)

LCA is a tool for systematic analysis of ecological features of products, and unit processes. Its importance has been grown in recent years as it helps to make environmental-based decisions. Detailed LCA framework for gasoline blending production study is shown in Fig. 4.

As per ISO a 14040 norm, LCA is performed in four phases:

1. Goal and scope
2. Inventory analysis
3. Impact assessment
4. Interpretation

Goal and scope

The goal and scope generally depend on the application, the geographical locations, and the time frame. The goal of the current study is to provide an outline of the cradle-to-gate LCA of different gasoline blending in an Indian context. The LCA included all raw materials and utilities. It excludes the construction, distribution, fugitive emissions, and use phases. In the present study, 1.0 t production of gasoline with different ratios (5-100 wt.%) of methanol, ethanol, and n-butanol as an additive is considered as a functional unit.

Inventory Analysis

The inventory describes the product system and its sub-processes by gathering data and calculating allocation. For inventory analysis of the gasoline blend, the mass balance is estimated on a according to per ton basis.. The primary data on the production of the additives is collected from the GaBi Indian database. For methanol and ethanol, data was directly collected from GaBi. The inventory was created for n-butanol as this was not available in the database.

Impact Assessment

In impact assessment, the information collected is analyzed for the probable environmental emissions. These impacts are expressed in equivalent units. In this paper, the CML 2001 method is used for emissions category representation.

Interpretation

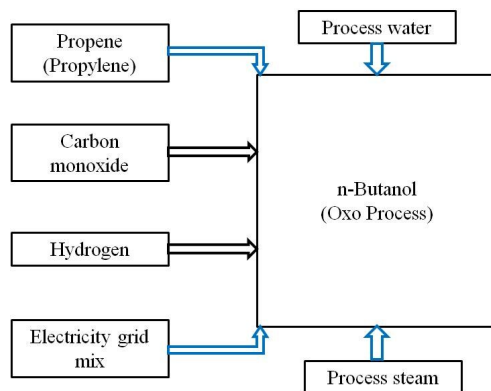In the interpretation phase, the results are analyzed including

Fig. 3: n-butanol production from oxo process.

Table 1: Properties of gasoline and alcohol.

Properties	Gross calorific value (MJ/kg)	Net calorific value (MJ/kg)	Density (kg/L)
Gasoline	47.1	43.9	0.732
Methanol	22.7	19.9	0.794
Ethanol	29.7	26.8	0.809
n-butanol	36	33.1	0.813

the relative contribution of individual process steps to the total with the above three phases. The conclusions are drawn depending on the findings of overall and component-based environmental impact.

RESULTS AND DISCUSSION

In order to convey the information included in the inventory and its significance to the environment, an impact assessment is carried out. The process flow scheme for the gasoline blend with different oxygenates is developed in GaBi. GaBi Professional software version 8.7 with the Indian Extension Database is used to analyze the environmental impacts.

Global Warming Potential (GWP)

Global warming is the increase in the warming of the troposphere due to the increase in anthropogenic greenhouse gases (GHG) in the atmosphere. The potential greenhouse effects of these gases are converted in reference to carbon dioxide (CO_2).

GWP of gasoline blending production with methanol (M), ethanol (E), and n-butanol (B) is shown in Fig. 5. From Fig. 5, it is clearly indicated that with increasing % of blending from 5% to 100%, GWP of methanol blended gasoline remains nearly constant. Whereas, GWP of ethanol and n-butanol blended gasoline gradually increases with increasing % blending. Gasoline blending with ethanol showed the highest GWP followed by n-butanol and methanol. The

reason could be that during ethanol production there are major two contributors to GWP, i.e., process steam from natural gas and ethene production. Whereas, in the case of n-butanol gasoline blend, n-butanol itself, propene (Pereira et al. 2015) and hydrogen production are major contributors to GWP as compared to gasoline. In the case of methanol gasoline blending, the primary pollution is because of natural gas production only (Lemonidou et al. 2003).

