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Introduction

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The history and development of the automotive fuels, gasoline and diesel together with their corresponding engines began more than 100 years ago and is still ongoing. During this entire period, engine and fuel technologies were mutually dependent on each other. However, with subsequent engine development, the fuel requirements changed and likewise, improvements in engine technologies were often only possible through specific progress in fuel technologies.

In the early days of both the spark-igniting Otto and the self-igniting diesel engines, the mere starting and running of the engines, i.e. the preparation of a combustible air–fuel mixture, were the main problems. The most important quality criterion for *gasoline* was that it had to evaporate easily: a finger dipped into the fuel had to dry rapidly in the open air.

Diesel fuel had to ignite, with little delay, at the end of the compression stroke of the engine, as a result of increases in heat and pressure. In contrast to gasoline volatility, the ignition quality of a fuel was much more difficult to determine and it took Rudolf Diesel and other inventors considerable time and experimental effort to establish that lamp kerosene and gas oil were the most suitable fuels.

Even after the initial breakthrough, both engines suffered from fuel-related problems, the spark-ignited engine from uncontrolled early "self-combustion" of the air–fuel mixture, which caused engine damage and severely reduced engine efficiency, and the diesel engine from the problem of exact metering of the small amounts of fuel at high pressure and high speed, as is required for fuel-injection systems.

It was about 50 years after the invention of the spark-ignited engine that a gasoline additive (tetraethyllead [TEL]) was found that reduced the uncontrolled combustion, known as engine knock, and thus allowed the construction of engines with much improved efficiency.

Small engines suitable for use in trucks became available about 30 years after the invention of the diesel engine, when high-pressure fuel-injection pumps had been developed, but it took another 10 years before Mercedes Benz installed the first diesel engine in a passenger car (PC).

Subsequently, prolonging engine life and safe operability under all climatic and driving conditions and increasing the yield of engine fuels from a given amount of crude oil by new refinery processes became important targets for fuel development until end of the twentieth century.

Since the 1980s the reduction of emissions during the production, transportation, storage and, of course, use of fuels in engines has become increasingly important. The primary objective for engine and fuel development was the reduction of air pollutants like NO_x, HC, and CO. In the last two decades concerns on global warming and global energy demand have added a new aspect, centering on fuel consumption and CO_2 emissions. The idea of fuel efficiency itself was not new; it can be traced back to the oil crises in the 1970s – a time where the search for alternative fuels also started. Some of the concepts, like methanol, have in the meantime gained only minor importance, whereas others like ethanol or biodiesel have been implemented commercially successfully as blending component with conventional fuels.

Engine fuels meanwhile have been optimized to a complex mixture of a large number of components meeting international standards and quality criteria. They guarantee maximum possible safety in the complete chain from production to their final usage, optimum engine operation over the whole engine lifetime, and, at the same time, minimum possible effects on the environment (Table 1.1).

To date a variety of engine fuels does exist, each having advantages and disadvantages. Some of them implemented in the market, some still at the beginning of their commercial life cycle and some still options on the shelf. The new edition of the Handbook of Fuels provides an overview on the available new, alternative fuel technologies as well as on developments of classical diesel and gasoline fuel to date. Moreover a comprehensive review and outlook of engine and exhaust control technology are presented, including hybrid and fuel cell concepts. Future mobility is one of the key challenges of our century and will continue to inspire innovation in automotive engine and fuel development – remaining a process

Table 1.1 Engine fuel, a highly complex mixture of various components produced to international standards, meeting engine, customer and environmental requirement.

Raw materials	Targets for fuel properties	Engine requirements
Crude oil – aromatics, olefins, paraffins, naphthenes (alcohols, ethers)	High energy content	Efficient engine performance
	Excellent combustion properties	Low fuel consumption
Additives based on organic chemistry	Low tailpipe emissions, incl. CO_2	Optimum drivability
Alternative fuels – natural gas biomass/ethanol vegetable oils or esters	Safe fuel handling	Long engine and exhaust gas aftertreatment life
	No deposits or corrosion in engine or fuel supply system	
Solar energy/electricity	Acceptable environmental properties and toxicity	Low emissions

that can be most efficiently undertaken in a joint approach in order to achieve maximum engine and fuel efficiency.

1.1 History of the Spark Ignited "Otto" Engine and of Gasoline

In 1876 Nikolaus August Otto developed a stationary, single-cylinder, four-stroke engine that ran on coal gas. This invention, which was later named after him, set a development in motion that substantially shaped the industrial age and has not yet come to a halt [1–5].

The development of motorized road transport was triggered by his dissatisfaction with the performance of the gas engines built in his factory in Deutz, near Cologne, which was founded in 1872. These heavy stationary engines used town gas as a fuel and operated at atmospheric pressure. They worked reliable and sold well in the beginning. However, all efforts to improve the incredibly low efficiency of these engines (3 horsepowers [hp] could be gained from an engine 4 m in height) failed, because a combustion process without compression was applied.

Nikolaus Otto therefore restarted tests that had been abandoned in 1862, that is, to develop a piston engine which combusts a gas–air mixture produced outside the engine after compression by the piston. This process would lead to much higher pressures in the combustion chamber and consequently to higher engine performance. This time he succeeded. In autumn of 1876 the first four-stroke engine was being operated and a year later the German "Reichspatent" 532 was issued. Overnight all previous gas engines were outdated because of the new "high-efficiency" engine.

The initial Otto four-stroke engine consisted of a single horizontal cylinder with a bore and stroke of 161 by 300 mm, resulting in a compression ratio of only 2.5. It used slide valves, reminiscent of the steam engine, and employed a flame for ignition. When operated on illuminating gas, it developed 2.2 kW, operating at 180 rpm. Its thermal efficiency of 14% seems low to the readers of today, but was two to three times that of a comparable steam engine. However, it was still monstrous, weighed 6.8 t, and was still fueled by town gas.

The technical director of the "Gas Engine Factory Deutz AG," Gottlieb Daimler, was not satisfied with the new engine. He envisaged a small high-speed four-stroke engine that could be built into a vehicle powered by a fuel, which could be transported at ambient pressure. The Deutz factory was not interested in this concept and consequently Daimler left the company and moved to Bad Cannstatt near Stuttgart, where he began his own studies, together with engineer Wilhelm Maybach. Right from the beginning Daimler intended to use a light mineral oil fraction, known as "ligroin," as the fuel, which up to that point had predominantly been used as a cleaning agent.

Within a very short period Daimler and the ingenious designer Maybach developed patentable basic technologies enabling their small four-stroke engine to run on a light gasoline fraction:

- The "uncontrolled glow tube ignition," a heated platinum tube pointing into the combustion chamber between the valves to ignite the compressed air–fuel mixture.
- The "curve groove steering" to open and close the exhaust valve.
- The "float carburetor" as a further development of the surface carburetor in which the intake air was sucked in through the "ligroin" (the light gasoline fraction).

The light gasoline, which had a density of well below 0.700 g/ml and a final boiling point of 85 °C, was sufficiently volatile to produce an ignitable air–fuel mixture in these early and simple carburetors [3]. The float carburetor had a considerable size as it was also used as a fuel reservoir, nevertheless it operated reliably. In 1885 the first vehicle from the workshop of Daimler and Maybach, the "Petroleum Reitwagen" (riding car) motorcycle (Figure 1.1), was on its "maiden trip" from Bad Cannstatt to Stuttgart-Untertürkheim. It can be considered as the prototype of the modern internal combustion engine (ICE), using a vertical cylinder and injecting gasoline through a carburetor (patented in 1887). A year later the gas engine manufacturer Carl Benz surprised the public with a three-wheeled "patented motor vehicle" (Figure 1.2) and in the same year Daimler installed his "small, lightweight, high-speed engine" in a four-wheeled motor carriage (Figure 1.3).

Meanwhile, four-cylinder in-line engines had been constructed and in 1893 Maybach developed the first spray-nozzle carburetor, which replaced the large
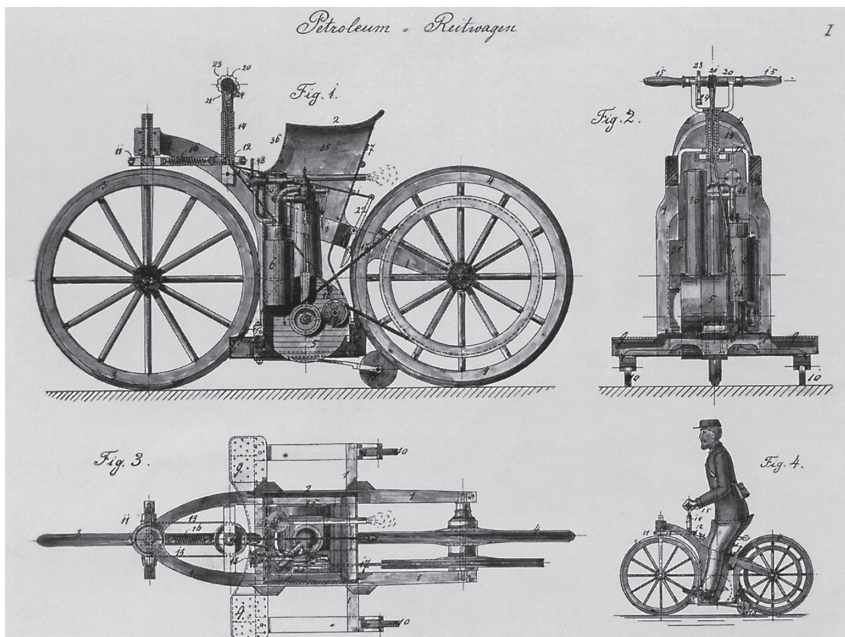

Figure 1.1 Technical data: air-cooled single-cylinder four-stroke engine, bore 58 mm, stroke 100 mm, swept volume 0.264 l, maximum output 0.5 hp., engine speed 600 rpm, weight 90 kg, vehicle speed 12 km/h. Source: Ref. [6].

Figure 1.2 The Daimler single-cylinder engine, patented in 1885.
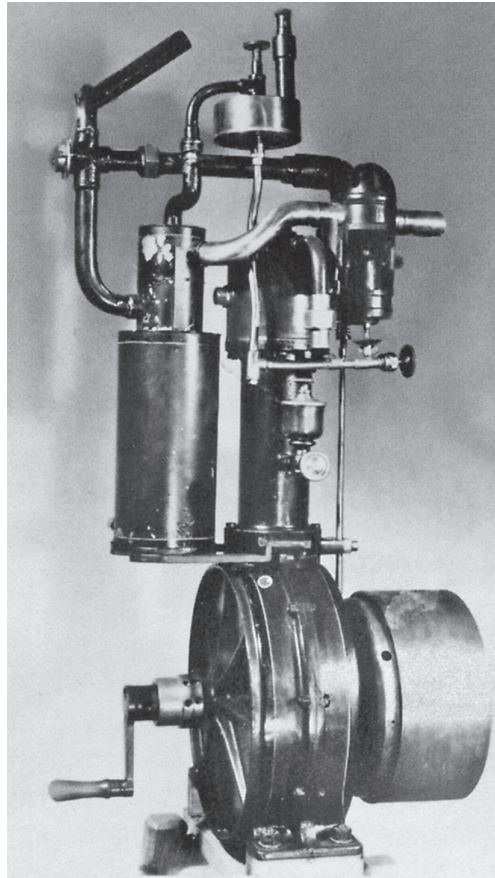
float carburetor. However, at that time it was not realized that this novel carburetor could also prepare ignitable air–fuel mixtures with gasolines of higher density and higher final boiling point and also of lower volatility. Therefore, the low-density light gasoline used up till then remained the preferred fuel for many years to come.

The invention of the spark ignition engine offered a variety of advantages for the early motorists over the steam car of the day. Among them were the almost instantaneous starting characteristics (a steam car needed up to 30 to 40 minutes in order to build up the necessary operational pressure) and its operation radius. This was three to four times longer than that of a steam powered vehicle – the driving factor being the need for water refill, because steam cars were not equipped with a condenser at the time. The major disadvantage of the combustion engine in its early days was the noise associated with each exhaust event, which could frighten passengers or people passing by. The add-on of mufflers to reduce the noise was therefore much appreciated by the public. An additional discomfort of the ICE was its need to be started by means of a hand crank. It could be an exhausting experience and engine backfire, a consequence of incorrect spark timing, lead to accidents involving broken arms or jaws.
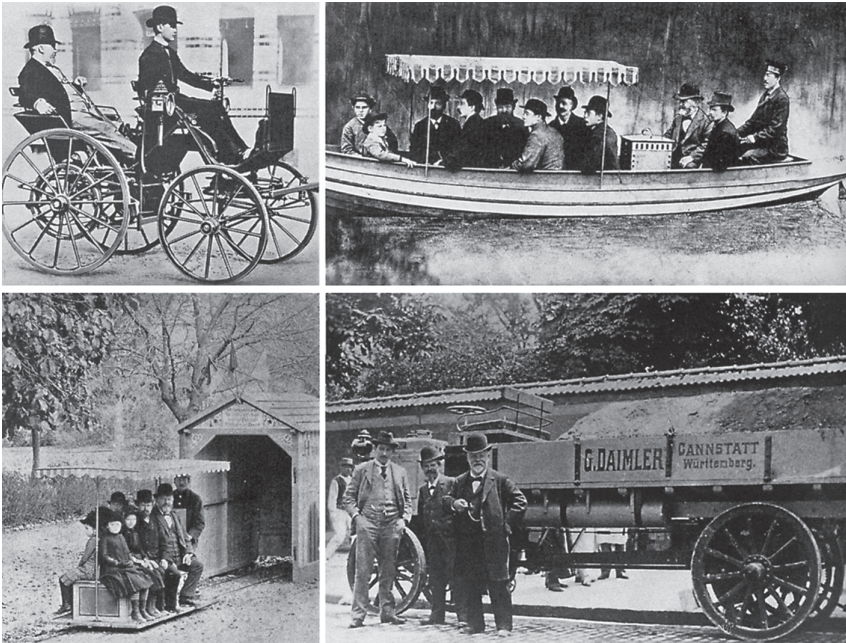
Figure 1.3 Daimlers "small, lightweight, high-speed engine" powering the first four-wheeled passenger car, "the motor carriage" from 1886; a boat (1886); a fair train (1887); a truck (1889).

The invention of the electric starter in 1911 by Charles Coleman and Charles Kettering made engine starting a much smoother and more pleasant experience for the driver.

The first recorded gasoline specification known to us originates from 1907. The US Navy specified a "high-grade refined gasoline, free from all impurities" with a Baumé density of 80°Be (about 0.673 g/ml). Motorists bought their gasoline at the pharmacy (the only place where it was available) and usually checked the density with a hydrometer.

When it was recognized that engines with spray-nozzle carburetors could also use heavier distillate gasolines, the fuel density was no longer the main specification parameter. It was discovered that the boiling characteristics of a gasoline was by far a better measure of its evaporation properties and hence new specifications were based on distillation properties.

Gasolines with a wider distillation range were also important from the supply point of view. With the rapidly increasing number of vehicles – in 1913 more than one million motor cars were registered worldwide – it became more and more difficult for the existing refineries to meet the demand for gasoline with the necessary quality (Figures 1.4–1.7). In the United States, for example, the required amount of gasoline was higher than the available quantity of distillates (straight-run products even by 1912). It was therefore imperative to produce more gasoline suitable for motor vehicles from the available crude oils, independent of their natural composition. This was even more important for
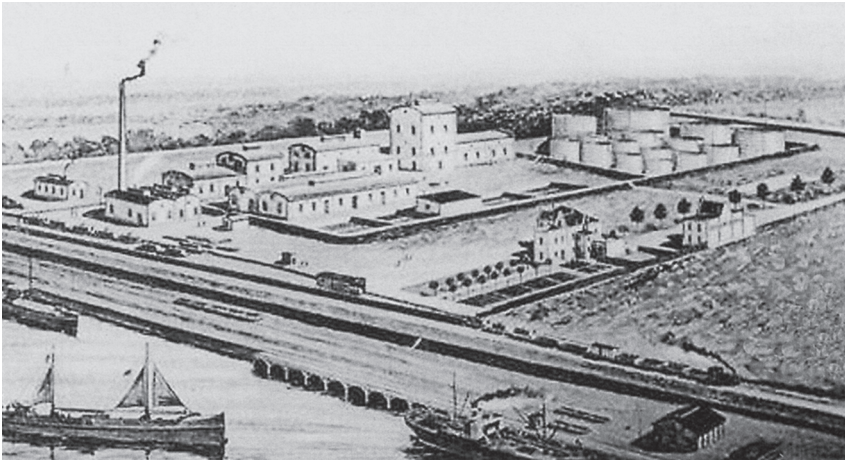
Figure 1.4 The "Gasoline Factory Rhenania" at Düsseldorf, Germany, in 1903. This was the first refinery of the Royal Dutch Petroleum Maatschappij, which merged with the Shell Transport and Trading Co. in 1907 to become the Royal Dutch/Shell group.
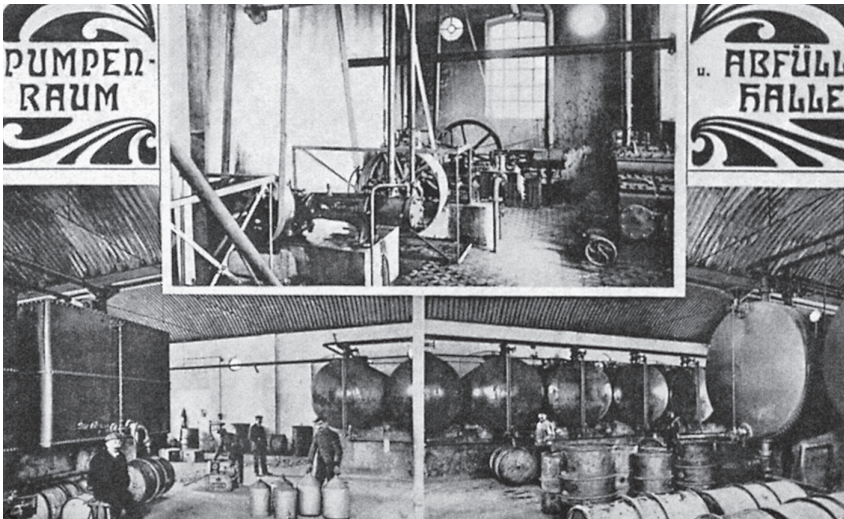
Figure 1.5 Pump room and filling of cans and drums with gasoline at the "Gasoline Factory Rhenania" in 1905.

Europe than for the United States, which at that time had nearly unlimited crude oil resources.

To solve the supply problem, a new refinery technology, thermal cracking, known in principle since 1869, was used from 1913 onwards to increase the production of gasoline from a given amount of crude oil. This technology rapidly increased in significance. However, unfortunately, it produced not only suitable light gasoline components from heavy crude oil constituents but also unstable olefins, which led to oxidation problems (gum formation

Figure 1.6 The chemical laboratory of the Rhenania gasoline works near Hamburg, Germany, founded in 1910.

and deposits). Hence oxidation inhibitors became the first implied gasoline additives.

The spray-nozzle carburetor and gasolines of suitable volatility ensured more precise mixture preparation and solved the early starting and running problems. However, the desire to increase the power output for a given engine size required higher cylinder pressures, which could be effected, for example, by increasing the compression ratio. This led to a new problem called engine knock. Although the phenomenon was not understood for some time, the results of the pinking combustion noise were obvious: overheating of pistons and valves causing severe engine damage.

Sir Harry Ricardo investigated the knock phenomenon and in 1910 he had discovered that addition of benzene prevented knock and engine damage. In his further studies he investigated the knock performance of gasolines with different compositions, their components and also that of specific hydrocarbons, alcohols, and other oxygenates. In general he found that branched and cyclic hydrocarbons, and also straight-chain hydrocarbons with olefinic double-bonds, produced less knock and would allow higher engine compression ratios than the straight-chain *n*-paraffins, which were predominantly present in distillate straight-run gasoline. These results were another reason for the development of refinery conversion technologies.

As the engine knock problem could not be solved at that time by selection of gasoline components and refinery technology, research was directed toward antiknock additives. In 1918, it was found that aniline had antiknock performance and by 1921 the General Motors researchers Midgley and Boyd, after testing a large number of substances, found that tetraethyl lead (TEL) had a very strong antiknock effect. This highly toxic material and its derivatives were used for about half a century as gasoline additives. Rising environmental concerns

Figure 1.7 1924 Shell began a "modern" service station network in Germany. A gasoline pump at the road side in Landau, in the upper Rhine area.

and the introduction of exhaust gas catalyst after-treatment systems eventually required unleaded gasolines to be introduced.

In the following decades, new refinery processes to increase the gasoline output and to raise the antiknock performance (octane quality) were developed:

- *Catalytic cracking* (1935) had, compared with thermal cracking, the advantage of produce less unstable olefins.
- *Isomerization and alkylation* (1938) increased the yield of branched paraffinic hydrocarbons with better octane quality.
- *Cyclization* (with simultaneous dehydrogenation) (reformate 1939, platformate 1949) produced aromatics with high octane quality (modern catalytic reformers can minimize the benzene concentration).

