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ORIGINAL PAPER

Cars and ground-level ozone: how do fuels compare?

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Abstract
Introduction An important question for policy-makers is how the main automotive fuels – diesel, gasoline, LPG (and increasingly, electricity) – compare in terms of ground-level ozone formation.
Methods Based on recent, equivalent emissions data, the study compares ozone formation on a per-kilometre basis of the main fuels: gasoline, diesel, liquefied petroleum gas and electricity (the latter in the United Kingdom).
Results Considering tailpipe emissions only, gasoline's and LPG's per-kilometre ozone impact is 44–88% of diesel's, while LPG's is slightly lower than gasoline's. If fuel production and tailpipe emissions are added together, the liquid fuels generate 48–80% of electricity's impact, i.e. the electric car's ozone impact is highest. The liquids' ozone-impact rankings are the same as for tailpipe only, from most to least: diesel, gasoline, LPG.
Conclusions Changing the fuel/energy type of a passenger car changes its emission inventory, so this could be a useful policy in combating ozone, i.e. governments could encourage some fuels/energies and discourage others. Based on the results shown above, a priority ranking of the main types, from best to worst in the United Kingdom, is: LPG, gasoline, diesel and battery electric. For electric, this ranking will vary in other regions, depending on the emissions of the power-generation grid. For the liquid fuels, the rankings are valid for Europe and North America in general. Impact assessment of ozone is complex, because the chemistry of its formation is complex. This complexity is only partially incorporated in existing impact assessment methods.

Keywords Ground-level ozone · Road transport · Gasoline · Diesel · LPG · Electricity

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1 The problem and puzzle of ground-level ozone

At ground level, ozone is a destructive pollutant that has high priority in environmental regulations. Unlike most priority pollutants, ozone is not emitted directly from fuel combustion, but is synthesised in the atmosphere from combustion and other emissions, via a complex web of chemical reactions. The results are at times surprising.

1.1 At ground, a destructive pollutant. On high, a cosmic shield

Ozone (O_3) is both a toxicant and a greenhouse gas. At ground level, it is a noxious pollutant. In the higher atmosphere, it is beneficial to life.

1.1.1 A bleaching agent

The pale-blue gas is classified as a bleaching agent. Inhalation of this bleach is harmful to animals and plants.

For humans, elevated levels of ambient ozone cause eye and respiratory irritation, indicated by symptoms such as cough, throat dryness, eye and chest discomfort, thoracic pain, and headache [1]. Lung function is also reduced, i.e. the lungs are less able to: move air in and out, and to put oxygen into and remove carbon dioxide from the blood. Ozone is also suspected to aggravate cardiovascular function, but this has not been proven conclusively.

Short-term (acute) effects of ozone exposure are undisputed. High ozone concentrations cause the above afflictions, which lead to increased hospital admissions and sometimes to death. A summary of 20 European epidemiology studies suggests that for every 5 ppbv increase in ambient ozone concentration, there is a 0.22% increase in daily deaths [2]. Long-term (chronic) effects of ozone exposure are less clear. Some studies suggest that it causes a permanent reduction in lung function, or that it contributes to heart disease and cancer. These claims are inconclusive [3], but not disproven. As [3] point out, the inconclusiveness could be due to: insufficient studies; and/or complications in the data. It can be complicated (and sometimes impossible) to isolate the effects of ozone from those of NOx and particles.

Precise deaths rates due to ozone are arguable. As officials of the UK Health Protection Agency point out, estimates of premature deaths can range from 100 to 10,000 per year, depending on where the threshold for ozone activity is set. Estimates of hospital admissions also vary by two orders of magnitude, from 230 to 23,000 [4]. If the high-end death rates are assumed, then ozone has a mortality rate that is in the same league as that of particle matter (PM2.5), 29,000 deaths per year, and NO_2, 23,500 deaths per year [5].

Ozone particularly preys upon the weaker people in society: the very young, the old and the poor. They are more susceptible to it, through lower defences and/or higher exposures [6].

1.1.2 A greenhouse gas

Ground-level ozone is a significant contributor to global warming. According to Intergovernmental Panel on Climate Change, IPCC [7], it is the third-largest source after carbon dioxide and methane (not accounting for black carbon), followed by halocarbons and nitrous oxide. Ozone generates about one-fourth the warming of carbon dioxide.