Most of the researchers reported GWP for methanol blended gasoline: 0.462 kg CO_2 eq.kg^{-1} CH_3OH and which is lower than the present study i.e. 0.832 kg CO_2 eq.kg^{-1} CH_3OH (Yadav et al. 2020). Whereas, for ethanol-blended gasoline, GWP was found to be 2.22 kg CO_2 eq.kg^{-1} g C_2H_5OH, which is quite higher as compared to cradle-to-gate in Western Europe i.e. 1.3 kg CO_2 eq.kg^{-1} (Muñoz et al. 2014). The higher values of GWP found in the present study for ethanol-blended gasoline may be due to the electricity generation from coal, transportation, and also the reporting method used (ReciPe). Individually, gasoline production has the lowest GWP (13.52 g CO_2 eq.MJ^{-1}) among methanol, ethanol, and n-butanol which is in the range of most results (10 and 15 g CO_2 eq.MJ^{-1} fuel) (Eriksson & Ahlgren 2013). Alcohol production from biomass particularly minimizes GHGs creation and result in global warming (Dalena et al. 2018).

Acidification Potential (AP)

Acidification potential is an increase in the acidity of the earth, a waterbody, or atmosphere due to human activities.
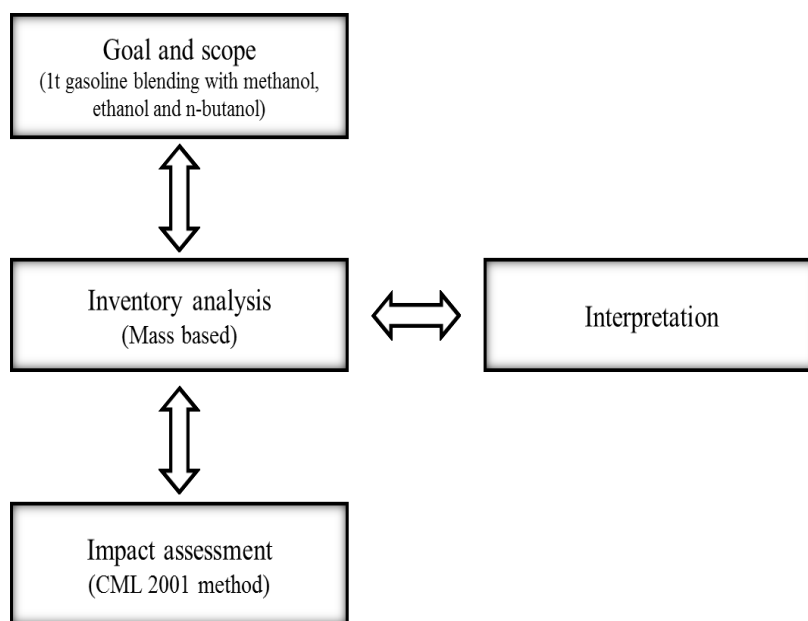

Fig. 4: LCA framework for gasoline blending production study.

The increase in the acidity of the air leads to an increase in the pH value.

The detailed AP from the production of gasoline blending with methanol, ethanol, and n-butanol is shown in Fig. 6. AP of methanol and ethanol-blended gasoline increases with increasing blending % from 5% to 15% whereas, decreases from 50% and onwards. The reason could be, independently gasoline production has the second most AP (6.3E-03 kg SO_2 eq.kg^{-1}). In the case of n-butanol blended gasoline, the authors found an almost constant AP irrespective of % blending. The higher values of AP in the case of n-butanol (7.88E-03 kg SO_2 eq.kg^{-1}) are on account of electricity and process steam generation. However, it was reported that n-butanol production from the petrochemical route had lower AP than bio-butanol due to the use of fertilizer during the agricultural stage (Pereira et al. 2015).

Eutrophication Potential (EP)

The EP is the excessive addition of nutrients such as nitrogen and phosphorus liberated into water and land. Phosphorus and nitrogen from agriculture, combustion processes, and industry effluents mainly cause eutrophication. Emissions of pollutants are converted into kg PO_4- eq. EP of methanol, ethanol, and n-butanol blended gasoline comparative results are shown in Fig. 7. As the percentage of methanol blending increases from 5% to 15%, EP showed higher values whereas, with an increase in methanol blending from 50% to 100%, EP decreases. However, for ethanol blending with gasoline, it is nearly constant. Discrete production comparison shows gasoline has the second most EP (0.418E-3 kg

Phosphate eq.kg^{-1}). EP of gasoline blended with n-butanol contributes 28 times higher followed by gasoline. The EP of alcohol blended gasoline from discrete production potential is lowest compared with methanol (0.16E-3 kg PO_4- eq.kg^{-1}) ethanol, and n-butanol. The highest EP (0.53 kg PO_4- eq.kg^{-1}) in the case of n-butanol may be contributed because of the electricity, process, steam generation, propene, and carbon monoxide. Pereira et al. (2015) reported that petrochemical n-butanol has lower EP compared to biobutanol. However, in the case of ethanol blending, it is due to process steam production only. Falano et al. (2014) also calculated EP for ethanol (PO_4- eq.) as 1.17 gm.L^{-1} which is nearly equal to 0.91 gm.L^{-1} calculated in the present study.