With new refineries and the new upgrading processes, it was possible to meet the worldwide increasing demand for gasoline in both quantity and quality. However, owing to political and other reasons, there were local differences. Germany,

for instance, had little access to crude oil, but a large coal production. During World War I, a gasoline consisting of 70% benzene, produced as byproduct of coal coking, and 30% ethanol from biomass was developed. In the 1920s, blends of either gasoline or benzene with alcohols were used and also gasoline–benzene mixtures.

By order of the German government, from 1930 onwards gasolines had to contain 2.5% ethanol. Under pressure from farmers, this fraction was increased to 10% in 1932. However, as a result of the increasing motorization, the necessary amounts of ethanol could not be supplied, and therefore from 1936 onwards mixtures of ethanol–methanol were also used increasingly. It is interesting that even at that time, problems of corrosion of aluminum alloys and zinc-coated iron, caused by methanol were being reported.

In spite of the significant substitution, a considerable amount of crude oil/gasoline had to be imported. During this period the first coal liquefaction plants were erected. The first unit working according to the Bergius–Pier coal hydrogenation method went on stream in 1936. The plant produced a gasoline with an octane quality (research octane number [RON]) of about 70. In comparison, the Fischer–Tropsch hydrocarbon synthesis supplied fuel with an RON of only 50 to 58. Therefore, it was still necessary to use alcohols and benzene to achieve an acceptable octane quality.

To control the gasoline quality problems in Germany at that time, in 1938 the motor industry was advised to limit the octane requirement of the engines to 74 RON. In May 1939 it was decided to add TEL to gasoline. The maximum concentration was 0.42 g/l and during World War II the minimum octane number of German gasoline was 74 RON.

At the end of the Second World War, nearly all refineries and units for the gasoline production in Germany had been destroyed. An office for mineral oil, founded shortly before the war, continued to work after the war to organize the short-term supply of oil products. After this office was closed in 1951 and international oil companies had built new refineries according to modern and international standards, the gasoline quality in Germany improved and reached that of the remainder of the western world in about 1960. The requirements of motorists and the motor industry, together with strong competition between the oil companies, led to an ever-increasing gasoline quality, which was achieved through new components and with gasoline additives. The aim was perfect engine operation under all climatic conditions, a long engine life, high power output, and high fuel economy. Milestones in the additive development were, apart from alkyl lead compounds and antioxidants, anti-icing additives to prevent carburetor icing, phosphorus-containing additives to reduce spark plug fouling brought about by highly leaded gasoline and various generations of detergents to keep carburetors, and later the complete inlet system, including the fuel injection nozzles, free from deposits.

When it was recognized that the increasing use of leaded gasoline caused worldwide lead contamination of air, water, and soil and that lead found its way into plants, animals, and humans, it was decided to reduce the maximum lead concentration in gasoline stepwise and to compensate the loss in octane quality by the addition of refinery components, however, not by benzene this time! In

Germany the maximum lead concentration was reduced to 0.4 g/l in 1972 and to 0.15 g/l in 1976. The introduction of unleaded regular grade (research octane number, RON 91) in Germany in 1985 allowed the motor industry to introduce vehicles with a catalytic system for treating exhaust gas. At that time unleaded regular grade was the most commonly used unleaded gasoline in the United States. A year later the unleaded so-called Euro premium (RON 95), a new grade, followed. The octane quality of 95, agreed between the oil and the motor industries, did not require investment in the refineries, because the composition was similar to that of leaded premium (RON 98), but of course without the lead. In 1989 the unleaded Super plus (RON 98) was introduced in small quantities. One of the components used as a lead replacement in this grade was methyl *tert*-butyl ether (MTBE). Vehicles without a catalyst could only use unleaded gasoline when their engines were equipped with sufficiently hard exhaust valve seats. The motor industry supplied the relevant information and the oil industry installed telephone hot lines to inform their customers accordingly.

The sale of leaded regular gasoline stopped in 1988 and of leaded premium in 1996. Long before that date the motor industry had equipped all new engines with hardened exhaust valve seats and the oil industry had developed additives (e.g. potassium based) to protect the exhaust valve seats of old engines so that leaded gasolines were no longer required.

Whereas pivotal engine parameters, like spark ignition timing or mixture preparation had originally been manually controlled, this had now changed to a high degree of automatization, making use of mechanics, pneumatics, and hydraulics. Today's engines with their sophisticated on-board computer and engine control functions are increasingly controlled electronically. These developments in engine technology have fostered a mutual dependency with the quality and development of gasoline to fire the engine.

From 1950 on, increasing attention was given to the further improvement of gasoline by the addition of small quantities of highly active additives.

Since 1970, measures for improving the environmental compatibility of automotive fuels have become increasingly important worldwide. The first step was the successive reduction in the lead additive content of gasolines. Later, completely unleaded gasolines were developed, which were a prerequisite for the introduction of catalytic converters.

In the mid-1980s homogeneous-charge spark-ignition gasoline engines (using carburetion, throttle-body-, or port-fuel-injection) were the dominant automotive engines. One of the key concerns of that time addressed the presence of deposits in various parts of the engine, e.g. carburetor, inlet valves, or fuel injectors, and understanding their impact on engine performance and engine-out emissions. These issues could be overcome by the invention and employment of suitable fuel performance additives. Each engine technology required new and tailored additives in order to overcome its specific deposit problematic(s). This is a continuous process and is still ongoing.

The 1970s also saw the early developments of exhaust gas recirculation (EGR), a technique deployed in order to reduce the engine-out NO_x emission. Reduction of hydrocarbon and carbon monoxide emissions could be realized by the implementation of simple computers to control engine and combustion as well
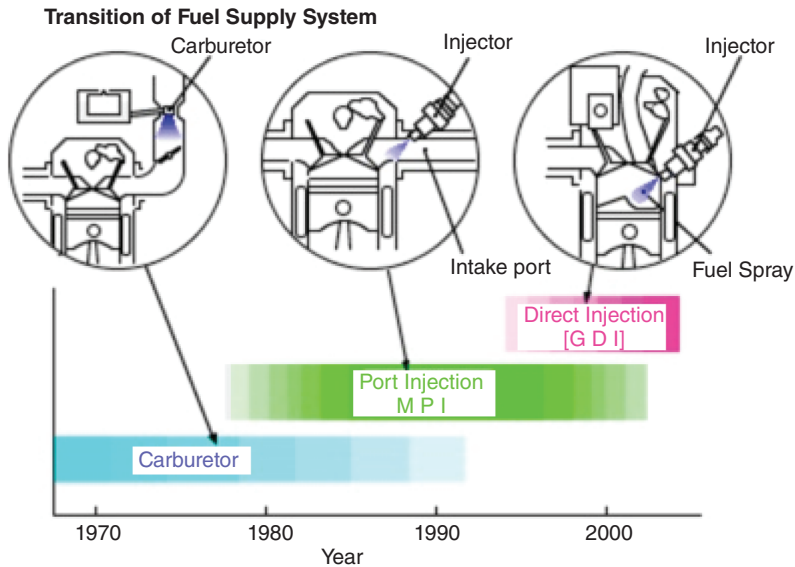
Figure 1.8 Historic overview on gasoline injection technology. Source: Ref. [8]. © 1996, Institution of Mechanical Engineers.

as the development of so-called oxygen sensors, which were able to improve the efficiency of the three-way catalyst via improved control of the air/fuel ration. A further decrease in engine emissions was effected by moving from carburetor to fuel injection technology (MPI = multi point injection). This enabled precise control of the amount of fuel delivered to the cylinder as well as further optimizations of combustion chamber design and the increase of the compression ratio. In 1980 6% of vehicles were equipped with fuel injection technology – in 1990 carburetors had seized to exist [4] (see Figure 1.8).

In the mid-1990s the first commercial vehicles were equipped with gasoline direct injection (GDI) technology. So far the fuel had been "indirectly" injected, i.e. on the back of the inlet valve and sucked in together with the air to enable perfect mixing of the two. Now the fuel was directly injected into the combustion chamber, after having been highly pressurized and being delivered by means of a common rail into the individual cylinders. Sophisticated combustion chamber design was needed in order to ensure an air-fuel flow through the combustion chamber delivering an ignitable mixture at the spark. Initially this technology did not allow the more efficient lean-burn mode and the engines were run stoichiometrically. Nowadays, it is possible to operate an engine at low loads with a stratified fuel charge (i.e. stoichiometric at the spark in order to enable ignition) and leaner around it. Direct injection technology enabled further improvements in engine efficiency based on the ability to precisely adjust fuel amount and injection timing to the actual load condition and by reducing or avoiding throttle and pumping losses experienced with MPI.

The worldwide rising demand for oil products as well as environmental challenges, such as global warming, have led to a strong push for the development

of new and more efficient automotive technologies as well as automotive fuels. In addition to direct injection technology, recent and ongoing trends are turbocharging in combination with engine downsizing as well as new valve timing and lift technology concepts. A recent study carried out by Environmental Protection Agency (EPA) [7] provides an analysis of turbocharged, downsized engine technology developments and future trends.

Variable valve timing (VVT) and variable valve lift (VVL) are techniques that enable a more precise control over the air entering the engine. VVT allows the timing of valve opening and closing to be varied. VVL systems also allow the length and/or height of the valve opening to be adjusted. At low engine loads a reduction in pumping losses is possible, as the throttle can open further. At high loads they increase airflow for more power, enabling engine downsizing for additional efficiency improvements. VVT can also be used to control levels of residual exhaust gases in the cylinder, providing additional combustion improvements and pumping loss reductions.

VVL/VVT also offers the ability to use of more efficient combustion cycles, such as the *Atkinson cycle.* An Atkinson-cycle engine trades off decreased power for increased efficiency. Essentially, the intake valve remains open for a longer time during the intake stroke and closes during the normal compression stroke. This allows more work to be extracted per volume of fuel as compared with a typical Otto-cycle engine.

Stop–start concepts seek to reduce fuel consumption in urban driving by shutting the engine off when the vehicle comes to a stop in traffic and restarting when acceleration is required, thereby reducing engine idling time.

Table 1.2 Distribution of MY2015 gasoline turbochargers [5].

Category	Turbo share (%)
Car	
Four-cylinder	60.8
Six-cylinder	4.5
Eight-cylinder	2.5
Other car	1.8
Truck	
Four-cylinder	17.9
Six-cylinder	11.9
Eight-cylinder	0.5
Other truck	0.1

In addition to the concepts for engine efficiency enhancement for internal combustion engines mentioned earlier, electric and hybrid mobility starts to play an important role. Hybrid concepts are discussed in more detail in Chapter 15.
Source: Ref. [5].

Cylinder deactivation allows the engine to significantly reduce pumping losses, as well as some reduction in heat transfer losses at low load conditions. The concept works by reducing the number of running active cylinders and increasing the load on the cylinders "working." This results in the reduction of the active displacement, thus increasing manifold pressure and reducing pumping losses through a lower pressure differential across the engine. It also reduces the heat transfer to the cylinder walls and head also helping engine efficiency to improve.

Table 1.2 [5] summarizes the prevalence of turbocharging in the vehicle population up to 2015. Further information on GDI, CVT, electric vehicles (EVs) plug-in hybrids, and fuel cell vehicles can be obtained from [7].

1.2 History of the Diesel Engine and of Diesel Fuel

For a long time the production and the processing of crude oil was dominated by the demand for gasoline, because the spark-ignited Otto engine was, apart from a few exceptions, the only engine available for mass motorization. When Rudolf Diesel experimented with his self-igniting engine at the Maschinenfabrik Augsburg Nürnberg (MAN) factory in Augsburg, Germany, nothing was known about the fuel requirements of his new engine and it seemed only natural that he tried gasoline as a fuel.

During the development of his engine, Diesel's aim was an engine operating as closely as possible to the Carnot cycle, with high gas temperatures after compression, which gives the maximum theoretical efficiency. This concept required higher compression ratios than used in the spark-ignited Otto engine to enable self-ignition of the fuel towards the end of the compression stroke, which is followed by controlled flame propagation during the expansion stroke, provided a suitable fuel is used.

The first ignition of Diesel's engine using gasoline on 10 August 1893 was so vehement that his device to measure the cylinder pressure was destroyed. This proved two things: firstly, the principle of self-ignition worked, and, secondly, gasoline was unsuitable as a fuel for a Diesel combustion process. Obviously the self-ignition properties of gasoline were inadequate, causing ignition delay, which was followed by unacceptably steep pressure rises.

Apart from finding a suitable fuel, there was also the problem of precise high-pressure fuel injection into the combustion chamber. Adequate pumps and nozzles were not available. Rudolf Diesel solved the problem by blowing the liquid fuel together with compressed air into the combustion chamber. This system worked reasonably well, but the air compressor reduced the engine efficiency and made the engine less suitable for use in vehicles.

Employing a mixture of gasoline and lamp oil and the air pressure injection system, Diesel finally succeeded and he got his engine (Figure 1.9) to run.

After 1897, experiments with other fuels started, which had little success at first. Later it was found that so-called middle distillates from crude oil, with a boiling range between 250 and 350 °C, were ideal fuels for the Diesel combustion

process. These middle distillates were blended in low concentrations into town gas, which explains the name "gas oil" for this mineral oil fraction.

However, it took more than 25 years until the diesel engine found its way into vehicles, after the development of reliable high-pressure liquid-fuel injection systems. In 1923–1924 MAN and Daimler–Benz introduced commercial vehicles with diesel engines and the first PC, a Mercedes 260 D followed in 1936, powered by a 2.6-L 4 cylinder pre-chamber engine producing 45 hp. at 3000 rpm.

By 1905, the production of turbochargers for diesel engines started to boost power output. A turbocharger pre-compresses the air to create more air mass for the combustion and subsequently delivers more power. Turbo intercoolers were also developed at that time. They cool down the compressed air from the turbocharger, like a heat exchanger, and increase density of the air prior to combustion.

The German engineer Prosper L'Orange patented a radically new design, including a pre-chamber (German Patent DRP No. 230517). The patent described the design of a pre-combustion chamber in the diesel engine, which allowed for partial combustion prior to the introduction to the cylinder and additional turbulence for a more complete mixing of the fuel and air. L'Orange obtained his patent on 14 March 1909. According to Daimler AG, this was an important milestone in diesel technology advancement because it "made possible the development of the diesel engine from being a stationary source of power to one with mobile applications." L'Orange provided additional, groundbreaking innovations, such as the funnel prechamber (1919), the pintle-type injection nozzle (1919), and the variable injection pump (1921).

The desire to improve the diesel engine's power-to-weight ratio produced several novel cylinder arrangements to extract more power from a given capacity. The Napier Deltic engine, with three cylinders arranged in a triangular formation, each containing two opposed-action pistons, the whole engine having three crankshafts, is one example. The Commer engine (designed for road vehicles) is another one. It had three horizontal in-line cylinders, each with two opposed action pistons and the engine had two crankshafts. While both these designs succeeded in producing greater power for a given capacity, they were complex and expensive to produce and operate, and when turbocharger technology improved in the 1960s, this was found to be a much more reliable and simple way achieving more power.

The most important fuel property, which was required to guarantee safe starting and smooth running of the engines without any harsh noises and steep pressure rises, was adequate ignition quality to ensure a short ignition delay after fuel injection into the hot and compressed air in the combustion chamber. Ideal fuels are those that are unsuitable for the spark-ignited Otto engine, i.e. straight-chain paraffins with a boiling range above that of gasoline. This widened the availability of fuels from crude oil without the use of conversion processes. Furthermore, for a long time the typical diesel fuel was a straight-run distillate, being a comparatively cheap fuel. The low cost and abundant supply of the diesel fuel and the high efficiency of the diesel engine were three important factors in the use of diesel engines in commercial road transport vehicles. The disadvantages of the diesel engines at that time, i.e. noise, smoke, a limited engine speed range, and

less power output for a given engine size and weight, were tolerated in heavy-duty machinery and vehicles.

Another important fuel property was cleanliness: the fuel had to be free from foreign matter particles and to be stable at high temperatures to protect the injection system against wear and deposits, which caused rough running, smoke, and power loss. Another important quality criterion was cold flow filterability and pumpability. Unfortunately the long-chain paraffins, with their excellent ignition properties, form wax crystals at low temperatures, which limit their use in winter periods.

During the Second World War, the development of diesel powered vehicles slowed down. It was too difficult to supply the troops with two different fuels. Because of the crude oil shortage in Germany, diesel fuel also had to be produced from alternative sources, namely, coal. Fortunately the Fischer–Tropsch process supplied a component with an even higher ignition quality than straight-run diesel fuel from crude oil. This component was blended with low quality tar oil to obtain a diesel fuel of reasonable quality. The steep increase of motorization in Germany after the war changed the situation completely. The consumption of diesel fuel rose from about $700\,000\,t$ per annum in 1942 to $4.8 \times 10^6\,t$ per annum in 1960. The demand for diesel fuel and light heating oil (which has about the same boiling range) rose further and could not any longer be met by straight-run products. The refineries had to invest in cracking units for the production of middle distillates. While the crack components improved the cold-flow performance, it became more difficult to obtain an adequate ignition performance of the final diesel fuel blend.

Over the years because of the improvement described earlier, diesel engines finally reached such a developmental stage that they could compete with Otto engines and increasingly found their way into passenger cars (PCs). This meant a more widespread use of diesel fuel, and although the engines behaved in a more "user-friendly fashion," for some time, diesel fuel kept its smelly and "sticky" appearance. This poor image was made even worse by the way diesel fuel was offered at the service stations, as it was intended for heavy-duty customers. The pumps were often hidden in a remote corner, the surroundings were scruffy and the supply nozzle sometimes too large for a PC. In some areas of the world, e.g. in the United States, the diesel fuel that was available was unsuitable for the new high-speed PC engines. Also, inadequate housekeeping at service stations caused problems. Water at the bottom of the tanks and attack of fungi and dirt in the fuel caused problems in the early diesel-powered PCs.

Technical requirements, environmental considerations, and customers needs demanded distinct improvements. This was partly achieved by PC friendly fuel pumps at the service station forecourts. However, the product also had to be improved through refinery technology and also by the use of new additives.

Fuel injection systems have changed significantly over time. Older engines made use of a mechanical fuel pump and valve assembly which is driven by the engine crankshaft, usually from the timing belt or chain. These engines used simple injectors that were basically very precise spring-loaded valves, which opened and closed at a specific fuel pressure. The slightly more modern method utilized a separate fuel pump, which supplied fuel constantly at high pressure

to each injector. Each injector then has a solenoid that was operated by an electronic control unit, which enabled more accurate control of injector opening times, resulting in better engine performance and fuel economy. This design was also mechanically simpler than the combined pump and valve design, making it generally more reliable, and less noisy, than its mechanical counterpart.

The increase of injection pressure from 600 bars in the 1980s (in-line injection pumps) to more than 2500 bar in latest common rail technology was dramatic.

Modern diesel engines make use of one of the following direct injection methods (Figure 1.10):

1. *Distributor pump direct injection:* The first direct injection diesel engines used a rotary pump very similar to indirect injection diesels; however, the injectors were mounted in the top of the combustion chamber rather than in a separate pre-combustion chamber. The disadvantage of this approach was the harsh noise and the particulate (smoke) emissions.
2. *Common rail direct injection:* In older diesel engines, a distributor-type injection pump, regulated by the engine, supplied fuel to injectors, which were simple nozzles through which the diesel was sprayed into the engine's combustion chamber. In common rail systems, the distributor injection pump was replaced by a high pressure pump which stores a reservoir of fuel at high pressure in a "common rail." Modern common rail diesel systems can generate pressure in excess of 2000 bar (29 000 PSI). The fuel is delivered to the cylinders via computer-controlled injector valves, each of which contains a precision-machined nozzle and a plunger driven by a solenoid, or by piezo-electric actuators. Modern injectors are typically equipped with five to seven extremely small and fully machined holes to ensure optimum atomization of the diesel fuel in the cylinders and, as a result, efficient air fuel mixing and combustion.
3. *Unit direct injection:* Unit direct injection also injects fuel directly into the cylinder of the engine. However, in this system the injector and the pump are combined into one unit positioned over each cylinder. Each cylinder thus has its own pump, feeding its own injector, which prevents pressure fluctuations and allows more consistent injection to be achieved. This type of injection system, also developed by Bosch, was used by Volkswagen AG in PCs (where it is called a "Pumpe-Düse System," literally a "pump-nozzle system") and by Mercedes Benz (PLD) and most major diesel engine manufacturers in large commercial engines (CAT, Cummins, Detroit Diesel). With recent advancements, the pump pressure has been raised to 2050 bar (205 MPa), allowing injection parameters similar to common rail systems.

These breakthroughs in technologies made direct fuel injection sufficiently smooth and offered additional reductions of emissions so that the highly efficient direct-injection diesel engines were suitable for PCs, and they have since replaced the previously used swirl and pre-chamber engines.