Ozone Depletion Potential (ODP)

This impact highlights the deterioration of the ozone layer of the stratosphere which protects living beings from ultraviolet rays. Halocarbons like chloro-fluoro-carbons or synthetic halogenated compounds prevent stratospheric ozone creation and thus limit the regeneration of the ozone layer.

ODP with respect to methanol, ethanol, and n-butanol blended gasoline is shown in Fig. 8. It was observed that with an increase in alcohol blending % in gasoline, the ODP also increases in the case of ethanol and n-butanol except for methanol. Methanol blended gasoline production does not show much effect even though it increases from 5% to 100%. ODP is maximum for the gasoline blending with ethanol followed by n-butanol mostly in case of higher blending from 50%, 85%, and 100%. Principally, ethene ($CH_2 = CH_2$), water (H_2O) and sodium hydroxide (NaOH) production are the
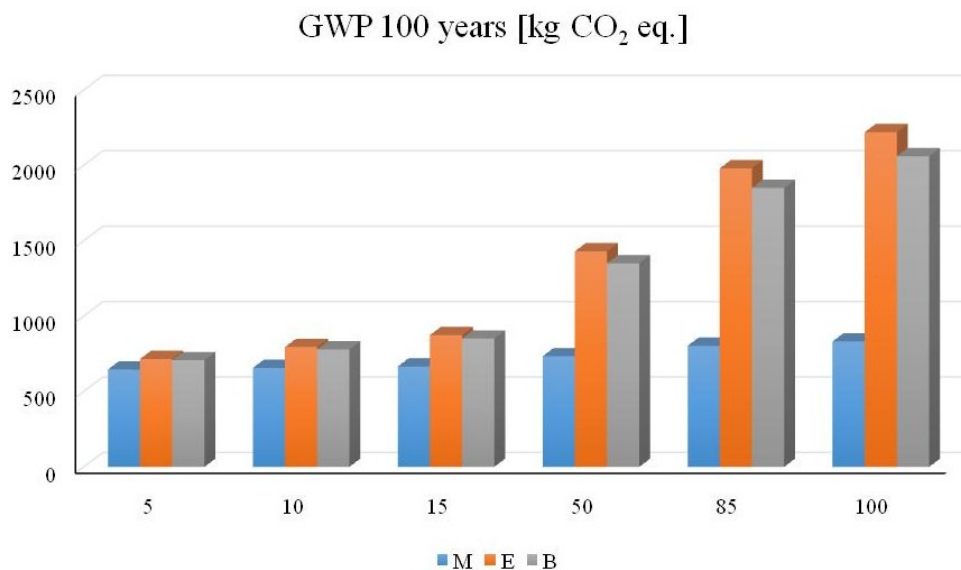

Fig. 5: GWP of gasoline blending production with methanol (M), ethanol (E) and n-butanol (B).

primary contributors that damage ozone. ODP for gasoline blending with n-butanol is mainly contributed by propene and electricity generation of n-butanol. Similar results were disclosed by Pereira et al. (2015), in the case of petrochemical route n-butanol production. The authors mentioned that the use of propylene and heat from natural resources during the production stage were major contributors. Independent production observed the lowest ODP for gasoline (8.19E-15 kg R11 eq.kg^{-1}).