Compared with the old days, diesel fuel is now a complex blend of hydrocarbon streams with a wider range of additives than in gasoline. The quality of diesel fuel is regulated in dedicated fuel standards.
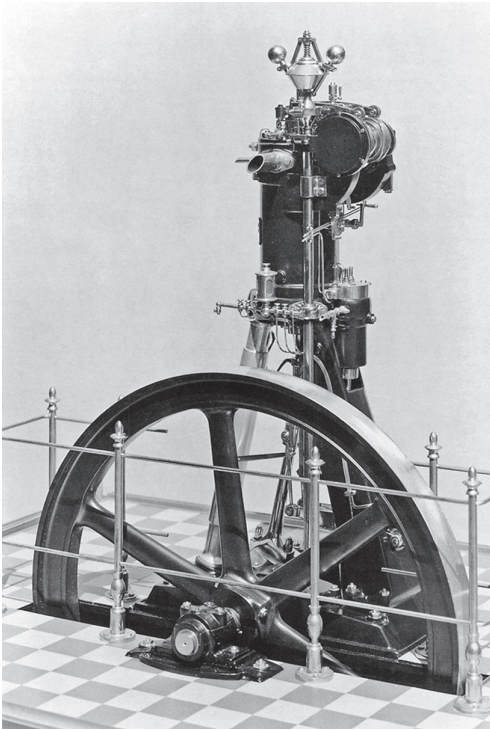
Figure 1.9 The first Diesel engine from MAN, a single cylinder of 19.6 l capacity, weighing about 5 t. It operated on a mixture of gasoline and lamp oil with a hitherto unknown high efficiency of 26.2%, producing 17.8 hp. at less than 200 rpm.
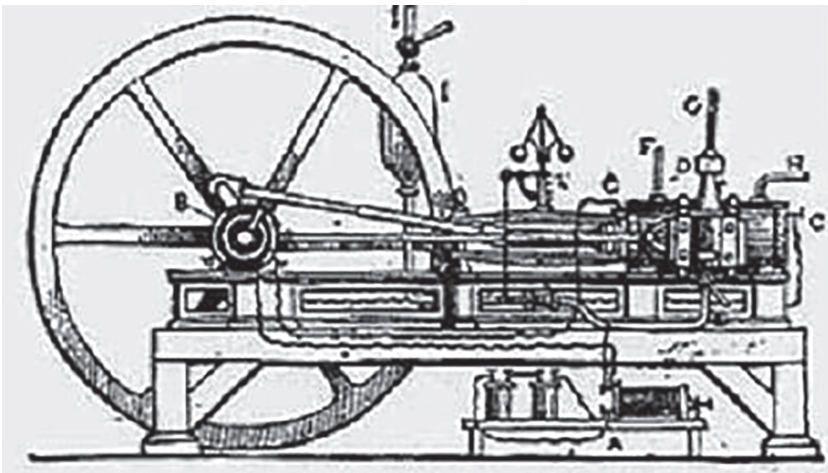
Figure 1.10 Lenoir engine. Source: Ref. [33].

New refinery technologies, synthetic fuels or components, new additives and fuel from biomass have helped to solve critical development tasks. Close cooperation between the oil and motor industries either bilaterally or industry-wide will ensure the necessary progress towards more efficient and environmentally acceptable diesel engines of the future.

1.3 History of Alternative Fuels

This chapter provides an overview of the history of the main alternative fuels discussed in this book. It aims to describe their histories from a global perspective, with a focus on Europe and the United States in most cases. Some of the alternative fuels and processes to produce them have been known to mankind since the Neolithic age – most of them are indeed much younger. Interest in the different chemicals has initially not been an intended use as a transportation fuel, but rather for domestic applications in heating, lighting, or cooking. Interestingly enough key engine engineers and inventors like Nikolaus Otto, Rudolf Diesel, and Henry Ford expressed a very early interest in some of the options generated from biomass and tested them in their first prototype cars. These inventors were not motivated by the potential sustainability of the fuels (the word "sustainable" probably did not exist at the time), but they saw a marketing advantage to consumers in a rural environment. So right from the beginning of the motoring age, alternative fuels had been on the radar as potential power sources. The success of the "in abundance" available, cheap petroleum-based products stopped their development very soon. Only in times of fuel shortages, e.g. during World War I and II and the oil crisis of the 1970s, did these fuels become attractive and researched alternatives again. It has to be remembered that biofuels accounted for c. 7% of the total fuel consumption in some countries in the 1930s. Their real success story nonetheless set in much later, with the awareness of the global climate challenge and the need for sustainable energy.

This chapter describes the early developments of key alternative fuels from their early days up into the late twentieth century.

1.3.1 Ethanol

Ethanol is one of the chemicals that were known to mankind already during the Neolithic age. Dried ethanol residue has been found on 9000 year old pottery in China [9]. Fermentation is the oldest way for the manufacture of ethanol and it has remained the traditional way of making alcoholic beverages. It is also the principal process by which the majority of ethanol fuels are produced nowadays.

Beer and wine typically do not develop alcohol content over 15 vol% alcohol, since a higher concentration makes it impossible for most yeasts to grow [10]. With the invention of the distillation process, it was possible to obtain higher ethanol concentration. Fermented solutions have been distilled since ancient times by Greeks and Arabs. Greek alchemists working in Alexandria during the first century CE carried out distillation and the medieval Arabs learned from the Alexandrians [11].

Ethanol's first use as transportation fuel was to power an engine in 1826, developed by the American Samuel Morey, which was designed to run on a mix of ethanol and turpentine. Nikolaus Otto, the inventor of the modern four-cycle ICE, used ethanol to fuel one of his early engines, dating back to 1876. Henry Ford's first vehicle, the quadricycle, ran on pure ethanol (1896) and the Ford

Model T was designed to run on (corn) alcohol, gasoline, or a combination of both fuels, an approach later to be called "flex-fuel," but in 1908 this term was not yet invented.

Ethanol as a conventional fuel substitute was of interest in varies countries around the globe dating back as far as to the early twentieth century. Differing in the potential feedstock for the ethanol production, these efforts were united in the goal to create independence from oil (imports), to strengthen the struggling agriculture and to mitigate fuel shortages as a consequence of the increasing car population – and sadly enough – the two world wars.

Europe. With few domestic oil reserves, Germany and France wanted to develop alternatives to petroleum derived products, making the countries more fuel independent. The usage of local crops like potatoes, grain, or beet was an attractive option to be developed. The foundations for ethanol usage as alternative fuel were laid in the 1890s, when alcohol-fueled engines started to be used in farm machinery in Europe. Research at the Experimental Mechanical Laboratory of Paris and at the Deutsche Landwirtschaftliche Gesellschaft in Berlin helped to pave the way for expanded use of alcohol fuel.

Germany created the world's first large-scale biofuels industry in the decades before World War I as a way to promote rural development and national self-sufficiency. Kaiser Wilhelm II is quoted in a newspaper interview to have said to "be enraged at the Oil Trust of his country" and offered prizes to his subjects and cash assistance "… to adapt [alcohol] to use in the industries" [12]. The German program involved tariffs on imported oil, farm distillery construction, promotion of ethanol fueled appliances, and research into ethanol fueled trucks, automobiles, and locomotives. In 1903, the Reichstag approved a tariff on oil to expand the farm ethanol production infrastructure. Potato alcohol was seen as the "final solution of the oil problem and the means by which the grasp of the great (Standard oil) monopoly will be broken." A network of small farm "Materialbrennereien" distilleries was put in place. Estimates of its size vary. By one 1906 account, some 72 000 distilleries operated, of which 57 000 were small farm "Materialbrennereien" stills producing a total of 27 million gallons (about 102×10^6 l) [12]. By 1938, Germany was producing about 267 million liters of ethanol, about two thirds from potatoes and the rest from grain, wood sulfite liquors, and beets. Some 89 million liters of methanol were produced from coal. All in all 54% of the pre-World-War II German fuel production was derived from non-petroleum sources, of which 8% was ethanol from renewable sources [13].

France was also seeking to energize its ethanol fuel research, and in 1901 the French ministry of agriculture offered prizes for the best alcohol-fueled engines and household appliances. An exhibit developed in Germany around 1900 displayed already a variety of alcohol powered application: cars, farm machinery, lamps, stoves, heaters, laundry irons, hair curlers, coffee roasters and every conceivable household appliance and agricultural engines.[1] The exhibit traveled to France, Italy and Spain between 1901 and 1904 and was then sent to

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1 https://digitalcommons.unl.edu/cgi/viewcontent.cgi?article=1005& context=journalismstudent – (accessed 29 July 2019).

the United States and displayed in Norfolk, Virginia, and Baltimore in 1907 and 1908, respectively.

Before World War I the French ethanol fuel program was supported by the Ministry of Agriculture and French biofuel production rose from 2.7 million gallons (about 10.2×10^6 l) in 1900 to 8.3 million (about 31.4×10^6 l) in 1905. Its main purpose was to help support the French sugar beet market and to minimize the rising surplus of domestic other crops. Moreover, the increase in oil imports from Russia and the United States, along with the lack of domestic oil reserves, started to become a major concern. However, without a contentious agrarian movement, the French did not embark on a large-scale distillery building program like the Germans.

After World War I, a French committee recommended that a "national fuel" of 40 to 50% blend of ethanol with gasoline should be created and in 1923 a law was passed (Article Six), mandating a blending level of 10%, later raised to 25% alcohol in 1928. Several ethanol fuels appeared on the market and were branded as "Carburant Poids Lourds" (truck fuel), "Tourisme" and "Supercarburant." Biofuels use peaked in 1935 at 406×10^6 l, accounting for over 7% of all fuel use but declined again towards 1937 due to poor harvests [13].

Concerns on oil resources was one major reason for interest in biofuels at the beginning of the twentieth century in the United Kingdom and a 1907 British commission noted that "a famine in petrol appears to be inevitable." Evaluating the possible substitutes, the commission stated: "Of all the liquid fuels which have been considered by the Committee, the one holding out the greatest promise is alcohol," since it was renewable and production could be "unlimited in any amount" [14]. The enthusiasm about biofuels was so widespread that a 1915 book, entitled "Modern Inventions", had a chapter entitled "Alcohol Motors and the Fuel of the Future" [15].

British foreign policy at the time focused on securing supplies of petroleum from the Middle East. However an Alcohol Motor Fuel Committee was created in 1914, whose task was to evaluate sources of supply, methods of manufacture, and costs of production for alcohol fuel. Seven years later the commission concluded that the cost of alcohol in comparison with petroleum made alcohol a likely fuel only in tropical and remote areas of the world where sugar cane, cassava, Jerusalem artichokes and other crops would give a high yield per acre [16].

One committee member was Harry Ricardo, who at the time was one of world's leading engine designers. In his pioneering 1923 book, "The High-Speed ICE", Ricardo said: "It is a matter of absolute necessity to find an alternative fuel. Fortunately, such a fuel is in sight in the form of alcohol; this is a vegetable product whose consumption involves no drain on the world's storage and which, in tropical countries at all events, can ultimately be produced in quantities sufficient to meet the world's demand, at all events at the present rate of consumption. By the use of a fuel derived from vegetation, mankind is adapting the sun's heat to the development of motive power, as it becomes available from day to day; by using mineral fuels, he is consuming a legacy – and a limited legacy at that – of heat stored away many thousands of years ago. In the one case he is, as it were, living within his income, in the other he is squandering his capital. It is perfectly well

known that alcohol is an excellent fuel, and there is little doubt but that sufficient supplies could be produced within the tropical regions of the British empire" [17]. Two years before he had already patented racing fuels containing ethanol (the composition was a mix of ethanol, methanol, and acetone, containing small amounts of water). These fuels were a huge success and used on racing tracks in Europe and the United States in the 1920s and 1930s respectively. As early as 1928 Harry Ricardo, National Distillers Co. and Shell Oil introduced an alcohol fuel blend in the United Kingdom called "Cleveland Discol." This fuel blend was a popular unleaded gasoline brand and was sold up to 1968 [18].

The French, British, and German biofuels research and laws had a worldwide impact. Engineers and chemists in Asia and Latin America who studied at European universities took home ideas about national self-sufficiency, fuel improvements, and support to local agriculture that would form the basis of biofuels programs in their own countries during the 1930s and into the 1970s.

Based on the research and investigations on ethanol as fuel for a broad range of applications in Germany, France, and the United States, it becomes obvious that countries that produced sugarcane had an economically interesting feedstock available right at their doorstep. Faced with the high cost of gasoline imports and having readily available sugarcane processing equipment, it was logical that developing nations would research and implement ethanol fuel options. Two that were especially active were Brazil and the Philippines.

Brazil is one of the main producers of ethanol fuel (second largest after the United States) and its ethanol history goes back a long way. The first modern commercial ethanol-only powered car, the Fiat 147, was launched on the Brazilian market in July 1979 [19].

Sugarcane had been cultivated in the country since the early sixteenth century. Its automotive fuel history started around 1919 when the governor of the northeastern state of Pernambuco ordered official vehicles to operate on ethanol and by 1921 distilleries in the state started producing ethanol on large scale. Brands of alcohol and gasoline blended fuel were marketed as Azuline and Motorin. Alcohol production grew and Brazil started a first national fuel program. Despite major progress and commercial success, there were still severe operational difficulties to overcome, which forced the redesign of the carburetor and the addition of additives to help with cold starting of the engine.

By 1931 a Brazilian law mandated gasoline importers to buy alcohol in volumes of 5% of their imports. At the time, gasoline cost about 41 cents per gallon, while alcohol was less than half the cost [20]. The number of Brazilian distilleries producing fuel-grade ethanol increased from just one in 1933 to 54 by 1945. Fuel alcohol production rose from $100\,000\,l$ in 1933 to 51.5×10^6 l in 1937, or about 7% of the nation's fuel consumption. To help keep track of research, promote biofuels, and lend technical assistance, Brazil's Instituto do Açúcar e do Álcool was established in 1933. Sales tax exemptions for blends and reductions on taxes of high compression motors for pure alcohol use were also implemented. Much of the interest in alcohol fuels came from sugarcane planters, who often used engines running on pure alcohol. But blends of various proportions were also marketed, including a 90% alcohol and 10% gasoline blend, a 70% alcohol and 30% diethyl ether blend, and a 12% alcohol and 88% gasoline blend.

During the first oil crisis of the 1970s, Brazil once again saw the value of domestically produced fuel and the government started promoting home-grown ethanol through the *Programa Nacional do Álcool*, a nation-wide program launched in 1975. Since 1976, blending gasoline with ethanol has been mandatory in Brazil, with the required percentage varying from 10% to 25%.

United States. The German and French push for an agriculturally based fuel generated a great deal of interest in the United States, leading at first to an 1896 congressional investigation and a decade of debate over the decision of having taxed industrial alcohol off the market.

In 1906 the tax was repealed. US President Theodore Roosevelt supported the repeal and said: "Standard Oil Company has, largely by unfair or unlawful methods, crushed out home competition. It is highly desirable that an element of competition should be introduced by … putting alcohol used in the arts and manufactures upon the free list."

Like their counterparts in Europe, American farmers hoped to recapture some of the markets being lost to the new automotive industry. If horses were being replaced by horseless carriages, perhaps farmers could at least grow the fuel to supply them. However, this proved more difficult than expected due to regulatory and market barriers. Regulatory barriers included the high cost of meeting regulations for "denaturing". Market barriers included the low price of kerosene and gasoline, especially in regions where serious competition emerged. The inventor Alexander Graham Bell expressed a typical, enthusiastic view, when he wrote in National Geographic in 1917 that ethanol "makes a beautiful, clean and efficient fuel… that can be manufactured from corn [maize] stalks, and in fact from almost any vegetable matter capable of fermentation… We need never fear the exhaustion of our present fuel supplies so long as we can produce an annual crop of alcohol to any extent desired."

During the 1920s ethanol was used as gasoline blending component in order to increase the octane level and subsequently reduce engine knocking. At the time the usage of ethanol as octane enhancer was discussed as a viable option, but was abandoned in favor of TEL. In the 1940s the US army operated the first ethanol plant in Nebraska. The produce was solely for the army. Up to the 1970s virtually no commercial fuel ethanol was sold to the general public in the United States.[2]

The US ethanol industry got strong momentum in the 1970s again, when petroleum-based fuel became expensive and environmental concerns involving leaded gasoline created a need for an octane enhancing component. And ethanol helped to fill the gap that the ban of TEL left. Corn became the predominant feedstock for ethanol production because of its abundance and ease of transformation into alcohol. Federal and state subsidies for ethanol helped keep the fuel in production when ethanol prices fell with crude oil and gasoline prices in the early 1980s.

With the phasing out of MTBE as an oxygenate in the United States and a desire to decrease dependence on imported oil and increase the use of environmentally

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2 https://digitalcommons.unl.edu/cgi/viewcontent.cgi?article=1005&
context=journalismstudent – (accessed 29 July 2019).

friendly fuels, the ethanol demand increased dramatically. As a consequence the first renewable fuels standard (RFS) was launched in 2005 and ethanol fuels grades E15 and E85 are well established. Moreover the United States is the world's largest ethanol producer since 2005, with an annual production volume of 15 500 million gallons ($58\,800 \times 10^6$ l), representing almost 60% of the volume manufactured per year worldwide.[3]

1.3.2 Methanol

The English name "*wood alcohol*" was coined when methanol was first discovered by destructive distillation of wood known as *pyrolysis*. Ancient Egyptians used a mixture of substances that included methanol in their embalming process. They already obtained the methanol from pyrolysis of wood. Pure methanol was not isolated until 1661 by Robert Boyle, who produced the chemical through the distillation of boxwood.

Since those very early days dramatic developments have taken place in terms of production technology. Methanol is nowadays produced from methane (synthetic methanol) or from biomass or carbon dioxide (biomethanol).

During the first oil crisis in the early 1970s, it was first proposed as an alternative to petroleum-derived fuels and as recently as 2011 a first biomethanol production plant was constructed in Sweden. The history of methanol as transportation fuel can be called an on–off relationship – with a promising outlook for the future. More details on the history of this alternative fuel and concepts for the transportation sector are provided in Chapter 9.

1.3.3 Vegetable Oils and Hydrotreated Vegetable Oils (HVOs) [21, 22]

Van den Abeele [23] provides an in-depth review of the history of vegetable oils for the transportation sector, covering in particular the early days. The idea to use vegetable oils to power diesel engines is more than a 100 years old. Rudolf Diesel already conducted first experiments with plant oils and one of his prototype engines presented at the World Exhibition in Paris in 1900 ran on peanut oil. He thought that this fuel type would make his engines more attractive to farmers having a source of fuel readily available. In 1912 during a presentation to the British Institute of Mechanical Engineers he remarked, "The fact that fat oils from vegetable sources can be used may seem insignificant today, but such oils may perhaps become in course of time of the same importance as some natural mineral oils and the tar products are now" [24].

Most major European countries with African colonies at the time – Belgium, France, Italy, and the United Kingdom – had an interest in vegetable oil fuels, based on the plants and seeds available locally. Vegetable oils were also used as emergency fuels and for other purposes during World War II. The appearance of petroleum-based fuel in larger quantities made vegetable oils unattractive as automotive fuels and interest in them vanished due to technical and commercial constraints.

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3 http://www.ethanolrfa.org/wp-content/uploads/2017/02/Pocket-Guide-to-Ethanol-2017 .pdf – (accessed 29 July 2019).

The major obstacle for usage of vegetable oils as transport fuels was their kinematic viscosity: it is about an order of magnitude greater than that of conventional, petroleum-derived diesel fuel. As a consequence the fuel was difficult to be injected and/or atomization of the fuel in the engine's combustion chamber was poor. Moreover, depending on the source and composition of the vegetable oil, stability could be an issue, both in terms of fuel storage and in operational problems resulting from engine deposits. Options to overcome these issues have been (and still are) usage of blends of vegetable oils with petroleum-derived fuel (rather than the neat vegetable oil itself) and/or the adaption of the engine hardware or development of dedicated engines. Early approaches are, for example, reports on the development of a new type of diesel engine for motor trucks able to run on a mix of 88% of soy bean oil and 12% of gasoline. Seven hundred trucks in Yunnan and Szechwan had been equipped and operated successfully at the time.

As a result of the energy crises of the 1970s, vegetable oils were revisited as alternatives to petrodiesel fuel, with work commencing in countries such as Austria, Germany, South Africa and the United States. The 1970s also saw the formation of the first commercial enterprise to allow consumers to run straight vegetable oil in their automobiles, i.e. the Elsbett company of Germany. The Elsbett engine is able to run on straight vegetable oil due to several specific design features, which help to overcome the technical constraints of vegetable oils. The engine is making use of preheating of the fuel and incorporates injectors with nozzles optimized towards neat vegetable oils (to improve fuel injection and atomization), a dedicated and heated fuel filter (to overcome cold temperature issues), and technical modifications to extend the glow-plug operation.[4]

The use of methyl esters of sunflower oil to reduce the viscosity of vegetable oil marked the beginning of the rediscovery and eventual commercialization of vegetable oil esters as biodiesel fuel. Use of these esters could overcome some of the critical issues, like high kinematic viscosity, which vegetable oils intrinsically exhibited. The rise of the fatty acid esters lead to the loss of interest in vegetable oils as transport fuels.

Hydrotreating of vegetable oils is a relatively new process and offers an alternative to esterification for producing biobased diesel fuels. Hydrotreated products are called HVO (hydrotreated vegetable oil) or renewable diesel fuels. The hydrotreatment process offers a product that can overcome key detrimental effects of vegetable oils, in particular poor injection/atomization due to high kinematic viscosity, deposit formation and storage stability problems or poor cold temperature properties. HVOs are straight chain paraffinic hydrocarbons that are free of aromatics, oxygen, and sulfur and have high cetane numbers.

1.3.4 Biodiesel/FAME

Nobody knows exactly when the term "biodiesel" was invented. Knothe [25] did research on its first scientific use and points it down to a Chinese paper published in 1988, Gerpen tracks it down to 1984 [26]. The appearance of this expression in

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4 http://www.repowernow.net/cars/diesel/elsbett.htm – (accessed 29 July 2019).

the scientific literature rose exponentially with the start of the 1990s and reflects the increasing interest in this type of liquid for use in ICEs.

The story of the liquid itself started much earlier, in 1853. At the time two chemists, E. Duffy and J. Patrick started research on the transesterification using vegetable oils [27]. Their aim at the time was not the production of fatty acid esters, but the production of soap. Their principal findings formed the foundation of usage of modern fatty acid esters (FAME) and their production by transesterification.

As with all aforementioned alternative biofuels, interest in biodiesel was first spurred by the agricultural community in the context of the industrial revolution and usage of machinery instead of horsepower and the aspiration of national governments to decrease dependency on fossil fuel sources and use of "home-grown" fuel as an alternative. Research and development got specific emphasis and momentum during times of fuel shortages like the two World Wars or the energy crisis of the 1970s. On the other hand, once fossil fuel became readily available again, this interest decreased due to commercial constraints, i.e. the low price of diesel fuel.

Although the performance of vegetable oils was described as satisfactory in many early publications, it was clear that one major technical obstacle for their usage as an automotive fuel was their intrinsic high kinematic viscosity. Papers from the early 1920s already describe operational issues like engine deposits [28, 29]. Solutions considered were engine modifications as well as options making use of pre-heating of the fuel or starting of the engine with conventional diesel. Advanced fuel injection timing was also considered as mitigation strategy and several feasibility studies were published during the 1930s and 1940s [30, 31]. A recognized alternative was the chemical modification of the vegetable oil itself. Early approaches pursued were transesterification, pyrolysis and cracking, which were later complemented by concepts like dilution with conventional diesel fuel, microemulsion and hydrogenation. Biodiesel – obtained by transesterification – represents one, admittedly the currently most successful concept, to overcome this issue.

It all started in 1937, when the Belgian G. Chavanne of the University of Brussels was granted a patent for a "Procedure for the transformation of vegetable oils for their uses as fuels" (Belgian Patent 422,877). This patent described the alcoholysis of vegetable oils using ethanol. This probably is the first account of the production of what is known as "biodiesel" today. A year later a bus fueled with palm oil ethyl ester traveled between Brussels and Louvain – a very early public transport trial. 1942 was the year of the first extensive article on the production and use of palm oil ethyl ester fuel [23]. It highlights the aforementioned bus trial, states the principal miscibility of esters with conventional petroleum-derived fuels and is most likely reporting the first cetane number measurements on biodiesel.

During the energy crisis of the 1970s, the potential value of vegetable oil esters inspired researchers worldwide. Different types of crudes were investigated in different parts of the globe and tests of a huge variety of vegetable oils were made, including amongst others rapeseed, sunflower, castor, palm, cottonseed, and olive oil. The end of the fuel shortage and the subsequent cheap abundance of fossil fuel that followed it led to a halt of activities in this research area.

Interest in fatty acid alkyl ester fuels arose again in the 1980s. The first patent applications on the use of vegetable oil methyl esters as diesel fuel substitute were filed in 1980 by Hartman and Tanaka. At the same time the University of Graz started first test runs on rapeseed methyl esters (RMEs) as diesel fuel. The growing interest in "biodiesel" is reflected by the steep rise in number of publications and patents from the 1980s onwards, rising from nearly zero in 1980 to about 250 in 2010–2012 [27].

The first report on use of esters in the United States dates back to 1982, when an SAE paper reported on the "Evaluation of Cottonseed Oils as Diesel Fuel." This and other early studies concluded that a significant factor that slowed down the development of vegetable oil esters was cost. In the 1980s vegetable oils were considerably more expensive than diesel and it was mentioned that the additional processing would only drive the cost higher.

In 1985 a small pilot plant opened in Silberberg, Austria, producing rapeseed oil methyl esters, using the low-pressure, low-temperature technology. First industrial plants followed in Austria and France. Throughout the 1990s, plants were opened in many European countries, including the Czech Republic, France, Germany and Sweden. In 2017 the four largest producers of biodiesel were the United States, Brazil, Germany, and Argentina (see Table 1.3).

Commercial production started in the United States in 1991 in Kansas City, Missouri. 1995 saw the first US vehicle trial when the University of Idaho provided biodiesel to Yellowstone National Park, which used the fuel in a truck that was driven several hundred thousand miles without damage to the engine and is still in use.

An important milestone was the first standard for RME, issued in 1991 by the Austrian Standardization Institute (ÖN C 1190) followed be the German DIN 51606. Both subsequently were superseded by the European standard EN 14214

Table 1.3 Leading biodiesel producers in 2017.

Country	Production, 10^9 l
United States	6
Brazil	4.3
Germany	3.5
Argentina	3.3
Indonesia	2.5
France	2.3
Thailand	1.4
Spain	1.3
China	1
Poland	1
Canada	0.5
Netherlands	0.4

Source: Data from [32].

in 2014. In 2000 biodiesel was successfully listed as only alternative fuel under EPA's Tier I and Tier II health effect testing under the Clean Air Act (CAA). And with ASTM D6751 the United States published the first biodiesel standard in 2002, followed by ASTM D7467 for blends of biodiesel with petrodiesel in the range from 6 to 20% in 2008.

Because biodiesel and other biofuels can help countries reduce their dependence on imported fossil fuels, and because biofuels frequently produce less air pollution, in particular less net carbon emissions than fossil fuels, governments around the world became very interested in promoting their use. They issued mandates requiring the use of biofuels and offered tax credits for their use. Examples are the European Union (EU)'s "directive for the promotion of the use of biofuels" published in 2003, which stimulated a growth of the biodiesel industry. The directive mandates a minimum market share of biofuel within the total amount of fuels sold on an annual basis. In 2005 the quota was set at 2%, rising to 5.75% in 2010. Together with bioethanol and ETBE, FAMEs are the pivotal contributors to achieve these goals. Another example is the Energy Policy Act of 2005 in the United States, which was intended to reduce the need for imported petroleum that required government fleets to purchase alternatively fueled vehicles. When the Department of Energy decided that biodiesel use could substitute a portion of a fleet's vehicle purchase requirement, biodiesel became an attractive option for some fleet operators.

As a result, production of biodiesel grew rapidly. In 2016 33.2×10^9 l of FAME were produced. Global biodiesel production and consumption is forecasted to rise by 14% from 2016 to 2020, driven by the fulfillment of current biofuel policies in the United States, Argentina, Brazil, Indonesia, and the EU.[5]

Nowadays, fatty acid methyl esters are mainly used in diesel blends or – less often – in pure form. The nature of the vegetable oil crude is varying and depends on factors like regional availability and seasonal/climate requirements (e.g. cold temperature or temperature/storage stability demands).

1.3.5 Liquefied Petroleum Gas (LPG)

Liquefied petroleum gas (LPG) was first mentioned in 1903 when the German pharmacist and chemist Hermann Blau developed successful experiments on the fractionation of gas mixtures. He was able to produce so-called Leuchtgas, a very pure product, which did not contain toxic CO, like the "Stadtgas" known previously. He patented his invention in 1903 and the first commercial plant was opened in Augsburg-Oberhausen.

On the American continent, the father of LPG is the American chemist and explosives experts for the Bureau of Mines, Walter Snelling. He detected mixtures of propane and butane as early as 1910, when he separated gaseous fractions from liquid petroleum ones, thus discovering the existence of propane. Two years later, in 1912, he started his first domestic propane installation and in 1913 he patented producing propane on an industrial scale. Later that year

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5 https://www.spglobal.com/platts/en – (accessed 28 June 2021).

the patent was bought by Frank Philips, the founder of the ConocoPhillips oil company.

LPG use at the time was practically null, but it soon became popular for domestic applications, i.e. lighting, heating, and cooking. LPG was being sold in cylinders as propane and butane or as a less refined mixture of both components. In 1928, LPG was first used as motor fuel (in a truck) and the first refrigerator using LPG as coolant was made. During the Olympic Games in Los Angeles in 1932, it was used for cooking and water heating. In the few following years, demand for LPG was further boosted by the popularity of airships, regularly traveling between Europe and the United States. Using gaseous fuel with roughly the same mass as air was very convenient for airships as it did not alter a zeppelin's overall weight the way liquid fuels did (airships would become considerably lighter once liquid fuels were burned away, thus forcing the release of hydrogen, which was extremely dangerous). Unfortunately, when the Hindenburg – the largest airship ever built – was destroyed in a disaster in 1937, killing 36 people, the zeppelin era ended abruptly.

As early as 1860 the first gas driven combustion engine was patented by the French inventor Etienne Lenoir, who used "Leuchtgas" instead of steam in order to fuel the two-stroke engine (Figure 1.10).

Subsequent developments by Siegfried Markus and George B. Selden paved the way for this technology towards the first major step-change, the invention of the Otto engine by Nikolaus Otto. The first stationary engines were run on gas and so did the first "vehicle", the two-wheeled "Reitwagen". First gas filling stations appeared in Europe in the 1930s in Italy. The first German gas station opened in 1935 in Hannover. By that time, LPG was sourced from petroleum as a condensate when compressing refinery off-gas to extract gasoil.

At the same time countries with a low supply of oil like England and Germany began experimenting with alternative fuels in search of alternatives to fuel their growing car fleets. Among others, experiments were conducted with natural gas (methane) and LPG.

In 1934 Germany kicked off a program to increase independence from oil imports. In 1935 several alternatively fueled vehicles (three diesel, one each fueled with methane, LPG, and methanol as well as two steam powered) were presented at the international automobile fair in Berlin. By 1942, Autogas systems had becoming very popular due to the shortage of conventional fuels. The supply of LPG was stretched beyond petroleum to biomass, i.e. it was synthesized from lignite. Unlike in other markets, German Autogas consumption was about 50 times higher than what was delivered to households for cooking.

These first generation systems were developed for spark ignition engines and heavy duty diesel engines (dual fuel). The liquefied fuel stored in cylinders or fixed tanks was fed in liquid phase to a converter which vaporizes the fuel and regulates the pressure to a set value. The gas is then fed into a mixer located in front of the throttle valve at the beginning of the intake system. The mixer restricts the diameter of the intake accelerating the flow of air at the narrowest point thus locally reducing the pressure. The mixture composition is regulated through the interaction between the regulated pressure and the amount of air flowing through the venturi nozzle. Manual adjustments can be made to ensure satisfying operation

across the entire speed and load ranges. The system was entirely mechanically operated and had very few moving parts.

The technical solutions elaborated in those years set the reference for decades to come. The simple technology spread to other countries and was used in the south of France and in Italy (with both LPG and natural gas). Its main principles are still used today on carbureted engines, like those found on motor scooters or in generators.

LPG became very popular since the mid-1950s in the Netherlands, with the import of American trucks able to run both on gasoline and LPG. Oil refiners (a prominent business branch in the Netherlands) saw an opportunity to generate extra income by marketing LPG, a byproduct in the oil refining process. This trend gained extra momentum by the Suez crisis in the 1950s and the oil crisis of the 1970s, which led to the increase of fossil fuel prices and resulted in LPG being established in the market, available at already 100 filling stations in the Netherlands in 1950. The Netherlands has the fifth-largest Autogas market in the EU after Poland, Italy, Germany, Bulgaria, and Romania, with a share of about 2% of the Dutch market and 40% of all fuel stations offering LPG.[6]

The consumer interest declined when the taxes on the fuel were increased. This changed at the beginning of the twenty-first century when interest in this energy source re-arose because of its favorable environmental impact and subsequently implemented tax incentives in many countries. There are an estimated 16.7 million vehicles driven by LPG worldwide [34], the majority of them in Europe and Asia.

1.3.6 Natural Gas

Naturally occurring natural gas was discovered and identified in America in 1626, when French explorers discovered natives igniting gases that were seeping into and around Lake Erie. In 1821, William Hart dug the first successful natural gas well in the United States in Fredonia, New York. Eventually, the Fredonia Gas Light Company was formed, becoming the first American natural gas distribution company. With its predominant usage in the nineteenth century being lighting houses and streets, the demand now also is covered by its production as a byproduct of coal.

Originating under the surface of the earth in geological formations, natural gas can be obtained in many ways and is often found in conjunction with crude oil. Various methods of processing natural gas from crude oil have been used since the middle of the nineteenth century. But during those early days most of it was flared off or just released into the atmosphere, as reliable pipelines were in short supply. Natural gas at that time was more considered to be an unwanted byproduct. In 1872, a five-mile, two-inch iron pipeline transmitted the first recorded natural gas from the site of Drake's oil discovery in Titusville to Newton, Pennsylvania. By 1891, a line longer than 100 miles connected Chicago to Indiana gas

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6 https://auto-gas.net/government-policies/autogas-incentive-policies/netherlands – (accessed 29 July 2019).

fields. At the turn of the century, natural gas continued to be used primarily for lighting.

In order to be used as a commercial product the issue of transportation had to be solved first – a considerable task as the leakage of the product posed considerable risks. Reliable welding and leak-proof pipe coupling technology developed in the early twentieth century and subsequently the pipeline network began to grow, resulting in the first relatively long-distance lines among major eastern cities in the United States.

Once the transportation issue was satisfactorily resolved, the suppliers began to work on the purification of the product: the conditioning of natural gas by removing unwanted byproducts like "drip gasoline" or "head gasoline" started. As natural gas was taken up for commercial and residential applications, stringent standards of cleanliness were developed and the natural gas industry became strictly regulated. Like crude petroleum products, natural gas undergoes processing and refining to strip the wellhead natural gas product down to the cleaner-burning, purified methane. The materials – particularly associated hydrocarbon liquids such as ethane, propane, and butane – are removed from natural gas so it can be safely transported and processed (including liquefaction and compression).

The "use" of natural gas can be tracked back as early as to the ancient Mesopotamia. Historical and biblical references to humankind's encounters with natural gas include burning springs created when something, perhaps lightening, ignited natural gas seeping out from the earth. The Chinese may have recognized the energy potential for natural gas as early as 900 BCE, but their first known natural gas well was drilled in 211 BCE. In later centuries, the Chinese adapted bamboo pipelines to transport it to provide fuel for boiling water, heating, and the first known instances of lighting. The Japanese dug gas wells as early as 615 CE.

Natural gas-powered vehicles can be traced back to World War I and World War II, when the shortage of gasoline was felt worldwide. Developed out of necessity, "Gas Bag Vehicles" began to appear in France, Netherlands, Germany, and England. Automobiles, buses and trucks were powered by "town gas" or "street gas," a byproduct of the process of turning coal into cokes (used to make iron), captured in a balloon that was usually carried on the roof of the vehicle.

Whereas today's natural gas passenger vehicles are equipped with advanced technologies that compress natural gas (CNG) so more fuel can be carried in the tank, the "gas bag" fuel tanks of the past needed to be much larger to house the uncompressed gas. For every liter of gasoline about two to three cubic meters of gas was needed, meaning that an extremely large fuel tank was needed for any sort of reasonable range. The solution came in the form of a gas storage bag mounted to the roof rack of the vehicle.

Natural gas vehicles (NGVs) are functionally similar to a standard gasoline engine. For this reason, after-market conversions, though costly, can be performed on standard engines, allowing for vehicles to run either solely on natural gas or as a bifuel vehicle, running on both natural gas and gasoline. Most NGVs use space-saving CNG which is stored in fiberglass-protected, composite-metal storage tanks that have evolved over the years into smaller, lighter-weight – yet safer – models, both for original model NGVs and after-market conversions.

Natural gas is used in vehicles running from PCs to heavy-duty trucks and forklifts to commercial fleets, taxies, and buses.

Today 25 million NGVs are in use throughout the world. The fastest growing region for NGVs is Asia-Pacific, followed by North America. Europe has recently been showing a decline in number of NGVs (−10% comparing April 2018 to 2016).[7] The Asia Pacific region is expected to dominate the market in future, due to the largest market share for commercial vehicle and fleet operators. For instance, China is showing growth in refueling infrastructure for CNG and LNG fuel. The America region is second to the Asia Pacific region in the automotive NGV market due to increase in the adoption rate of the natural gas driven vehicle.

Like the vehicle population the number of NG filling stations has been steadily growing and amounted up to 24 036 in 2017, the largest numbers being available in China and Pakistan. 14% of the NG filling stations can be found in Europe, with Italy and Germany exhibiting the largest networks.[7,8,9] In Germany, for instance, seven CNG/LNG filling stations are installed per 100 km motorway.

Today natural gas is wielded and processed on large scale: in 2016 a total of almost 3.6×10^9 m^3 were produced worldwide.[10] The top five natural gas-producing countries were the United States, Russia, Iran, Qatar, and Canada. Applications include domestic heating, generating electricity and as fuel for ICEs (both stationary and mobile). It represents one of the world's most important energy source, but despite its many advantages it still remains a niche as transportation fuel. Though it has been used to fuel cars for decades, the number of NGVs in production remains low and seems to suggest that it will never displace the percentage of standard gasoline engines of which it would be capable. Yet, natural gas reserves could serve as a bridge to future energy technologies.

One of the key drivers for the growing interest in natural gas as transportation fuels is its greenhouse gas (GHG) balance. Since natural gas contains less carbon than other transportation fossil fuels, it produces up to 20% less GHG emissions than comparable gasoline vehicles and up to 15% less than comparable diesel vehicles. Natural gas therefore is still an attractive future alternative energy for the transportation sector, from CNG-powered plug-in hybrids to fuel cells to hydrogen. Presently, natural gas is the largest feedstock for natural gas production, which means that most of the hydrogen used to fill hydrogen-powered vehicles hitting the market is a byproduct of natural gas. Incidentally, though natural gas is a nonrenewable resource, its primary component, methane, can be derived from biomass such as sewage and landfills. There is also the realization that NGVs make a lot of sense for countries rich in gas resources, which is reflected in the number of vehicles and filling stations in countries like China or Russia.

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7 http://www.iangv.org/current-ngv-stats – (accessed 29 July 2019).
8 http://energyeducation.ca/encyclopedia/Natural_gas_refueling_station – (accessed 29 July 2019).
9 http://www.eafo.eu/infrastructure-statistics/natural-gas – (accessed 29 July 2019).
10 https://www.statista.com/statistics/264101/world-natural-gas-production-by-country – (accessed 29 July 2019).

The interested reader is referred to references [35–37] for a more detailed description of the history of CNG as transportation fuel.

1.4   Emission Regulations Worldwide

This section focuses on the tank-to-wheel (TTW) part of the overall emissions generated by a specific type of fuel, i.e. it is restricted to the part which is associated with the combustion of the fuel. It should be noted that the well-tank aspect is also regulated. As with the TTW emissions no global standards exist (a definition of TTW and WTT is given in Section 1.5). In Europe well-to-tank (WTT) emissions are regulated via RED (renewable energy directive) and FQD (fuel quality directive) standard, in California LCFS (low carbon fuel standard) is applied.[11,12,13]

Transport emission standards are the legal requirements regulating air pollutants released into the atmosphere by vehicles. Emission standards set mandatory limits on the permissible amount of specific air pollutants emitted out of the tailpipe. They do also regulate the specific test procedure the vehicle is subjected to in order to determine its emissions. These tests seek to mimic specific operational conditions, e.g. city or extra-urban driving under the tightly controlled conditions of a chassis dynamometer ("rolling road"). Mandatory emission limits have tightened over time and testing procedure have become more stringent. This push has led to a variety of innovations improving combustion, overall engine efficiency, and different strategies in exhaust after-gas treatment, which are described in more detail in Chapter 3.

The objective in setting limits for vehicle emissions is to improve air quality and to protect human health. Despite globalization, emission standards for the transportation sector are not regulated on a global basis. This section is focusing on the standards for Europe, North America, and Japan. It seeks to provide a comprehensive outline of the historical development of the standards in these three regions as well as an overview on latest legislation. The interested reader can find full data for individual countries in footnote[14,15,16].

The enhancement of vehicle standards typically also demanded fuel quality improvements, which were subsequently mandated in changed fuel specifications. In some cases the fuel modifications were necessary to allow the introduction of vehicle technologies that were required to meet the new vehicle emission standards. One prominent example is the adoption of Euro I standards for gasoline vehicles equipped with three-way catalyst, requiring the use of unleaded gasoline.

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11 https://ec.europa.eu/energy/en/topics/renewable-energy/renewable-energy-directive – (accessed 21 March 2021).

12 https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:32009L0030 – (accessed 21 March 2021).

13 www.arb.ca.gov/fuels/lcfs/lcfs.htm – (accessed 21 March 2021).

14 http://www.theicct.org/issues/co2-standards – (accessed 21 March 2021).