Abiotic Depletion Potential (ADP) for Elements and Fossil

The impact category was subdivided into two categories, using two sets of ADPs: the ADP for elements and the ADP for fossil fuels. Fig. 9 (a) and (b) shows ADP (element) and ADP (fossils), respectively, for gasoline blending with methanol, ethanol, and n-butanol. ADP element for gasoline blending with n-butanol is mainly due to propene production. It is also seen that gasoline production has the lowest
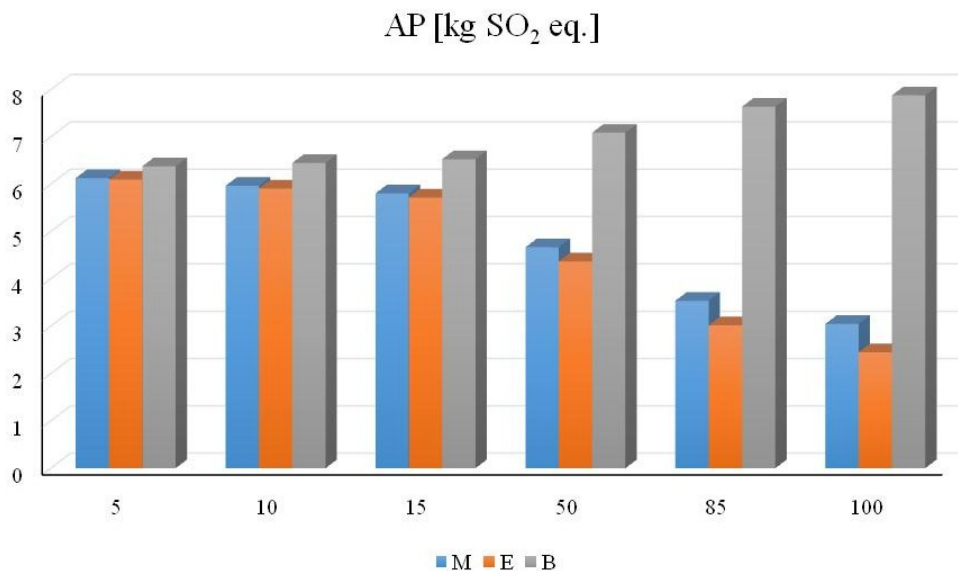
AP [kg SO$_2$ eq.]

Fig. 6: AP of gasoline blending production with methanol (M), ethanol (E), and n-butanol (B).
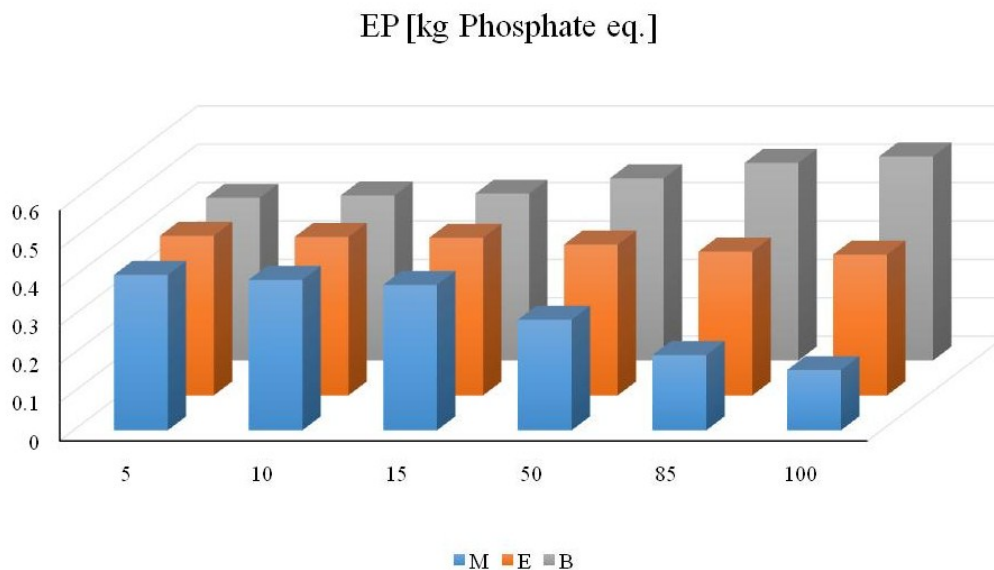
EP [kg Phosphate eq.]

Fig. 7: EP of gasoline blending production with methanol (M), ethanol (E) and n-butanol (B).

ADP elements. ADP (element) is the highest for gasoline blending with ethanol followed by methanol. As % blending of alcohol increases, ADP also increases. Initial 5% to 15% alcohol blended gasoline showed the lowest ADP and increased afterward with increasing alcohol blending %. This is mainly on account of ethane and sodium hydroxide production (Li et al. 2018).

ADP fossil impact for alcohol gasoline blending is nearly constant up to 15%. Ethene and process steam from natural gas are major contributors to ethanol ADP (fossil) impact. ADP (fossil) for gasoline blending with n-butanol is mainly contributed by propene and hydrogen production of n-butanol and the equal contribution by gasoline. It is also observed that individually, gasoline production has the third most (49.1 MJ.kg^{-1}) ADP fossils. In the present study (India), ADP (fossil) is calculated around 36.4 GJ.t^{-1} methanol which is coherent with the 33.4 GJ.t^{-1} methanol reported by Li et al. 2018.