15 https://www.epa.gov/regulatory-information-topic/regulatory-information-topic-air#transport – (accessed 21 March 2021).

16 http://www.meti.go.jp/english – (accessed 21 March 2021).

Vehicle emissions contribute to the increasing concentration of gases that are leading to climate change. The principal GHGs associated with road transport are carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O). One example: Road transport is the third largest source of UK's GHGs and accounts for more than 20% of total emissions. Of the total GHG emissions from transport, over 80% are due to CO_2 emissions from road vehicles.

Road transport also remains the main source of many local pollutants including carbon monoxide (CO), nitrogen oxides (NO_x), and particulate matters (PMs). Within urban areas, the percentage of contributions due to road transport is particularly high – in London road transport contributes almost 80% of particulate emissions. There is a growing body of evidence to link vehicle pollutants to human ill health including the incidence of respiratory and cardio-pulmonary disease and lung cancer.

The measurement technologies for the air pollutants CO, NO_x, and CO_2 are well established and mandated since the early 1990s, whereas the evaluation of PM is again a field of ongoing research – despite the fact that an accepted and mandated measurement technology does exist. The EU has based its regulation by setting number-based emission standards. The set limits are based on "solid" particles larger than 23 nm''. Euro 5b/6 light duty diesel vehicles need to comply with a maximum of 6×10^{11} particles/km, corresponding to a mass about 0.15 to 0.7 mg/km, depending on DGN. Gasoline fueled cars had to fulfill an interim standard of 6×10^{12} through 2017 and have to comply to the more stringent diesel standard since.

The US California Air Resources Board (CARB) standard is on the other hand mass based. For light-duty vehicles (LDVs) it was set to a maximum of 1.8 mg/km as of 2017. By 2025 it will be tightened to 0.6 mg/km. It should be noted that meaningful filter mass measurements, the currently mandated PM technology, are very difficult at levels corresponding to these number standards. In addition it is discussed if and how particles of smaller size down to 10 nm should be assessed as well. This particle size may be even more relevant in terms of impact of human health. A more detailed introduction into this field of research can be found at[21].

In addition to the aforementioned so-called regulated emissions, a number of unregulated emissions are also generated during combustion. These air pollutants are typically individual chemical species that are suspected or have a proven potential detrimental impact on the environment, in particular on the human health. Some prominent examples of this category of emissions are aldehydes (e.g. formaldehyde, acetaldehyde), unsaturated hydrocarbons (e.g. 1,3-butadiene), aromatics (e.g. benzene, toluene, xylenes) or polycyclic aromatics.

The generation of these types of emissions is not as universal as the previously described regulated emissions, which are formed in engines running on fossil fuel. In contrast unregulated emissions depend on the type of fuel combusted or engine or exhaust regulation control system used. Research on this type of emissions is ongoing, one focus area being biofuels.[17] The measurement of these

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17 https://www.vtt.fi/inf/julkaisut/muut/2014/VTT-R-03970-14.pdf – (accessed 21 March 2021).

types of pollutants needs to be very specific and is in most cases not (yet) standardized. It is nonetheless important to note that some of these species may become important for future fuel regulation. An extensive literature review on air pollutants (regulated and nonregulated, impact on environment and analytical techniques for their detection) is provided in[18].

The US EPA's Mobile Source Air Toxic (MSAT) list from 2007 discusses a number of them: benzene, 1,3-butadiene, formaldehyde, acetaldehyde, acrolein, polycyclic organic matter (POM), naphthalene, diesel exhaust, and gasoline PM. Benzene, 1,3-butadiene, formaldehyde, and benzo[a]pyrene have been classified as human carcinogens [38, 39]. Acetaldehyde [40] has been classified as a possible carcinogen. Many other compounds, such as polycyclic aromatic hydrocarbons (PAHs) and nitro-PAHs, have been classified as proven, probable or possible carcinogens to humans. Ethene forms potential damages for plants and contributes to the formation of ozone and peroxyacetyl nitrate (PAN) [41]. Risk factors for calculating the cancer potency of exhaust gas are defined by many organizations. OEHHA (2009) defined cancer unit risks and potency factors for 107 carcinogenic substances or groups of substances. The US EPA IRIS (Integrated Risk Information System) is a human health-assessment program that evaluates quantitative and qualitative risk information for effects that may result from exposure to environmental contaminants.

Volatile organic compounds (VOC) contribute to the formation of ground-level ozone together with nitrogen oxides (NO_x) in the presence of heat and sunlight. Exhaust and evaporative emissions from gasoline cars are the dominant source of VOCs within the transport sector, contributing 19% to the total VOCs emitted in 2010. Ozone causes adverse health effects, for example, irritation of the respiratory system, coughing, throat irritation, reduction of lung function, and induced asthma.

Ammonia originates mainly from agricultural sources. However, concern of traffic sources in the production of ammonium aerosols increased as the use of urea-based selective catalytic reduction (SCR) systems for NO_x control for diesel vehicles became common. Ammonia is also formed by the three-way catalysts of the spark-ignited gasoline cars. Nitrous oxide (N_2O), a strong GHG, is also induced by catalyst chemistry under lean conditions [42, 43].

1.4.1 Europe

In 1992, the EU implemented the first emission standard, the Euro 1 standard. Five more standards have been deployed since and details can be seen in Table 3.1 in Chapter 3.

Emission standards: Euro I standard regulations were identically for gasoline and diesel cars. This changed from Euro II stage onwards: EU regulations introduced different emission limits for diesel and gasoline vehicles. Diesels have more stringent CO standards but are allowed higher NO_x emissions. Gasoline powered vehicles were exempted from PM standards through to the Euro IV stage,

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18 http://www.diva-portal.org/smash/get/diva2:802086/FULLTEXT01.pdf – (accessed 21 March 2021).

but vehicles with direct injection engines are subject to a limit of 0.005 g/km for Euro V and Euro VI . A particulate number standard (P) or (PN) has been introduced in 2011 with Euro V b for diesel engines and in 2014 and with Euro VI for gasoline engine (see Table 3.1 of Chapter 3).

Some of the important regulatory steps implementing emission standards for LDVs were:

- Euro II standards (also known as EC 93): Directives 91/441/EEC (PCs only) or 93/59/EEC (PCs and light trucks)
- Euro II standards (EC 96): Directives 94/12/EC or 96/69/EC
- Euro II/IV standards (2000/2005): Directive 98/69/EC, further amendments in 2002/80/EC
- Euro V/VI standards (2009/2014): Regulation 715/2007 and several comitology regulations

Awareness of global warming and the subsequent need to reduce GHG emission kicked off initiatives to reduce CO_2 emissions, which will also help in the field of energy security. In the EU, transport CO_2 emissions have risen by 29% since 1990 with 12% of the total EU emissions now arising from the exhaust of cars.[19,20,21,22,23,24]

The EU first introduced mandatory CO_2 standards for new PCs in 2009, followed by regulations for vans in 2011. The 2009 regulation set a 2015 target of 130 g/km for the fleet average of all manufacturers combined. Individual manufacturers were allowed a higher CO_2 emission value, depending on the average vehicle weight of their fleet. The heavier the average weight of the cars sold by a manufacturer, the higher the CO_2 level allowed. A similar CO_2 standard for new light-commercial vehicles was introduced in 2011. It set a target of 175 g/km for 2017.

At the end of 2013, the European Parliament and the Council of the EU reached an agreement regarding two regulatory proposals that will implement mandatory 2020 CO_2 emission targets for new PCs and light-commercial vehicles in the EU. The PC standards are 95 g/km of CO_2, phasing in for 95% of vehicles in 2020 with 100% compliance in 2021. The light-commercial vehicle standards are 147 g/km of CO_2 for 2020. This means a fuel consumption of around 4.1 l/100 km of petrol or 3.6 l/100 km of diesel.

The 2015 and 2021 targets represent reductions of 18% and 40%, respectively, compared with the 2007 fleet average of 158.7 g/km [36]. The next step in the reduction of CO_2 limits in the EU is expected for 2025, current proposals ranging

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19 https://ec.europa.eu/clima/policies/transport/vehicles/cars_en – (accessed 21 March 2021).
20 https://iea-amf.org/app/webroot/files/file/Country_Annex_Reports_2016_pdf/Annex_44.pdf – (accessed 21 March 2021).
21 http://www.cambridgeparticlemeeting.org/sites/default/files/Presentations/2017/CPM_Kittelson_2017_Particle%20number%20measurements%20Correcting%20for%20losses%20at%2010%20nm%20or%20smaller.pdf – (accessed 21 March 2021).
22 https://www.concawe.eu/wp-content/uploads/2018/02/Real-driving-emissions.pdf – (accessed 21 March 2021).
23 https://www.acea.be/uploads/press_releases_files/RDE-compliant_diesels_November_2018.pdf#page=9 – (accessed 21 March 2021).
24 https://theicct.org/publications/laboratory-road-2014-update – (accessed 21 March 2021).
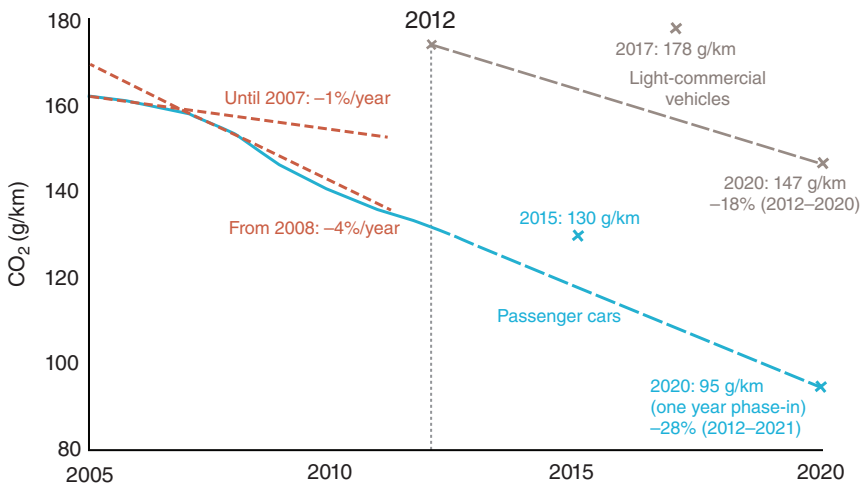
Figure 1.11 Historical development and future targets for CO_2 emission levels of new passenger cars and light-commercial vehicles in the EU. Effects of phase-in, super-credits and eco-innovations not shown here. (http://www.theicct.org/sites/default/files/publications/ICCTu95gram_jan2014.pdf – accessed 06 June 2019).

between 68 and 78 g/km. A synopsis of the historical development and future targets for CO_2 emissions for new passenger cars and light commercial vehicles in the EU are depicted in Figure 1.11.

Implementation of CO_2 targets is not restricted to Europe, but can be observed on a global basis. The EU PC standard of 95 g/km for 2020 can be compared with similar targets for the United States (93 g/km for 2025 PCs), Japan (105 g/km by 2020) and China (117 g/km by 2020). The chart does not take into account any credits (such as super-credits or eco-innovations) or differences in (real-world) enforcement. An overview on the historic development of CO_2 levels for selected countries as well as a synopsis of enacted targets can be found at reference[25].

Regulating Bodies: EU legislative procedures require that any regulatory proposal by the European Commission is to be discussed and voted on in the European Parliament, as well as in the Council of the EU. Once this has successfully been achieved, the National governments of the EU are bound to implement them into National law. In order to enhance the speed of implementation, national governments have the freedom to launch tax incentives, one prominent example being the taxation on low sulfur fuels in Germany at the beginning of the century.

Test Procedures. Emissions are tested over a chassis dynamometer test cycle and expressed in g/km (except PN, which is expressed in 1/km). Over the course of time, there have been several changes to the regulatory emission test cycles in Europe:

● *ECE 15 + EUDC:* The original EU test cycle (also known as the MVEG-A test), including urban and extra-urban segments, performed from a hot start.

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25 https://www.researchgate.net/figure/2-Historical-fleet-CO2-emissions-performance-and-current-standards-g-CO2-km-normalized_fig5_323551777

- *New European driving cycle (NEDC):* Effective from 2000 (Euro III), the ECE 15 + EUDC test was modified to eliminate the 40 s engine warm-up period before the beginning of emission sampling. This modified cold start test was referred to as the NEDC or as the MVEG-B test.
- *WLTP:* The worldwide harmonized light vehicles test procedure (WLTP) and the corresponding worldwide harmonized light vehicles test cycle (WLTC) replaced the NEDC procedure. The transition from NEDC to WLTC is planned as follows:
- *September 2017:* WLTP type approval testing is introduced for new car types. Cars approved using the old NEDC test can still be sold.
- *September 2018*: All new vehicles must be certified according to the WLTP test procedure.
- *January 2019*: All cars at dealers should have WLTP-CO_2 values (with some exceptions for a limited number of vehicles in stock). National governments should adjust vehicle taxation and tax incentives to WLTP values.

During the transition period, European Commission will convert the NEDC-based CO_2 targets to WLTP targets of comparable stringency.

More details on the NEDC and WLTP driving cycle can be found at footlinks[26,27]. The Euro V/V implementing legislation introduced new PM and PN emission measurement methods developed by the UN/ECE Particle Measurement Programme (PMP). The new PM mass measurement method is similar to the US 2007 procedure. The regulatory PM mass emission limits were adjusted to account for differences in results using the old and the new method. PN emissions are measured over the NEDC/WLTC test cycle using the PMP PN method (UN/ECE Reg. 83 Suppl. 7).

Real driving emissions (RDE). In addition to laboratory testing on the chassis dynamometer, vehicle emissions must nowadays also be tested on the road. The RDE testing requirements have been introduced through several regulatory amendments, with the first RDE package published in March 2016, the second in April 2016, and the third in July 2017. The RDE test is performed during vehicle operation using a portable emissions monitoring system (PEMS). The RDE test must last from 90 to 120 minutes. The route must include three segments: urban (<60 km/h), rural (60–90 km/h) and motorway (>90 km/h), in that order and with respective shares of one third. Each segment must cover a distance of at least 16 km.

NO_x emissions must be measured on all Euro VI vehicles (PCs and light-commercial vehicles). On-road PN emissions are to be measured on all Euro VI vehicles, which have a PN limit set (diesel and GDI). CO emissions also have to be measured and recorded on all Euro 6 vehicles. PEMS data must be processed using two methods: the CO_2 moving average window (EMROAD) and the power binning (CLEAR). RDE emission limits are defined by multiplying the respective NEDC emission limit by a conformity factor for a given emission. The conformity factor is subject to an ongoing debate and with RDE package

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26 https://dieselnet.com/standards/cycles/ece_eudc.php – (accessed 19 March 2021).
27 https://dieselnet.com/standards/cycles/wltp.php – (accessed 19 March 2021).

4 the conformity factor was lowered from 1.5 to 1.43 recently. RDE testing requirements are introduced in phases until 2020.

As of November 2018, the fourth package of RDE legislation was published in the Official Journal as part of Regulation (EU) 2018/1832.

A comprehensive review of the uncertainty margins when measuring emissions with PEMS led to a decrease of the NO_x margin from 0.50 to 0.43 at the Euro VId step in 2020. For PN, the PEMS error margin remained unchanged. The two different evaluation methods (i.e. moving average windows (EMROAD) and power binning (CLEAR)) that were allowed under RDE package 3 were proven to lead to diverging results. Consequently one simple evaluation method is now implemented, using the raw emissions for a good percentage of RDE trips, while it balances the raw emissions against CO_2 for the more demanding trips (compared with WLTP). A specific evaluation factor is used in the case of plug-in hybrid vehicles (PHEV); it compares CO_2 emissions during the RDE test and CO_2 emitted over the WLTP test in charge-sustaining mode. The evaluation methodology will be reviewed against technological progress.

A new methodology for checking in-service conformity (ISC) is introduced. Testing compliance will now be made by both the manufacturer and the granting type-approval authority (GTAA), while other entities can make use of an accredited laboratory or a technical service to make tests that are taken fully into account in assessment of compliance. Starting in 2020, GTAA are required to test at least 5% of their approvals per year and per manufacturer. The new ISC also foresees the possibility to perform evaporative and cold temperature tests.

New statistics are implemented to ensure that testing is representative and balanced. The new rules also lead directly to further investigation when vehicles emit amounts significantly over the limit.

All data will be publicly available in order to allow testing and the GTAAs need to report yearly on the ISC performed the previous year in order to improve transparency of the system.

Regulation (EU) 2018/1832 also includes the second act on the WLTP, which aligns EU requirements with the fourth amendment to the Global Technical Regulation (GTR) No. 15.

This includes a substantial upgrade of almost all annexes and appendices of GTR No. 15: e.g. a new gear shift calculation tool, an improved procedure for the determination of the vehicle's road load (with coast down but also with wind tunnel), the correction of several inconsistencies in the previous versions and improved calculation of the test results. The new amendment also brings new EU specific elements, such as the correction of the CO_2 test results based on the difference between the actual speed profile and distance vs. the set points, the revised ambient temperature correction test (ATCT) procedure at 14 ˚C, the revised inducement procedure for urea SCR systems and the revised provisions for CO_2 calculation in the cases of multi-stage and individual type approvals.

The second WLTP act also introduces the new evaporative emissions procedure from GTR No. 19.

Finally, Regulation (EU) 2018/1832 introduces requirements to install standardized and accessible on-board fuel and energy consumption monitors (OBFCM) on all new vehicle types from 2020 and on all new vehicles from 2021.

This obligation applies only to vehicles running with liquid fuels (gasoline, diesel and biofuels). The accuracy of the OBFCM is checked at type-approval on the basis of the WLTP tests.

In 2017 two Concawe studies were commissioned to gain information on on-road emissions performance of latest Euro VI b and diesel PCs. Expected challenges with the new certification procedures relate to three areas. Firstly urban driving conditions may not allow SCR aftertreatment devices to reach an efficient operating temperature. Secondly the high load conditions that may lead to exhaust temperatures too high for lean NO_x traps to be effective and thirdly low-temperature conditions, which may limit the use of EGR (a well-established NO_x emission reduction technology). The test results with state-of-the-art Euro VI diesel PCs show that they are able to meet near future NO_x emission limits of moderate RDE testing commensurate with Euro VI d. Combinations of emissions control technologies, for example, long and short route EGR, large volume SCR and possibly LNT, will be required. Vehicles equipped with urea SCR systems can reduce tailpipe NO_x, bay reactive urea reductant dosing varying the urea consumption, thereby enabling acceptable NO_x emissions even over high-duty drive cycles.[28]

End of 2018, the European Automobile Manufacturers' Association (ACEA) issued a press release emphasizing the low on-road NO_x emission of recent diesel vehicles certified to the Euro VI d-temp standard.

ACEA said that some 270 new types of diesel cars type-approved against the latest Euro 6d-temp standard were introduced on the European market over the past year. The new data shows that all of these diesel cars performed well below the NO_x threshold of the RDE test, which applies to all new car types since September 2017. What is more, already today most of these vehicles showed results that are below the stricter NO_x threshold that will be mandatory from January 2020 (https://www.acea.be/industry-topics/tag/category/real-driving-emissions-test).

These findings were supported by third parties. The German motor club ADAC published an updated overview of the NO_x performance of new diesel cars. Based on 243 tests of Euro VI c and Euro VI d-temp vehicles, real-world NO_x emissions measured on the ADAC Ecotest are in average at 81 mg/km, very close to the Euro VI limit of 80 mg/km. This represents a tremendous improvement of real-world NO_x emissions compared with Euro V and Euro VI b cars performance.

Defeat Devices. For LDVs, EU regulations define a "defeat device" as: "any element of design which senses temperature, vehicle speed, engine speed (RPM), transmission gear, manifold vacuum or any other parameter for the purpose of activating, modulating, delaying or deactivating the operation of any part of the emission control system, that reduces the effectiveness of the emission control system under conditions which may reasonably be expected to be encountered in normal vehicle operation and use." The regulations prohibit defeat devices but provide situations under which the prohibition does not apply. This allows manufacturers the option of disabling emission control system components to protect the engine/vehicle and to facilitate starting.

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28 https://www.concawe.eu/wp-content/uploads/2018/02/Real-driving-emissions.pdf

1.4.2   United States

The history of emission legislation in the United States dates back to the 1960s. The State of California first legislated tailpipe emission standards for 1966 model year cars sold in that state, followed by the United States as a whole in model year 1968. Also in 1966, the first emission test cycle was mandated by the State of California. As in Europe over time the standards were progressively tightened year by year, prescribed by the EPA. In contrast to European jurisdiction, US standards regulate gasoline and diesel cars jointly, whereas the EU developed separate, individual regulations for the two.

Regulating Bodies: In the United States, overall responsibility belongs to the EPA. Its authority to regulate engine emissions – and the air quality in general – is based on the CAA, most recently amended in 1990. Fuel economy standards are developed by the National Highway Traffic Safety Administration (NHTSA), an agency within the US Department of Transportation (DOT). The State of California has the right to adopt its own emission regulations, which are often more stringent than the federal rules. Engine and vehicle emission regulations are adopted by the CARB, a regulatory body within the California EPA. California is the only state vested with the authority to develop its own emission regulations. Other states have a choice to either implement the federal emission standards, or else to adopt California requirements.

Emission Standards. The current emission standards for LDVs are the Tier 3 standards, which follow the earlier Tier 2, Tier 1, and pre-Tier 1 federal emission regulations. The emissions are evaluated in the so-called federal test procedure (FTP-75) driving cycle.[29] Fuel economy for light-duty vehicles is assessed in the Highway Fuel Economy Test (HWFET or HFET), also a chassis dynamometer protocol. As a highway test cycle, it complements the FTP-75 one, targeting city driving.[30]

Two sets of standards had been defined for LDVs in the Clean Air Act Amendments (CAAA) of 1990: Tier 1 and Tier 2. Once Tier 2 standards were fully phased-in, the EPA adopted the Tier 3 emission regulations.

During the phase-in period, manufacturers are required to certify an increasing percentage of their new vehicle fleet to the new standards, with the remaining vehicles still certified to the preceding tier of emission regulations.

The US emission standards establish certification emission limits applicable to each certified vehicle as well as fleet average standards. The fleet average standards – for NO_x at the Tier 1 and Tier 2 stages and for non-methane organic gases (NMOG) + NO_x at Tier 3 area – form significant difference of the US emission regulations from other light-duty emission standards for air pollutants across the world, which typically include only certification type approval limits.

- **Tier 1 standards** were published in 1991 and phased-in progressively between 1994 and 1997. Tier 1 standards applied to all new LDV of less than 8500 lb.

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29 https://dieselnet.com/standards/cycles/ftp75.ph
30 https://dieselnet.com/standards/cycles/hwfet.php

Table 1.4 EPA Tier 1 emission standards for passenger cars and light-duty trucks, FTP 75, g/mile.

Category	50 000 miles/5 yr						100 000 miles/10 yr[a]					
	THC	NMHC	CO	NO_x[b] diesel	NO_x gasoline	PM[c]	THC	NMHC	CO	NO_x[b] diesel	NO_x gasoline	PM[c]
Passenger cars	0.41	0.25	3.4	1.0	*0.4*	0.08	—	0.31	4.2	1.25	*0.6*	0.10
LLDT, LVW < 3750 lbs	—	0.25	3.4	1.0	*0.4*	0.08	0.80	0.31	4.2	1.25	*0.6*	0.10
LLDT, LVW > 3750 lbs	—	0.32	4.4	—	*0.7*	0.08	0.80	0.40	5.5	0.97	*0.97*	0.10
HLDT, ALVW < 5750 lbs	0.32	—	4.4	—	*0.7*	—	0.80	0.46	6.4	0.98	*0.98*	0.10
HLDT, ALVW > 5,750 lbs	0.39	—	5.0	—	*1.1*	—	0.80	0.56	7.3	1.53	*1.53*	0.12

Abbreviations: LVW, loaded vehicle weight (curb weight + 300 lbs); ALVW, adjusted LVW (the numerical average of the curb weight and the GVWR); LLDT, light light-duty truck (below 6000 lbs GVWR); HLDT, heavy light-duty truck (above 6000 lbs GVWR).
a)   Useful life 120 000 miles/11 yr for all HLDT standards and for THC standards for LDT.
b)   More relaxed NO_x limits for diesels applicable to vehicles through 2003 model year.
c)   PM standards applicable to diesel vehicles only.
Source: EPA, Tier 1 emission standards for passenger cars and light-duty trucks, FTP 75.

gross vehicle weight rating (GVWR). The Tier 1 emission standards are summarized in Table 1.4. Car and light truck emissions were measured over the FTP 75 and expressed in g/mile.

- **Tier 2 standards** were adopted in 1999, with a phase-in from 2004 to 2009. They extended the light-duty emission standards to medium-duty passenger vehicles (MDPVs) with GVWR between 8500 and 10 000 lbs. This category includes primarily larger sports utility vehicles (SUVs) and passenger vans. The EPA Tier 2 program uses a three-tiered compliance strategy. Pre-production evaluation is used to certify vehicles prior to sale. A production evaluation is used on the assembly line for early evaluation of production vehicles. Finally in-use evaluation is used to verify properly maintained vehicles after several years of use. The Tier 2 emission standards are structured into eight permanent and three temporary certification levels of different stringency, called "certification bins," and an average fleet standard for NO_x emissions. Vehicle manufacturers have a choice to certify particular vehicles to any of the available bins. When fully implemented in 2009, the average NO_x emissions of the entire LDV fleet sold by each manufacturer had to meet the average NO_x standard of 0.07 g/mile. The temporary certification bins (bin 9, 10, and an MDPV bin 11) with more relaxed emission limits were available in the phase-in period only

and ceased to exist after the 2008 model year. *Tier 2* vehicles are those meeting the requirements of one of the available bins *and* that are used to meet the requirement that a percentage of the fleet have average NO_x emissions of 0.07 g/mile. The emission standards for all pollutants (certification bins) when tested on the FTP are shown in Table 1.5.

- **Tier 3 standards** were finalized in 2014, to be phased-in between 2017 and 2025, Tier 3 regulations additionally include emission standards for chassis-certified heavy-duty vehicles up to 14 000 lbs. The approach of the Tier 3 standards is similar to the Tier 2 ones – manufacturers must certify vehicles to one of seven available "certification bins" and must meet a fleet-average emission standard for their vehicle fleet in a given model year. The standards are more stringent than Tier 2 standards and include a number of other important changes: Both the certification limits (bins) and the fleet average standards are expressed using the sum of $NMOG + NO_x$ emissions, the fleet average $NMOG + NO_x$ emissions must reach 30 mg/mile (Bin 30 = Tier 2 Bin 2) by 2025, and the required emission durability has been increased to 150 000 up from 120 000 miles. Gasoline vehicles are tested – for exhaust and evaporative emissions – using gasoline containing 10% of ethanol (E10) (Table 1.6).

Tier 3 PM standards are prescribed per each certified vehicle separately and they are not fleet average standards. However, due to uncertainties in regards to future vehicle technologies (e.g. gasoline direct injected engines or start-stop systems) and to PM measurement methods at ultra-low levels, EPA adopted a five year phase-in period for PM standards. The phase-in schedule is based on percentage of sales. A relaxed in-use FTP PM standard of 6 mg/mile applies during the phase-in period.

Fuel economy in new LDVs has been regulated since the 1970s by corporate average fuel economy (CAFE) standards administered by the NHTSA, an agency within the DOT. The first GHG regulations for motor vehicles were adopted in 2002 in California. At the federal level, GHG emission standards and harmonized CAFE legislation for LDVs were adopted by the EPA and the NHSTA in 2010 and 2012. The aimed results are an average industry fleetwide level of 163 g/mile of carbon dioxide (CO_2) in model year 2025, which is equivalent to 54.5 miles per gallon (mpg) (if achieved exclusively through fuel economy improvements).

The standards were established in two phases:

- Phase 1 – Model years 2012–2016
- Phase 2 – Model years 2017–2025

California emission regulations for LDVs are known as so-called *low emission vehicle* (LEV), ultra low emission vehicles (ULEV), super ultra low emission vehicles (SULEV), and zero emission vehicles (ZEV) standards. The succession of California standards has generally preceded and exceeded the respective federal standards.

While the California LEV II program was similar in structure to the federal tier 2 legislation, there were a number of differences regarding overall fleet emission stringency, durability and categorization of vehicles. One difference is that the

Table 1.5 Tier 2 Emission standards, FTP 75, g/mile.

Bin#	Intermediate life (5 years/50000 miles)					Full useful life				
	NMOG[a]	CO	NO$_x$	PM	HCHO	NMOG[a]	CO	NO$_x$[b]	PM	HCHO
Temporary bins										
11 MDPV[c]						0.280	7.3	0.9	0.12	0.032
10[d)e)f)g)]	0.125 (0.160)	3.4 (4.4)	0.4	—	0.015 (0.018)	0.156 (0.230)	4.2 (6.4)	0.6	0.08	0.018 (0.027)
9[d)e)g)h)]	0.075 (0.140)	3.4	0.2	—	0.015	0.090 (0.180)	4.2	0.3	0.06	0.018
Permanent bins										
8[e]	0.100 (0.125)	3.4	0.14	—	0.015	0.125 (0.156)	4.2	0.20	0.02	0.018
7	0.075	3.4	0.11	—	0.015	0.090	4.2	0.15	0.02	0.018
6	0.075	3.4	0.08	—	0.015	0.090	4.2	0.10	0.01	0.018
5	0.075	3.4	0.05	—	0.015	0.090	4.2	0.07	0.01	0.018
4	—	—	—	—	—	0.070	2.1	0.04	0.01	0.011
3	—	—	—	—	—	0.055	2.1	0.03	0.01	0.011
2	—	—	—	—	—	0.010	2.1	0.02	0.01	0.004
1	—	—	—	—	—	0.000	0.0	0.00	0.00	0.000

a) For diesel fueled vehicle, NMOG (non-methane organic gases) means NMHC (non-methane hydrocarbons).
b) Average manufacturer fleet NO$_x$ standard is 0.07 g/mile for Tier 2 vehicles.
c) An additional temporary bin restricted to MDPVs expires after model year 2008.
d) Bin deleted at end of 2006 model year (2008 for HLDTs).
e) The higher temporary NMOG, CO, and HCHO values apply only to HLDTs and MDPVs and expire after 2008.
f) Optional temporary NMOG standard of 0.195 g/mile (50 000) and 0.280 g/mile (full useful life) applies for qualifying LDT4s and MDPVs only.
g) −50 000 mile standard optional for diesels certified to bins 9 or 10.
h) Optional temporary NMOG standard of 0.100 g/mile (50 000) and 0.130 g/mile (full useful life) applies for qualifying LDT2s only.
Source: EPA, Tier 2 Emission standards, FTP 75.

Table 1.6 Tier 3 fleet average NMOG + NO$_x$ SFTP standards (mg/mile).

Vehicle category	2017[a]	2018	2019	2020	2021	2022	2023	2024	2025
LDV, LDT1	86	79	72	65	58	51	44	37	30
LDT2, LDT3, LDT4, MDPV	101	92	83	74	65	56	47	38	30

a) For LDVs and LDTs over 6000 lbs GVWR and MDPVs, the fleet average standards apply beginning in MY 2018.
Source: EPA, Tier 3 fleet average NMOG+NOx FTP standards.

federal approach uses eight certification "bins" (with Tier 2 Bin 5 being similar to California's LEV, and Tier 2 Bin 2 similar to SULEV) to allow averaging across a greater level of emission diversification in the fleet. Another important difference is that, when fully implemented in 2010, the federal fleet average NMOG emissions – around 0.090 g/mile, based on Bin 5 – can be more than twice as high as the LEV II NMOG levels. However the overall, recent tendency has been that of increasing harmonization, i.e. the regulations for Tier 3 are closely aligned for California and the federal US.

The major regulatory steps in the evolution of California emission standards include:

- **Tier 1/LEV I** California emission standards extended through the year 2003.
- **LEV II** California regulations were phased-in through model years 2004–2010.
- **LEVIII** California regulations are phased-in through model years 2015–2025.

The same standards for gaseous pollutants are applied to diesel- and gasoline-fueled vehicles. PM standards applied to diesel vehicles only. Emissions are measured over the FTP-75 test. The additional SFTP procedures were phased-in in California between 2001 and 2005.

Car manufacturers were required to produce a percentage of vehicles certified to increasingly more stringent emission categories, according to schedules based on vehicle fleet emission averages for each manufacturer. After 2003, Tier 1 and TLEV standards were eliminated as available emission categories. Tables 1.7–1.9 provide an overview on the LEV standards for LDVs:

There are several changes when moving from LEV I to LEV III standards: NMOG and NO_x standards are combined into one $NMOG + NO_x$ standard. A more stringent combined $NMOG + NO_x$ fleet average requirement for 2015–2025 model years was implemented and the durability requirements for emission control systems were extended from 120 000 to 150 000 miles.

The PM emission standards and time schedule are shown in Table 1.9. The LEV III proposal originally proposed a particulate emission compliance option using a solid particle number (SPN) limit. This suggestion was withdrawn from the final regulation. The proposed limit was 3×10^{12} 1/mile. SPN emissions were to be measured over the FTP-75 cycle using a sampling approach patterned after the European PMP method.

The United States have two sets of parallel standards: firstly the so-called CAFE standards (corporate average fuel economy), which are adopted by the National Highway Traffic Safety Administration (NHTSA). NHTSA is an agency within the DOT. Secondly, greenhouse gas emissions as adopted by the EPA exist. The first CAFE standards were set in the 1970s, as a consequence of the oil crisis at the time. The Energy Policy Conservation Act of 1975 established the first CAFE standards for light-duty vehicles. For cars these standards aimed to double the fleet average fuel economy from 13.6 miles per gallon in 1974 to 27.5 mpg by 1985. In 2007, more progressive fuel economy and GHG emission regulations were implemented: The Energy Independence and Security Act (EISA) of 2007 mandated a 40% increase in fuel economy by 2020. As of model year 2011 onwards a combined average fuel economy of 35 mpg was aimed for. In 2007, the U.S. Supreme Court also stated that CO_2 is a pollutant under the Clean Air Act (CAA)

Table 1.7 LEV emission standards for light-duty vehicles; FTP-75, g/mile.

Category	50 000 miles/5 yr					100 000 miles/10 yr				
	NMOG[a]	CO	NO$_x$	PM	HCHO	NMOG[a]	CO	NO$_x$	PM	HCHO
Passenger cars										
Tier 1	0.25	3.4	0.4	0.08	—	0.31	4.2	0.6	—	—
TLEV	0.125	3.4	0.4	—	0.015	0.156	4.2	0.6	0.08	0.018
LEV	0.075	3.4	0.2	—	0.015	0.090	4.2	0.3	0.08	0.018
ULEV	0.040	1.7	0.2	—	0.008	0.055	2.1	0.3	0.04	0.011
LDT1, LVW < 3750 lbs										
Tier 1	0.25	3.4	0.4	0.08	—	0.31	4.2	0.6	—	—
TLEV	0.125	3.4	0.4	—	0.015	0.156	4.2	0.6	0.08	0.018
LEV	0.075	3.4	0.2	—	0.015	0.090	4.2	0.3	0.08	0.018
ULEV	0.040	1.7	0.2	—	0.008	0.055	2.1	0.3	0.04	0.011
LDT2, LVW > 3750 lbs										
Tier 1	0.32	4.4	0.7	0.08	—	0.40	5.5	0.97	—	—
TLEV	0.160	4.4	0.7	—	0.018	0.200	5.5	0.9	0.10	0.023
LEV	0.100	4.4	0.4	—	0.018	0.130	5.5	0.5	0.10	0.023
ULEV	0.050	2.2	0.4	—	0.009	0.070	2.8	0.5	0.05	0.013

Abbreviations: LVW, loaded vehicle weight (curb weight + 300 lbs); LDT, light-duty truck; NMOG, non-methane organic gases; HCHO, formaldehyde.
a)   NMHC for all Tier 1 standards.
Source: EPA, LEV Emission standards for light-duty vehicles, FTP-75.

Table 1.8 LEV II emission standards for passenger cars and LDVs < 8500 lbs (LDT1 and LDT2), FTP-75, g/mile.

Category	50 000 miles/5 yr					120 000 miles/11 yr				
	NMOG	CO	NO$_x$	PM	HCHO	NMOG	CO	NO$_x$	PM	HCHO
LEV	0.075	3.4	0.05	—	0.015	0.090	4.2	0.07	0.01	0.018
ULEV	0.040	1.7	0.05	—	0.008	0.055	2.1	0.07	0.01	0.011
SULEV	—	—	—	—	—	0.010	1.0	0.02	0.01	0.004

Source: EPA, LEV II emission standards for passenger cars and LDVs < 8500 lbs (LDT1 & LDT2), FTP-75.

and as a consequence needs regulation by EPA. NHTSA and EPA finalized new and harmonized CAFE and GHG emission rules in 2010 for model year 2012 to 2016 light-duty vehicles, targeting an average CAFE fuel economy of 34.1 mpg (6.9 l/100km) and CO_2 emissions of 250 g/mile in model year 2016 vehicles. As of 2012, mandatory limits were extended to model year 2017 bo 2025 vehicles, with a projected average standard of 163 g/mile. For 2012–2016 standards vehicle size dependent CO_2 emission-footprint curves were defined. This approach is based on the reformed CAFE standards for light trucks. The rationale is, the larger the

Table 1.9 LEV III particulate matter emission standards, FTP-75.

Vehicle type	PM limit (mg/mile)	Phase-in
PCs, LDTs, MDPVs	3	2017–2021
	1	2025–2028
MDVs 8501–10 000 lbs	8	2017–2021
MDVs 10 001–14 000 lbs	10	2017–2021

Source: EPA, LEV III particulate matter emission standards, FTP-75.

vehicle footprint, the higher the corresponding vehicle CO_2 emission target. In consequence each manufacturer has to comply to an individual target, defined by his vehicle production slate. The EPA CO_2-equivalent fuel economy figures are different from the CAFE ones because EPA allows additional CO_2 credits for air conditioning improvements. The CO_2 emissions are tested over the EPA-2-cycle test, a combination of FTP-75 and HWET test. In addition maximum limits for N_2O and methane emissions were defined (measured in the FTP-72 test): a maximum of 0.010 gpm for N_2O and a maximum of 0.030 gmp for methane. The regulation also includes a system of averaging, banking and trading of credits. Credit trading is allowed among all vehicles a manufacturer produces as well as between companies. Also credits for improvements, e.g. for advanced GHG/fuel economy technologies such as electric/plug-in hybrids, electric/fuel cell vehicles or alternative fuel based vehicles are realized.

In 2002, California adopted the first GHG control regulation legislation, which became effective from 1 January 2006.

The standards phase-in schedule is from 2009 to 2016, as shown in Table 1.10. The average reduction of GHGs from new California cars and light trucks is

Table 1.10 California fleet average GHG emission standards.

Time frame	Year	GHG standard, g CO_2/mile (g CO_2/km)		CAFE equivalent, mpg (l/100 km)	
		PC/LDT1	LDT2	PC/LDT1	LDT2
Near term	2009	323 (201)	439 (274)	27.6 (8.52)	20.3 (11.59)
	2010	301 (188)	420 (262)	29.6 (7.95)	21.2 (11.10)
	2011	267 (166)	390 (243)	33.3 (7.06)	22.8 (10.32)
	2012	233 (145)	361 (225)	38.2 (6.16)	24.7 (9.52)
	2013	227 (142)	355 (221)	39.2 (6.00)	25.1 (9.37)
Medium term	2014	222 (138)	350 (218)	40.1 (5.87)	25.4 (9.26)
	2015	213 (133)	341 (213)	41.8 (5.63)	26.1 (9.01)
	2016	205 (128)	332 (207)	43.4 (5.42)	26.8 (8.78)

Source: EPA, California fleet average GHG emission standards.

expected to be about 22% in 2012 and about 30% in 2016, compared with model year 2004 vehicles (however, in absolute terms GHG emissions would still increase due to projected growth in vehicle population and miles traveled).

The GHG standards are incorporated into the California LEV legislation. There are two fleet average GHG requirements: (i) for passenger car/light-duty truck 1 (PC/LDT1) category, which includes all PC/LDTs below 3750 lbs. equivalent test weight (ETW); and (ii) for light-duty truck 2 (LDT2) category, including light trucks between 3751 lbs. ETW and 8500 lbs. gross vehicle weight (GVW). In addition, MDPVs from 8500 to 10 000 lbs. GVW are included in the LDT2 category for GHG emission standards.

The GHG standards are defined in grams per mile of CO_2-equivalent emissions (g CO_2/mile in Table 1.10), calculated from the following formula:

$$CO_2\text{-Equivalent} = CO_2 + 296 \times N_2O + 23 \times CH_4 - AC \text{ Allowances}$$

1.4.3 Japan

Japan introduced fist engine emissions standards for new on-road LDVs and heavy duty engines in the late 1980s. The Japanese standards, however, remained very moderate through the 1990s. A step-change occurred in 2003, when very stringent 2005 emission standards for both light and heavy vehicles were adopted. At the time they came to power, the 2005 heavy-duty emission standards ($NO_x = 2$ g/kWh, PM = 0.027 g/kWh) were the most stringent diesel emission regulation in the world. Effective 2009, these limits were further tightened to a level in-between the US 2010 and Euro V requirements, and the 2016 limits are comparable in stringency with the US 2010 and Euro VI standards. While certification testing has been conducted over Japan-specific drive cycles, emissions will be tested using the worldwide harmonized light vehicles test procedure (WLTP) starting in 2018.

Two emission programs were implemented in the early 2000s to reduce emissions from *in-use diesel vehicles*: the "Automotive NO_x and PM Law" and the "Tokyo retrofit program." As the vehicle population targeted by these programs is no longer in circulation, the regulations have only historical significance.