Human Toxicity Potential (HTP)

HTP is a continuous toxicological impact on humans. The effect of percentage gasoline blending with methanol, ethanol, and n-butanol is shown in Fig. 10. For gasoline blending with n-butanol, n-butanol contributes 73% higher than gasoline. Process steam generation mainly contributes to n-butanol followed by propene and electricity generation. However, Pereira et al. (2015) reported that petrochemical n-butanol production had similar HTP compared with biobutanol.

Individually, gasoline production has the second most HTP. It is interesting to observe that the methanol addition helps to reduce the HTP impact. Khoo et al. (2016) also found (HTP) 0.4 kg DCB eq..kg^{-1} CH$_3$OH which is higher than 0.075 kg DCB eq..kg^{-1} CH$_3$OH calculated in the current study. Falano et al. (2014) calculated HTP (DCB eq.) for ethanol as 142 gm.L^{-1} and in the present study, it is around 192 gm.L^{-1}. The difference in the impact values may be due to the different sources of electricity generation, fuel composition, and also the reporting method used.

Terrrestric (TETP), Marine Aquatic Ecotoxicity Potential (MAETP), and Photochemical Ozone Creation Potential (POCP)

TETP, MAETP, and POCP are the highest for gasoline blending with n-butanol as shown in Fig. 11, 12, and 13, respectively. TETP for gasoline blending with n-butanol, gasoline, and n-butanol contribute nearly in equal proportions, i.e., 1.6E-3 and 1.8E-3 kg DCB eq.kg^{-1} respectively. Process steam and electricity generation contribute to n-butanol TETP impact. Similarly, electricity generation is also the main contributor to methanol production.

MAETP for gasoline blending with n-butanol, n-butanol contributes 5.6 times higher than gasoline. Process steam and electricity generation mainly add to this impact.

POCP impact is related to ozone creation and it is expressed in the amount of ethene equivalent emitted. Khoo et al. (2016) reported POCP 1.25E-4 kg ethene eq.kg^{-1} CH$_3$OH,
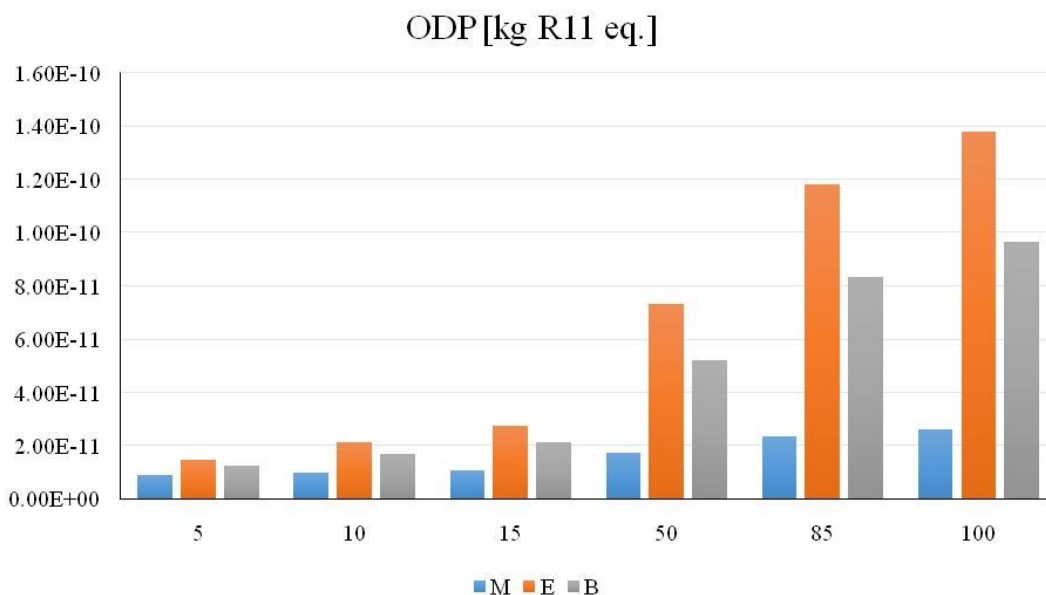

Fig. 8: ODP of gasoline blending production with methanol (M), ethanol (E), and n-butanol (B).

which is lower than 3.1E-4 kg ethene eq.kg^{-1} CH$_3$OH calculated in the current study. POCP for gasoline blending with n-butanol is mainly contributed by gasoline (6.48E-4 kg ethene eq.kg^{-1}) followed by n-butanol (5.57E-4 kg ethene eq.kg^{-1}). POCP of gasoline is added due to volatile organic compound emissions in transport and distribution (Furuholt 1995). In n-butanol, propene and hydrogen production contribute to the impacts compared to biobutanol (Pereira et al. 2015).

CONCLUSION

A detailed investigation of the environmental impact and implications of blending gasoline with (varying percents) methanol, ethanol, and n-butanol for the first time in India is presented in this paper on LCA methodology. The alcohol production through the chemical route, i.e., methanol from natural gas reforming process, ethanol from hydrogenation with nitric acid, and n-butanol from oxo process are considered.
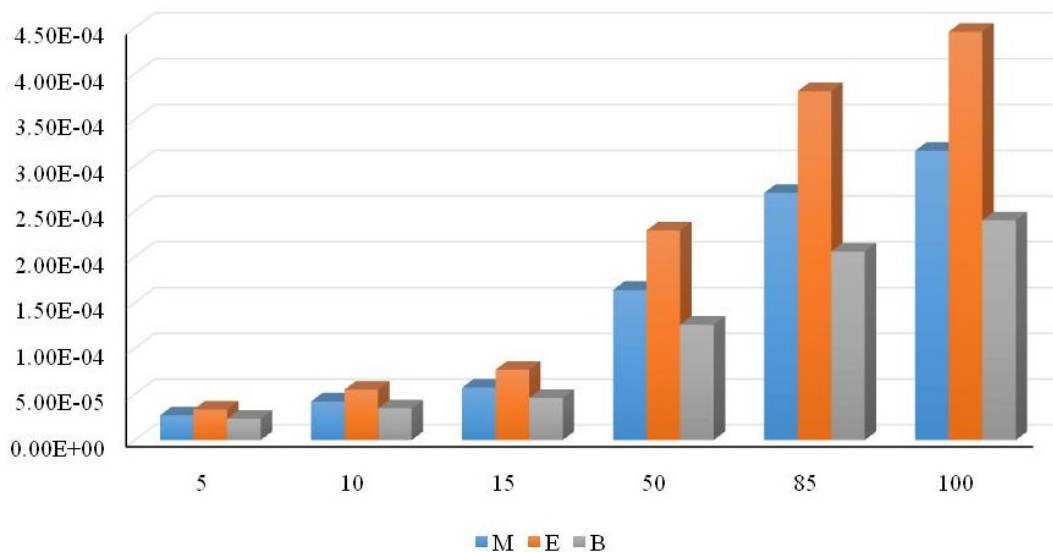

Fig. 9 (a): ADP (elements) of gasoline blending production with methanol (M), ethanol (E), and n-butanol (B).
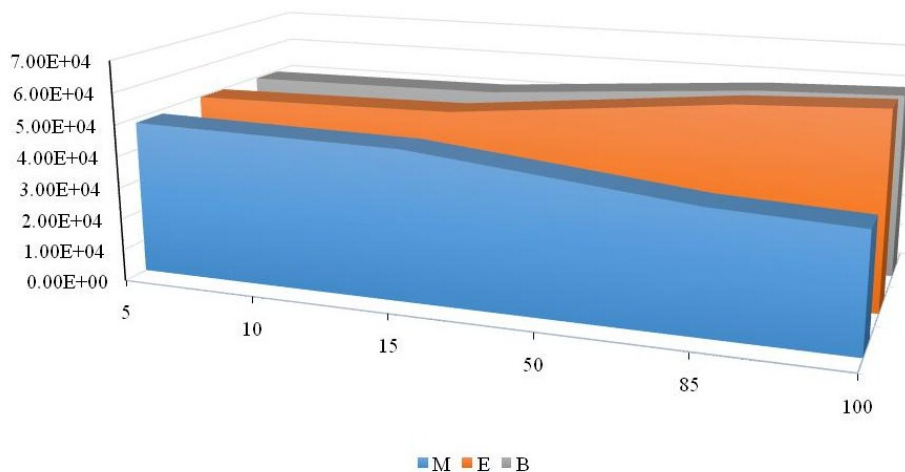

Fig. 9 (b): ADP (fossil) of gasoline blending production with methanol (M), ethanol (E), and n-butanol (B).