Regulating Bodies: The Automotive NO_x and PM law regulation designated a total of 196 communities in the Tokyo, Saitama, Kanagawa, Osaka, and Hyogo Prefectures as areas with significant air pollution due to nitrogen oxides emitted from motor vehicles. Under the Law, several measures had to be taken to control NO_x from in-use vehicles, including enforcing emission standards for specified vehicle categories. The regulation was amended in June 2001 to tighten the existing NO_x requirements and to add PM control provisions. The amended rule was called the "*Law Concerning Special Measures to Reduce the Total Amount of Nitrogen Oxides and Particulate Matter Emitted from Motor Vehicles in Specified Areas*" or in short the *Automotive NO_x and PM Law*. The amended regulation became effective in October 2002.

The Tokyo government and several neighboring prefectures adopted diesel emission regulations, which required retrofitting of older in-use diesel vehicles with PM control devices (catalytic converters or particulate filters), or else

replacing them with newer, cleaner models. The Tokyo retrofit requirements became effective in October 2003.

Japanese emission standards for engines and vehicles and fuel efficiency targets are jointly developed by a number of government agencies, including the Ministry of the Environment, the Ministry of Land, Infrastructure, and Transport and the Ministry of Economy, Trade, and Industry. In developing engine emission standards and policies, the Ministry of the Environment relies on recommendations of its advisory body known as the Central Environment Council (CEC).

Engine and vehicle emission standards are developed under the authority of the "Air Pollution Control Law," while fuel efficiency targets are adopted under the "Law Concerning the Rational Use of Energy" (Energy Conservation Law).

Emission Standards. As in the EU individual standards are set for gasoline and for diesel fueled vehicles. Japanese regulation differentiates between two types of exhaust emission, denoted as "mean" and "max." The "mean" standards are to be met as a type approval limit and as a production average. The "max" standards are to be met generally as an individual limit in series production and as type approval limit if sales are less than 2000 per vehicle model per year. The "max" standards are shown in brackets in the following Tables 1.11 and 1.12. Implementation dates in the tables refer to new vehicle models. Current production models are often allowed a delay of up to one year.

Table 1.11 Japanese emission standards for diesel passenger cars, g/km.

Curb weight (kg)	Date	Test	CO mean (max)	HC mean (max)	NO$_x$ mean (max)	PM mean (max)
<1250[a]	1986		2.1 (2.7)	0.40 (0.62)	0.70 (0.98)	
	1990	10–15 mode	2.1 (2.7)	0.40 (0.62)	0.50 (0.72)	
	1994		2.1 (2.7)	0.40 (0.62)	0.50 (0.72)	0.20 (0.34)
	1997		2.1 (2.7)	0.40 (0.62)	0.40 (0.55)	0.08 (0.14)
	2002[b]		0.63	0.12	0.28	0.052
	2005[c]	JC08[d]	0.63	0.024[e]	0.14	0.013
	2009[d]		0.63	0.024[e]	0.08	0.005
>1250[a]	1986		2.1 (2.7)	0.40 (0.62)	0.90 (1.26)	
	1992	10–15 mode	2.1 (2.7)	0.40 (0.62)	0.60 (0.84)	
	1994		2.1 (2.7)	0.40 (0.62)	0.60 (0.84)	0.20 (0.34)
	1998		2.1 (2.7)	0.40 (0.62)	0.40 (0.55)	0.08 (0.14)
	2002[b]		0.63	0.12	0.30	0.056
	2005[c]	JC08[d]	0.63	0.024[e]	0.15	0.014
	2009[f]		0.63	0.024[e]	0.08	0.005

a) Equivalent inertia weight (EIW); vehicle weight of 1265 kg.
b) 2002.10 for domestic cars, 2004.09 for imports.
c) Full implementation by the end of 2005.
d) Full phase-in by 2011.
e) Non-methane hydrocarbons.
f) 2009.10 for new domestic models: 2010.09 for existing models and imports.
Source: Japanese emission standards for diesel passenger cars. Licensed under CC BY 3.0 SA.

Table 1.12 Emission standards for four stroke gasoline passenger vehicles.

Date[a]	Test mode	Unit	Maximum allowable emissions[b]			
			CO	NMHC[c]	NO$_x$	PM
1973	10 mode	g/km	26.0	3.80	3.00	—
1975[d]	10 mode	g/km	2.70	0.39	1.60	—
	11 mode	g/test	85.0	9.50	11.0	—
1976	10 mode	g/km	2.70	0.39	1.20	—
	11 mode	g/test	85.0	9.50	9.00	—
1978	10 mode	g/km	2.70	0.39	0.48	—
	11 mode	g/test	85.0	9.50	6.00	—
2000	10 mode	g/km	1.27 (0.67)	0.17 (0.08)	0.17 (0.08)	—
	11 mode	g/test	31.1 (19.0)	4.42 (2.20)	2.50 (1.40)	—
2005	10–15 mode + 11 mode	g/km	1.92 (1.15)	0.08 (0.05)	0.08 (0.05)	—
2008	10–15 mode + JCo8C	g/km	1.92 (1.15)	0.08 (0.05)	0.08 (0.05)	—
2009	JCo8H + C	g/km	1.92 (1.15)	0.08 (0.05)	0.08 (0.05)	0.007 (0.005)

a) Application to new models produced by domestic manufacturers. Applicability to continuing models and imports typically delayed by one to two years.
b) Values in parentheses denote regulatory (average) limits.
c) HC prior to 1999.
d) Inertial weight greater than 1000 kg.
Source: JAPAN: LIGHT-DUTY: EMISSIONS, GASOLINE VEHICLES, Four stroke gasoline passenger vehicles. Licensed under CC BY 3.0 SA.

Emission standards for new diesel fueled vehicles are listed in Table 1.11, assessed in the 10–15 mode cycle (Figure 1.11). This test cycle superseded the older 10-mode cycle effective November 1991 for domestic cars and April 1993 for imports. The 2005 regulation introduced a new JC08 mode cycle, which was phased-in by 2011. Vehicles are tested using 50 ppm S fuel for the 2005 standards. From 2018 onwards the emissions will be tested over the WLTP cycle. Table 1.12 summarizes the standards for gasoline powered PCs.

In Japan, CO_2 emissions from the transport sector accounted for about 19% of total CO_2 emissions, of which 90% generated from road transport. While total worldwide CO_2 emissions from the transport sector have been increasing, CO_2 emissions from Japan's transport sector peaked in 2001 and have since been on a steady decline[31]. The sector's total CO_2 emissions in 2001 amounted to 267 million tons, but by 2007 this decreased to 246 million tons, over-achieving the projected reduction target for 2010 (240 to 243 million tons).

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31 https://www.japanfs.org/en/news/archives/news_id030151.html – (accessed 21 March 2021).

An overview on the evolution on PC fuel economy from the start of the century onwards shows that the fuel economy improved steadily over the year.[32] The 2010 target value for PC average fuel efficiency of 14.8 km per liter was already exceeded in 2008 and by 2012 the 2020 target of 20.3 km/l was almost reached.

Japan's fuel efficiency standards are set under the so-called Top Runner Program. Under this system, targets are based on the values achieved by the most energy-efficient products on the current market. In setting Top Runner Standards for vehicles, the average fuel efficiency performance (FEP) of vehicles currently on the market is taken as the starting point and the average is raised to the level of top-runner vehicles. Subsequently, new target FEPs are determined by considering the potential progress of technology expected to be widely introduced in the near future. Thus, fuel efficiency standards have been raised continuously.

The 2015 fuel efficiency standard has been set at 18.6 km/l for PCs in the 10-15 mode test cycle, with provisions for increasing stringency. Worldwide, only Japan has fuel efficiency standards for each car category ranging from light to heavy motor vehicles.

1.4.4   China

Vehicle exhaust has become a major source of air pollutants in China, as it has become one of the fastest growing fleets of motor vehicles in the world. Since the late 1970s, the number of vehicles in China has increased dramatically and vehicle pollution is estimated to be the cause of

- about 70% of the smog that casts a haze over certain Chinese cities. Chinese can check the levels of ozone and particulate material (PM2.5) in Beijing, Chengdu, Guangzhou, Shanghai, and Sheyang every hour at the Twitter site https://twitter.com/BeijingAir.
- about 87% of carbon monoxide and 92% of nitrogen oxides.

In some cities, vehicle exhaust emissions have become the prime air pollution source, such as in Beijing. These developments have put emphasis and speed on the development and implementation of stringent emissions standards in order to improve air quality. As a result the nationwide standard China 6 was agreed, one of the world's most stringent standards, with an envisaged nationwide implementation date of 2020 (China 6a) and 2023 (China 6b), respectively.

Regulating Bodies. Vehicle emission standards are issued jointly by the Ministry of Environmental Protection (MEP) and the Standardization Administration of China (SAC), the former being responsible for the development, drafting and approving of the standards. In addition to the nationwide responsible MPE, regional and local environmental protection bureaus exist.

Once a nationwide standard is issued, cities and regions in China may implement it in advance of the nation timetable once the State council gives approval for this measure. Beijing has traditionally been China's leader in implementing

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32 http://www.theicct.org/sites/default/files/info-tools/pvstds/Japan_PVstds-facts_jan2015.pdf – (accessed 21 March 2021).

emissions standards. Its example has been followed by other major cities like Shanghai or Guangzhou with similar air quality issues.

Emission Standards. Modern, nationwide vehicle emission control programs date back in China as long as to the late 1990s. First goals were the elimination of leaded gasoline, followed by the implementation of progressively stringent tailpipe emission standards for LDVs. China's emissions standards were nearly identical to Europe's in terms of its limit values and test cycles. LDV categories were also based on the European classification, with some deviations. As motor cycles form an important part of the vehicle population in China, separate standards exist and can be found at[33].

An overview on the implementation dates is given in Table 1.13. It has to be noted that the nationwide date generally refers to new type approvals. First registration of existing vehicle models is typically allowed for one more year. For China 5 and 6 standards, a single implementation date for new type approvals and first registration will be provided.

Deviations from the national implementation roadmap are possible. Beijing introduced the Euro IV standard in advance on 1 January 2008, and became the first city in mainland China to adopt this standard. It has also had a ban on the sale and registration of light-duty diesel vehicles since 2000.

Table 1.13 Timeline for nationwide implementation of light duty emission standards.

Stage	Standard	Implementation Date (type approval)	Implementation Date (all vehicle sales and registrations)
China l	**GB 18352.1-2001**	1 Jan 2000 (Type 1) 1 Jan 2001 (Type 2)	1 Jul 2000 (Type 1) 1 Oct 2001 (Type 2)
China 2	**GB18352.2-2001**	1 Jul 2004 (Type 1) 1 Jul 2005 (Type 2)	1 Jul 2005 (Type 1) 1 Jul 2006 (Type 2)
China 3	**GB 18352.3-2005**	1 Jul 2007 (no EOBD req.) 1 Jul 2008 (EOBD req. for Type 1) 1 Jul 2010 (EOBD req. for all others)	1 Jul 2008 (no EOBD req.) 1 Jul 2009 (EOBD req. for Type 1) 1 Jul 2011 (EOBD req. for all others)
China 4		1 Jul 2010	1 July 2011 (gasoline) 1 Jul 2013 (diesel)[a]
China 5	**GB 183525-2013**	1 Jan 2016 (gasoline) 1 Jan 2017 (diesel)	1 Jan 2017 (gasoline)[b] 1 Jan 2018 (diesel)[b]
China 6	**GB 18352 5-2016**	n/a	1 Jul 2020 (China 6a) 1 Jul 2023 (China 6b)

a) This date represents a two-year delay from the date specified in the original standard.
b) The implementation timeline of China 5 light-duty gasoline vehicle and light-duty diesel bus in the eastern 11 provinces (Beijing, Tianjin. Hebei. Liaoning. Shanghai, Jiangsu. Zhejiang. Fujian. Shandong. Guangdong and Hainan) is 1 April 2016

Source: China – Light Duty Emissions, Licensed under CC BY 3.0 SA.

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33 https://www.transportpolicy.net/region/asia/china/ – (accessed 19 March 2021).

The most advanced China 6 standard will take effect beginning on 1 July 2020. Unlike the previous standard phases, which closely followed the European emission standards, the China 6 standard combines best practices from both European and US regulatory requirements in addition to creating its own. The standards are fuel neutral, i.e. they apply to gasoline and diesel vehicles.

Key features are:

- China 6b targets a reduction of 50% for hydrocarbons, 40% for NO_x, and 33% for PM over Euro 6 levels.
- RDE requirements will apply as of July 2023 (the China 6b implementation date). Cold starts will be excluded and only the moving average window data evaluation method will be used.
- The WLTP testing cycle will be mandatory.
- In contrast to Europe or the USA an N_2O limit is introduced.
- China 6 OBD requirements are based on the California OBD II regulations.
- The evaporative emission limit (SHED) is tightened from 2.0 g to 0.70 g for type 1 and 2 category I vehicles, 0.90 g for type 2, category II and 1.20 g for type 2 category III vehicles.
- Emissions durability requirements have been enhanced from 100 000 to 160 000 km.
- Start of a multi-component compliance program involving agency- and manufacturer-run emission tests during pre-production, production and in-use stages and China's first emission warranty and defect reporting program.

Separately, the government plans to raise the fuel efficiency target level to 20 km per liter of fuel, 40% tougher than the current level, by 2020. In order to meet this target, it is expected that car manufacturers will have to sell more electric and compact cars. Also the utilization of energy saving technologies like idle stop-start is expected to contribute and exceed over 50%. Figure 1.12 provides an overview on the market penetration of different energy conservation technologies in the Chinese market [44].

China relies on a twofold strategy of "restrictive and instructive policy." The former regulates and reduces emissions and fuel consumption of vehicles, whereas the latter encourages consumers to buy energy-saving vehicles via tax incentives, in particular small displacement ones (max. 1.6 l engine).

A recent study concluded the fleet fuel consumption goal of 5 l/100 km set for 2020 will be impossible to reach based only on the promotion of either energy-saving technology of conventional vehicles or on new energy vehicles [44]. Only a coordinated approach of both seems to be the solution as can be seen in Figure 1.12.

1.5 Well-to-Wheel Analysis of Alternative Fuels

For each of the described alternative fuel options, a detailed well-to-wheel (WTW) analysis is provided in the individual chapter relating to a specific option.
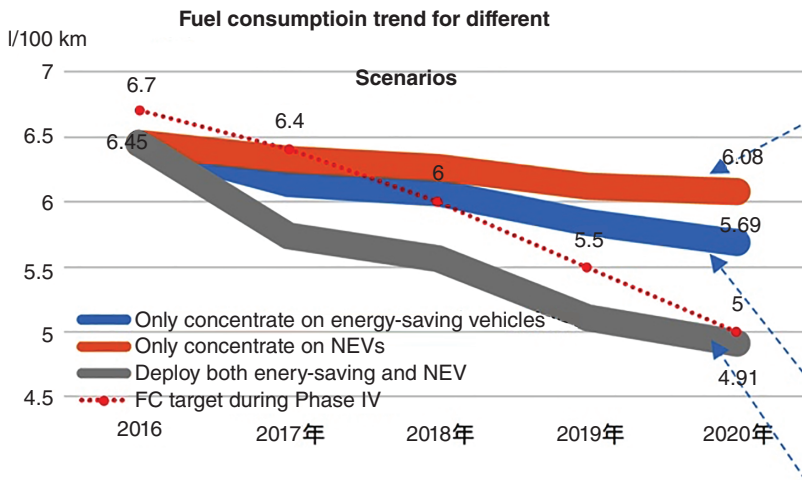
Figure 1.12 Fuel consumption trends for different scenarios from [44].

This section seeks to provide a short introduction in the topic of this type of analysis, helping to clarify the different approaches existing and specifically detailing the methodology used in the most recent WTW and TTW JRC studies, as these are primarily used for the assessments in this book (JRC = European Commission Joint Research Centre, Institute for Energy and Transport). These studies represent a sound and broadly accepted scientific reference. Moreover they cover a broad span of different fuel alternatives, all evaluated with the same approach, using the same fuel pathways, vehicle configuration, and simulation methodology, which makes the comparison of results very relevant and meaningful.

A number of different terms are frequently used in the context of assessing the "environmental impact" of a specific fuel option: WTW as well as TTW, life cycle analysis, ecobalance or cradle-to-grave analysis.

1.5.1 Life-cycle Assessment

Life cycle assessment (LCA) (also known as life-cycle analysis, ecobalance and cradle-to-grave analysis) is "a technique to assess environmental impacts associated with all the stages of a product's life from raw material extraction through materials processing, manufacture, distribution, use, repair, maintenance and disposal or recycling" [46].

Cradle-to-grave is the full life cycle assessment from resource extraction (="cradle") to the use and disposal phase (="grave"), i.e. for fossil fuels it would embrace the full life cycle from drilling to combustion in an engine.

In contrast to this, *cradle-to-gate* is only assessing a part of the product life cycle, restricted to the resource extraction (*cradle*) to the factory gate (refinery, for example). In the context of fuels it would not deal with for example tail-pipe emissions. It would assess the energy and GHG emissions associated with oil drilling to the production of fuel, leaving the refinery gate.

Cradle-to-cradle on the other hand is a specific variant of the cradle-to-grave assessment, where the last step in the product cycle is a recycling process (going back to cradle, as the term suggest). This assessment methodology extends the environmental impact of products by beyond production, operation and disposal practices. For the assessment of alternative fuel options, it is not relevant, as the fuels cannot be recycled after use.

1.5.2 Well-to-Wheel

WTW is the specific LCA used for transport fuels and vehicles. It seeks to estimate GHG emissions and energy efficiency for a specific fuel option, typically also with a defined time horizon. WTW differs from the approach taken by a life cycle analysis (LCA), as it does not consider energy and emissions involved in the building of facilities and the vehicles or end of life aspects. WTW breaks down the overall life cycle in distinct steps, an example is shown in Figure 1.13.

The first step is the "well," i.e. would deal with the oil drilling or with the growing of a specific crop. Followed by the production of a road fuel (e.g. in a refinery), delivery to depot(s), filling station and final combustion in the engine of a vehicle. The analysis includes all transportation steps necessary, from raw materials to the final fuel.

WTW is assessing a fuel until it is burned in an engine. It is composed of two complimenting parts: WTT and TTW. The former estimates energy use and GHG emissions for a fuel option until it reaches the vehicle (i.e. fill-up of vehicle
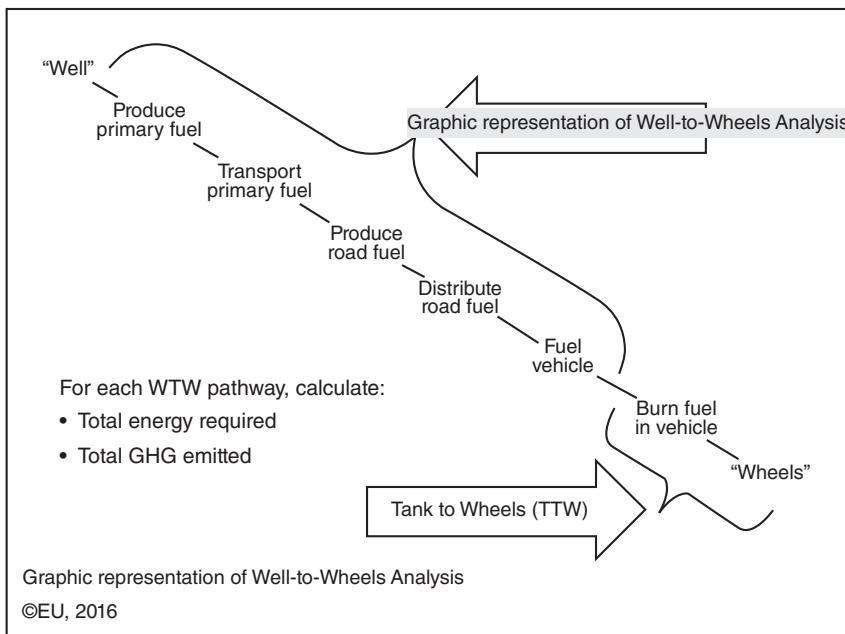
Figure 1.13 Steps of the WTW and TTW analysis. Source: Ref. [47]. Licensed under CC BY 4.0.

at a filling station). The TTW part then evaluates the impact of the combustion in the vehicle.

Results derived from a WTW analysis can differ from the ones obtained with the LCA methodology as the latter is taking into account more emission sources. For example, LCA also includes the GHG and energy necessary for the construction of the production units and the manufacturing of the vehicles. The differences can be significant. For example, when assessing the GHG emissions of a battery electric vehicle (BEV) in comparison to a conventional ICE vehicle, the WTW (accounting only the GHG for manufacturing the fuels) estimates that an electric vehicle can save 50–60% of GHG [45]. A hybrid LCA-WTW method, considering also the GHG due to the manufacturing and the end of life of the battery, estimates much lower GHG emission savings (c. 10–13% lower, compared with the WTW) [48].

It is worth noting that neither LCA nor WTW seek to estimate "costs to society" such as health, social or other speculative cost areas are made. LCA, in contrast to WTW, does often evaluate sustainability aspects like land use change (LUC) or water footprint.

1.5.3 Boundary Conditions of the JRC Study [46, 49, 50]

The authors of the study deliberately chose to favor WTW over LCA methodology, arguing that although LCA follows a standardized ISO methodology, it is more complex to implement. This is particularly difficult for new processes, where system boundaries need to be defined and data describing (a vast amount of) LCA variables can be lacking. The experts also state that US studies support the view and that the impact of vehicle production and end of lifetime make a significant contribution – but as it is a fairly constant contribution, it supports the validity of the WTW approach.

The WTW methodology makes the implicit assumption that energy use and GHG emissions are independent on where they occur. As GHG act globally this assumption is clearly true. The situation is nonetheless different for parameters like air pollution and water use, which depend more on local conditions and effects. The JRC study does discuss these effects and quantifies them where possible, but did not include these figures and considerations in the outcome, due to the large uncertainty associated.

Biofuels represent a very challenging topic. Due to the complex agricultural part, it is particularly difficult to produce reliable GHG and energy balances. In addition to the impact of fossil energy used in producing and processing the crop, GHG emissions are emitted over the growing period as nitrous oxide (N_2O), a gas with 298 times the GHG potency of CO_2, as nitrogen from fertilizer and natural sources is broken down in the soil. N_2O emissions depend on soil type, fertilizer addition, type of crop, and also weather, so they are difficult to estimate with accuracy. A detailed description of the model used for the estimate of N_2O emissions can be found at [46]. It does account, for example, of the fact that crops come from inside and outside the EU. Soils emit some N_2O even if they are not farmed. These can be quite significant, especially for organic soils. In previous versions of this study, the N_2O emissions were subtracted for a reference case of unfertilized

grassland to obtain the N_2O emissions directly attributable to biofuel production. With the new GNOC modeling tool used in the current version, a reference case was no longer needed, because the emissions from an unfertilized control plot are subtracted internal to the model.

The second factor affecting agricultural production is LUC. LUC refers to effects arising from the growing of the biofuel crop on the land, e.g. crop cultivation may directly change the soil carbon reservoir, when forest or grassland is converted to agricultural use. In some cases the carbon store may increase where a perennial crop replaces annual arable crops. Such DLUC can take several years to decades to reach equilibrium and it should be noted that the effects can be very large. Changes in land use may also be affected indirectly by biofuels through the expansion of croplands to replace the land lost to food production. GHG emissions may result from removal of existing forests or other vegetation and changes in the soil carbon reservoir of new land changed for agricultural use. These effects are referred to as Indirect Land Use Change (ILUC). LUC is discussed in more detail in Table 9.2. Both direct land use change (DLUC) and indirect land use change (ILUC) can be important in understanding the impact of biofuels, but they are difficult to estimate and still the subject of debate and research and were not included in the study for these reasons.

The JRC study provides an assessment of a wide range of alternative fuel options (fully commercialized or at least available on large-scale), and Table 1.14 provides a concise summary.

For the vehicle calculations, the JRC study used a common vehicle platform representing the most widespread European segment of passenger vehicles. Full details on vehicle data, performance and simulation can be found in [46]. As can be seen from Table 1.15, a number of powertrain options were assessed. As for production plants it was assumes that all vehicles meet current and incoming future regulations. Emissions ware based on the European NEDC test cycle as the WLTP procedure was not yet fully defined at the time the study was deployed.

All energy contents used exclude the heat generated after the combustion process by the condensation of water vapor arising from the hydrogen content of the material. For materials containing water (crops, animal feed, wood or manure) is used, the lower heating value (LHV) of the dry-matter content of the material. Some other studies have subtracted from this LHV also the heat needed to evaporate the water content. In the case of wet material, like animal slurry, the heat content then apparently becomes negative, and one cannot apply the principle of conservation of heat in a process (for example, wood apparently gains energy from nowhere as it dries out during storage).

Many pathways considered in the study include long-distance shipping of gases or liquids. In this case published data for a type of ship consistent with the length of the envisaged trip and the material being carried was used. Such ships normally return empty and the corresponding fuel consumption was accounted for as well.

1.5.4   Summary of Results of the JRC Study

In principle, the study concludes that a shift to renewable/low fossil carbon routes may offer a significant GHG reduction potential but generally requires

Primary energy resources and automotive fuels.

Table 1.14 Overview of alternative fuel options and pathways considered in the JRC study.

Resource	Fuel	Gasoline, diesel (2010 quality)	CNG/ CBG/ SNG	LPG	Hydrogen (comp., liquid)	Synthetic diesel	DME	Ethanol	MT/ETBE	FAME/FAEE	HVO	Methanol	Electricity	Heat
Crude oil		X											X[e]	X[f]
Coal					X[a]	X[a]	X					X	X	X
Natural gas	Piped		X		X[a]	X	X					X	X	X
	Remote		X[a]		X	X[a]	X[a]		X			X	X	X
Shale gas			X											
LPG	Remote[c]			X										
Biomass	Sugar beet							X	X					
	Wheat							X	X					
	Barley/rye							X						
	Maize (Com)		X[b]					X[d]						
	Wheat straw							X						
	Sugar cane							X						
	Rapeseed									X	X			
	Sunflower									X	X			
	Soy beans									X	X			
	Palm fruit									X	X			
	Woody waste				X									X
	Farmed wood				X	X		X				X	X	X
	Waste veg oils									X	X			
	Tallow									X	X			
	Organic waste		X[b]									X	X	X
	Black liquor				X	X	X					X	X	
Wind			X		X								X	
Nuclear													X	
Electricity					X									

a) With/without CCS.
b) Biogas.
c) Associated with natural gas production.
d) EU and US sources.
e) Heavy Fuel Oil.
f) Heating Oil.

Source: Ref. [46]. Licensed under CC BY 4.0.

Table 1.15 Overview of automotive fuels and vehicle combinations.

Automotive fuels and powertrain combinations.												
Powertrain / Fuel	PISI	DISI	DICI	Hybrid DISI	Hybrid DICI	PHEV20 DISI	REEV80 SI	PHEV20 DICI	REEV80 CI[a]	BEV	FCEV	REEV80 FC[b]
Gasoline												
Gasoline E10 (market blend)												
Gasoline E20 (high RON)												
Diesel												
Diesel B7 (market blend)												
LPG												
CNG												
E85												
MTBE												
ETBE												
FAME												
DME												
Syndiesel												
HVO												
Electricity												
Compressed hydrogen												
Cryo-compressed hydrogen												

All configurations modeled for both 2010 and 2020+ (except when stated otherwise).

Color coding.

□ Modeled in detail with the vehicle simulation tool.

□ Exceptions:

REEV80 CI[a] and REEV80 FC[b] only modeled for 2020.

REEV80 CI[a] modeled for two different layouts.

□ Derived from simulations using the relevant fuel properties.

The vehicle/powertrain configurations are:

- **PISI/DISI:** port injection/direct injection spark ignited engine
- **DICI:** direct injection compression ignited engine
- **PHEV20:** plug-in hybrid vehicle with an electric driving range of 20 km (NEDC)
- **REEV80:** range extended electric vehicle with an electric driving range of 80 km (NEDC)
- **BEV:** battery electric vehicle
- **FCEV:** fuel cell electric vehicle
- **REEV80 FC:** range extended fuel cell vehicle with an electric driving range of 80 km (NEDC) and a Fuel Cell as a Range Extender.

Source: Ref. [46]. Licensed under CC BY 4.0.

more energy (and also costs). The fact that there is not a *single fuel pathway offers a short term route to high volumes of "low carbon" fuel will create more diversity,* i.e.

- Contributions from a number of technologies/routes will be needed.
- A wider variety of fuels may be expected on the market.
- Blends with conventional fuels and niche applications should be considered if they can produce significant GHG reductions at reasonable cost.
- Optimum use of renewable energy sources such as biomass and wind requires consideration of the overall energy demand including stationary applications.
- Large-scale production of synthetic fuels or hydrogen from coal or gas offers the potential for GHG emissions reduction, but only if CO_2 can be captured and stored.

1.5.4.1 Alternative Liquid Fuels

A number of alternative liquid fuels (and production schemes) are available – either as neat fuel option (not possible in all cases) or to be used in blends with conventional fuels, in the existing infrastructure and vehicles.

The study concludes that fossil energy and GHG savings of conventionally produced biofuels such as ethanol and biodiesel are critically dependent on manufacturing processes and the fate of co-products. The lowest GHG emissions are obtained when co-products are used for energy production.

The biggest uncertainty in the determination their GHG balance was the determination of nitrous oxide emissions from agriculture. LUC may also have a significant impact on the WTW balance. Due to the difficulties described in the previous section, the authors decided to model only biofuels produced from land already in arable use.

FAME and HVO. When upgrading a vegetable oil to produce an automotive transport fuel, the two established routes of transesterification and hydrotreating are broadly equivalent in terms of GHG emissions. Taking into account the energy contained in the biomass resource, the total energy involved is two to three times higher than the energy involved in making conventional fuels. These pathways are therefore fundamentally inefficient in the way they use biomass.

ETBE can be an alternative to the use ethanol in gasoline. The fossil energy and GHG gains are commensurate with the amount of ethanol used. The study evaluated a variety of processes for the conversion of the biomass into ethanol and states that they have an attractive fossil energy and GHG footprint.

XTL. The evaluation of synthetic diesel produced from natural gas (gas-to-liquid [GTL]) or coal (coal to liquid [CTL]) revealed that GHG emissions from GTL diesel are slightly higher than those of conventional diesel, while those from CTL diesel are considerably higher. The relatively new processes for the manufacturing of biomass to liquid (BTL), using biomass as starting point, offer lower overall GHG emissions, but suffer still from high energy use. Such advanced processes have the potential to save substantially more GHG emissions than current bio-fuel options, which is one of the major conclusions of the authors.

Dimethyl ether (DME). DME can be produced from natural gas or biomass with lower energy use and GHG emissions results than other GTL or BTL fuels. DME is judged by the study as the sole product, for which the yield of fuel for use for diesel engines is high. It should be noted that the use of DME as automotive fuel would require modified vehicles and infrastructure similar to LPG.

The evaluation of the "black liquor" route yielded higher wood conversion efficiency compared to direct gasification in those situations where it can be used and is particularly favorable in the case of DME.

1.5.4.2 Alternative Gaseous Fuels

As of today the WTW GHG emissions for *CNG* have been determined to be between gasoline and diesel. Beyond 2020, greater engine efficiency gains are predicted for CNG vehicles and WTW GHG emissions are expected to approach those of diesel. WTW energy use will remain higher than for gasoline. *LPG* provides a small WTW GHG emissions saving compared with gasoline and diesel.

The authors highlight in the study that the origin of the natural gas and the supply pathway are critical to the overall WTW energy and GHG balance. The production of *biogas*, particularly from waste materials, was found to have a very low GHG impact, independent of whether the biogas is used to fuel cars or produce electricity. Considering the production of synthetic gas (SNG) from wind electricity and captured CO_2 (from CCS) revealed the low GHG emissions but also the energy need of the processes.

All options discussed in this section would require dedicated vehicles as well as infrastructure for supply and distribution, including re-fueling.

1.5.4.3 Electricity and Hydrogen

Externally Charged Vehicles and Fuels. The study considered a wide range of options for vehicles, including plug-in hybrids. The authors conclude that the electric propulsion on the vehicles is efficient. Nonetheless, the overall energy use and GHG emissions depend critically on the source of the electricity used, i.e. where electricity is produced with lower GHG emissions, electrified vehicles give lower GHG emissions than conventional ICEs, with BEVs giving the lowest emissions. Where electricity production produces high levels of GHG emissions, the plug-in hybrid (PHEV)20 configuration emits less GHG than the other electric vehicles (XEVs). This is because a plug-in hybrid involves less electric driving than the BEV and range extended electric vehicles (REEV). The differences in performance between PHEV and REEV technologies are primarily a function of the different assumed electric range (20 km vs. 80 km) rather than a differentiator between the technologies themselves.

Fuel Cell Vehicles (FCEV) and Hydrogen. Again a variety of potential hydrogen production routes exist and the results are critically dependent on the pathway selected. The developments in fuel cell system, tank, and vehicle technologies are expected to allow fuel-cell vehicles to become more efficient in the 2020+ timeframe and increase their efficiency advantage over conventional vehicles. In case hydrogen is produced from natural gas the authors were able to demonstrate in the earlier version of the report that WTW GHG emissions savings can only be

achieved if hydrogen is used in FCEV. When moving to the 2020+ horizon, this option is considered to have the potential to produce half the GHG emissions of a gasoline vehicle.

Electrolysis using EU-mix electricity or electricity from natural gas results in GHG emissions two times higher than producing hydrogen directly from natural gas and gives no benefit compared with a gasoline vehicle. Hydrogen from nonfossil sources (biomass, wind, nuclear) offers low overall GHG emissions. For hydrogen as a transportation fuel virtually all GHG emissions occur in the WTT portion, making it particularly attractive for CO_2 capture and storage. Using hydrogen as a cryo-compressed fuel increases GHG emissions by about 10% compared with the compressed gaseous form with 70 MPa.

1.5.4.4   2020+ Horizon

Figure 1.14 provides a summary of the TTW simulations, illustrating expected changes resulting from the development of vehicle technology. As can be seen, developments in diesel and gasoline vehicle technology will yield significant improvements, contributing to a significant reduction in the TTW part of the equation. Hybridization of the conventional engine technologies is also expected to provide further energy and GHG emission benefits. The efficiency gap between spark ignition and compression ignition vehicles is forecasted to be narrowing, especially for hybrid versions using methane (CNG, CBG, SNG) and LPG fuels.
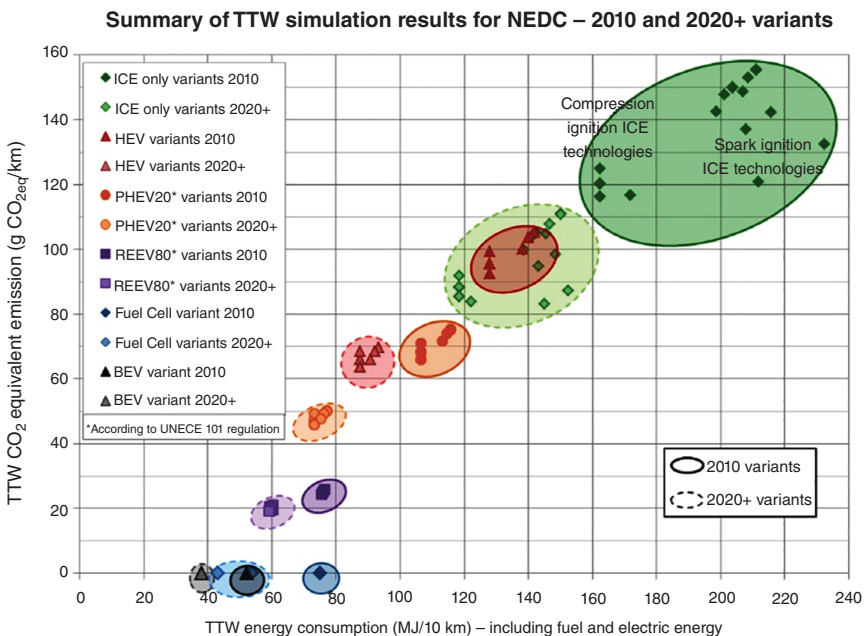

Figure 1.14 Summary of TTW simulation Results for NEDC for 2010 and 2020+ variants. ICE = internal combustion engines; HEV = hybrid electric vehicles; PHEV20 = plug-in hybrid vehicles; REE80 = range extended electric vehicles; BEV = battery electric vehicles. Source: Ref. [46]. Licensed under CC BY 4.0.

Moving to the 2020+ horizon, it is expected that CNG as transportation fuel will only provide small improvements, because its global GHG balance is close to that of the gasoline and diesel fuels it would replace.

The improvements expected in FCEV efficiency, production of hydrogen from natural gas by reforming and its use in FCEV are projected to have the potential to save as much GHG emission as substituting coal by natural gas in power generation. Using farmed wood to produce hydrogen by reforming will save as much GHG emission per hectare of land as using the wood to produce electricity in place of coal. It is expected that it will also be able to save more GHG emissions per hectare than producing conventional or advanced biofuels.

Considering wind electricity for transport fuels (via hydrogen production), in combination with use in FCEV, leads to the assumption that this pathway will be more efficient than the application of synthetic diesel or methane in ICE-based vehicles. According to the study using wind electricity to produce hydrogen and using it in FCEV saves slightly less GHG emissions than substituting NG CCGT electricity. Using wind electricity as a substitute for coal electricity is judged to be the most efficient option for GHG savings by the JRC study.

Figure 1.15 summarizes the WTW results in terms of energy use and GHG emissions for the non-hydrogen pathways. The results are shown for 2020+ vehicles, except for conventional gasoline and diesel where the 2010 results from the reference baseline: the dotted lines mark the performance of a 2010 gasoline vehicle. The energy figures include all energy, both fossil and renewable. In general, those options that have low GHG emissions have high total energy use. Although GHG emissions are of prime concern today, energy conservation and efficient use of energy resources are and will remain important goals.
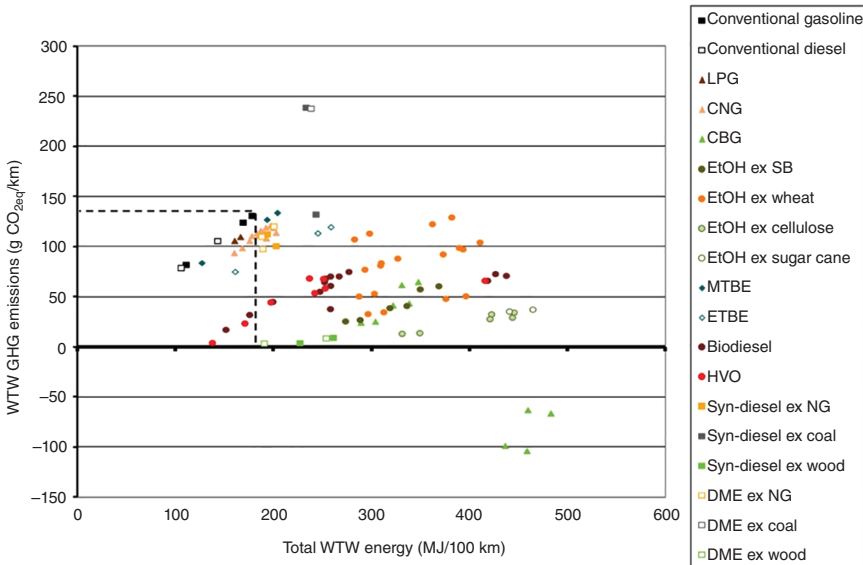
Figure 1.15 Energy use and GHG emissions for non-hydrogen pathways (2020+ vehicles). Source: Ref. [46]. Licensed under CC BY 4.0.
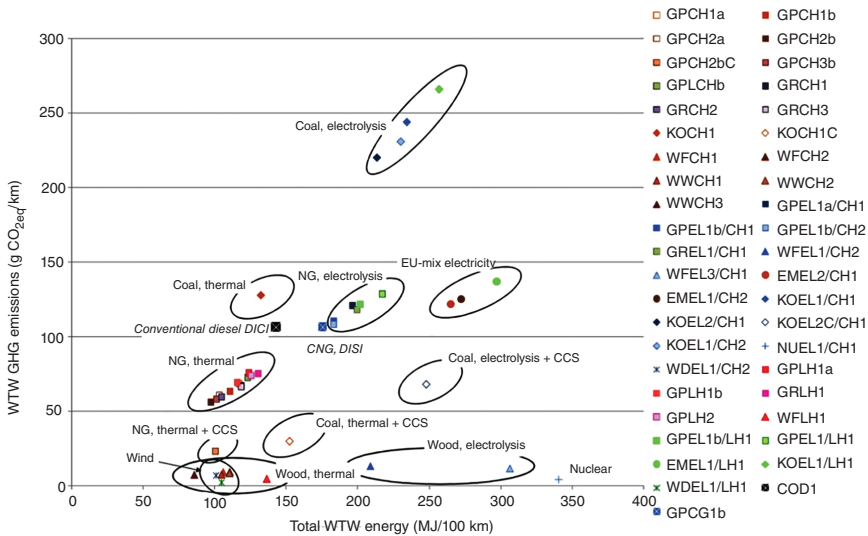
Figure 1.16 Energy use and GHG emissions for hydrogen pathways.

A similar overview for hydrogen pathways is given in Figure 1.16. The graph illustrates the diversity of routes and the wide range of energy consumption and GHG emissions (the data points for conventional diesel in a 2020 direct injection compression ignited engine (DICI) vehicle (diesel engine with direct injection) and CNG in a 2020 direct injection spark ignited engine (DISI) vehicle [spark ignition vehicle with direct injection] are also shown for reference).

From an energy and GHG point of view, there are favorable and unfavorable ways of producing hydrogen. GHG reduction tends to be at the cost of extra energy although the high efficiency of the fuel cells can compensate for the high hydrogen production energy. The electrolysis routes whereby primary energy is first turned into electricity and then electricity into hydrogen are energy intensive. Even when combined with an efficient converter such as a fuel cell, the energy consumption remains higher than for conventional fuels and powertrains. Pathways involving liquid hydrogen are slightly less favorable than the equivalent with compressed hydrogen. More detailed information on all pathways is provided in [46, 49].

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