The results show that the gasoline blending with methanol has the lowest impact and is nearly constant from 5 to 15%. For gasoline with ethanol as an additive, the GWP, ODP, and ADP rise with increasing ethanol addition. Meanwhile, increasing ethanol addition reduces the AP and TETP impact of gasoline. Similarly, n-butanol has higher environmental impacts such as AP, EP, HTP, TETP, MAETP, and POCP compared to methanol and ethanol. Gasoline production has the second-most AP, EP, HTP, and TETP impact. Thus, out of 11, 6 of the environmental impact categories of n-butanol as an additive were consistently higher than that of methanol and ethanol.

It is not possible to state which alcohol or which route (petrochemical or biomass) is environmentally friendly overall. Blending during the production, use, and end-of-life cycle in India must be examined economically, environmentally, and sustainably. Gasoline blending appears to be more sustainable only when the additives are produced through the biological route for cleaner energy. However, the overall efficiency, energy use, and economic evaluation can play a deciding role.

DISCLAIMER

The results are based on information and data from the GaBi Indian database.
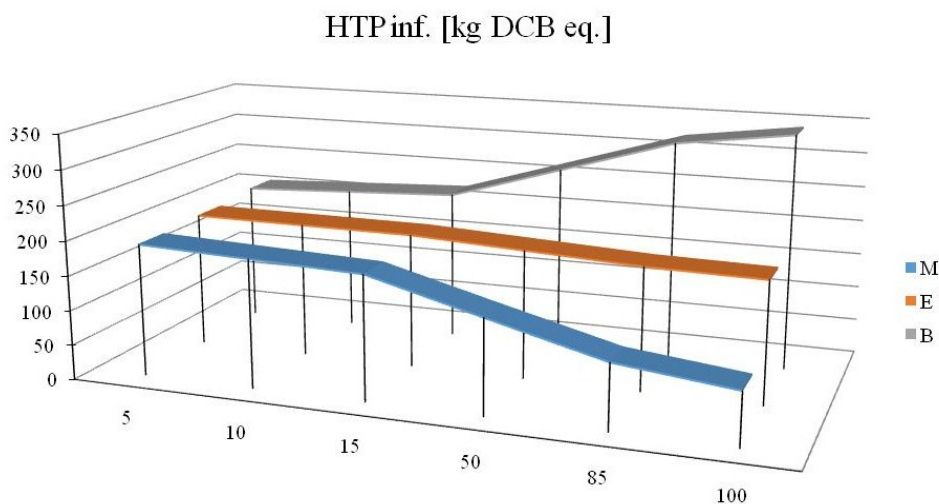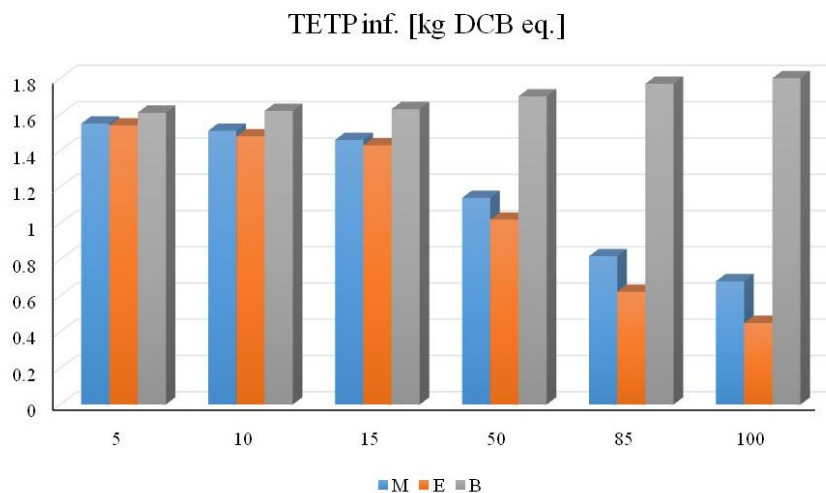
HTP inf. [kg DCB eq.]

Fig. 10: HTP of gasoline blending production with methanol (M), ethanol (E), and n-butanol (B).

TETP inf. [kg DCB eq.]

Fig. 11: TETP of gasoline blending production with methanol (M), ethanol (E), and n-butanol (B).

MAETP inf. [kg DCB eq.]
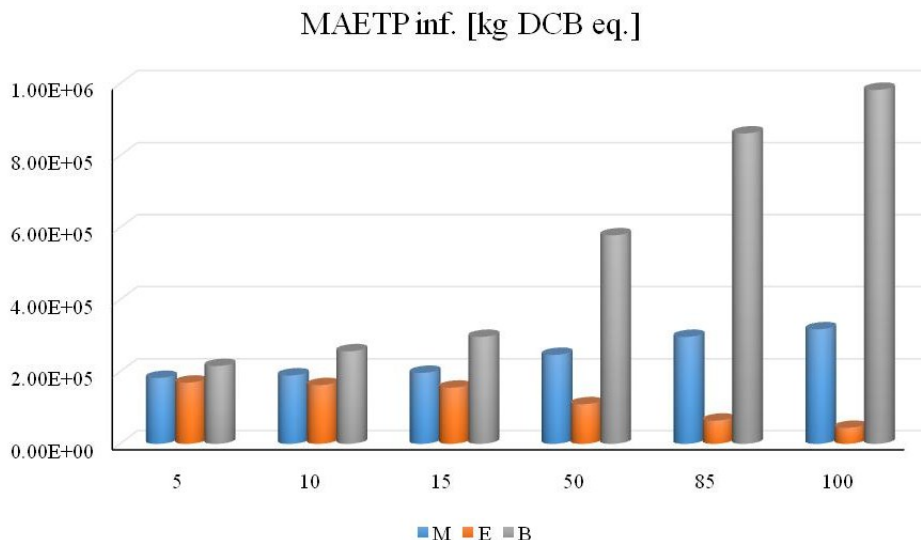
Fig. 12: MAETP of gasoline blending production with methanol (M), ethanol (E), and n-butanol (B).

POCP [kg Ethene eq.]
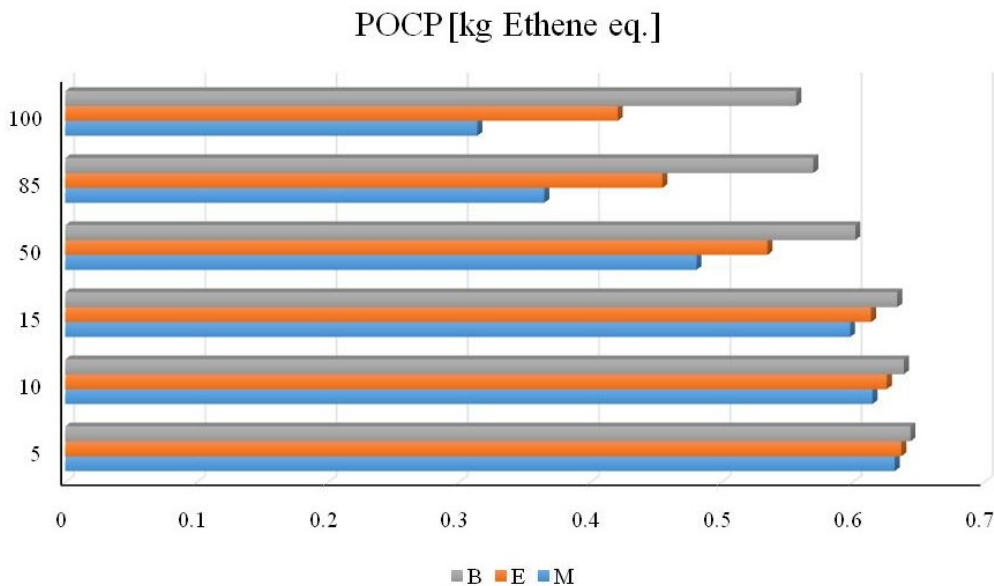
Fig. 13: POCP of gasoline blending production with methanol (M), ethanol (E), and n-butanol (B).

ABBREVIATIONS

ADP   -   Abiotic depletion potential
AP   -   Acidification potential
DCB   -   Di-chloro Benzene
EP   -   Eutrophication potential
GHG   -   Greenhouse gas
GWP   -   Global warming potential

HTP   -   Human toxicity potential
LCA   -   Life cycle assessment
MAETP   -   Marine aquatic ecotoxicity potential
NG   -   Natural gas
ODP   -   Ozone depletion potential
POCP   -   Photochemical ozone creation potential
TETP   -   Terrestrial ecotoxicity potential

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