Nonetheless, ozone is not regulated under the UN Framework Convention on Climate Change (Kyoto and Paris Protocols). This is probably because it is a secondary pollutant, and these Protocols focus on primary pollutants. Likewise, ozone appears not to have a recognised global warming potential (GWP). Most greenhouse gases are assigned a GWP by the UNFCCC, but ozone is not, and a literature search has generated no GWP estimates for it. Even a 2016 authoritative estimate listed its GWP as 'NA'.[1] This lack of GWP might be due to ozone's secondary status. It might also be due to ozone's low persistence at ground level. It lasts hours-to-days, unlike the other main GHGs that last for years.

[1] http://cdiac.ornl.gov/pns/current_ghg.html

1.1.3 An ultraviolet-radiation shield

About 90% of earth's ozone is found 20–50 km above the earth's surface, in the stratosphere. At that height, ozone protects life, because it absorbs shortwave radiation from the sun, such as gamma rays, x rays and ultraviolet (UV) rays. These rays can penetrate living cells, causing damage ranging from DNA mutations to sunburn.

As these rays enter the stratosphere, they react with the molecular oxygen there, converting it into two individual oxygen atoms, which then react with other oxygen molecules to make ozone.

$$O_2 + \text{cosmic rays} \rightarrow O + O$$

$$O + O_2 \rightarrow O_3$$

Subsequently, some of the ozone is converted back, i.e.

$$O_3 + \text{cosmic rays} \rightarrow O + O_2$$

This final reaction captures more of the rays than does the oxygen conversion to ozone. Nearly all gamma rays and x rays, and most of the UV rays, do not make it to the earth's surface. They are intercepted mainly by ozone but also by molecular oxygen.

The result of these reactions is an equilibrium of molecular oxygen and ozone, with ozone concentrations of around 1000–10,000 ppbv and molecular oxygen concentrations of around 210 million ppbv. Ironically, although there is 20–200 times more oxygen than ozone in this part of the stratosphere, it is still called the 'ozone layer'.

Ozone depletion (ODP) refers to destruction of stratospheric ozone. Hydrocarbon-halogen compounds (say, fluorocarbons) can float up to the stratosphere, where their free-radical halogens are set free by UV and other high-energy radiation. These highly-reactive halogens convert ozone back to oxygen, leading to ‚holes‘in the ozone concentrations, particularly over the polar regions. This allows significantly higher amounts of UV rays into the lower atmosphere.

1.2 Ozone has regulatory priority

Ozone limits are expressed in two units of concentration. Most of the developed world has regulatory limits for ambient levels of ground-level ozone. The World Health Organisation (WHO) also issues recommended limits. Among air-quality pollutants (i.e. excluding carbon dioxide), ozone has second-highest priority among European regulators, behind the first priority, particle matter (PM).

1.2.1 Two common units for ozone concentration

Within the European Union, ambient-air quality standards for ozone are expressed as mass of ozone per volume of air. Their common units are micrograms per cubic metre, usually written as $\mu g/m^3$. A microgram is 1.0×10^{-6} g (one thousandth of a milligram), and a cubic metre is 1000 l. United States' standards, many health assessments and some monitoring-programmes use either 'parts per million by volume' (ppmv) or 'parts per billion by volume (ppbv). The actual conversion for ozone is 2.1 $\mu g/m^3$ = 1 ppbv.

1.2.2 Ozone ambient air-quality guidelines and limits

For regulators, probably the most important concentration for ozone is the WHO's air quality guideline of 70 $\mu g/m^3$ (35 ppbv) ozone, averaged over 8-h. This is the approximate concentration at which ozone affects human health.

This 35 ppbv value was set in 2004 by the Joint Task Force on the Health Aspects of Air Pollution, an ongoing scientific-collaboration of the WHO and the UN Economic Commission for Europe (UNECE). Actually, the Task Force initially declared a range of 25–35 ppbv, but then settled on a fixed value of 35 ppbv. The Task Force found [8]:

> "…a statistically significant increase in mortality risk estimates…at ozone concentrations above 50–70 $\mu g/m^3$; and according to the opinion of experts present at the seventh meeting of the Task Force, more reliable estimates from atmospheric models were available for concentrations above 70 $\mu g/m^3$."

This 'health-effect threshold' has become anchored in most subsequent policy analyses of ozone. It has led to the adoption of 'SOMO35', an indicator of ozone exposure that is widely used in Europe. SOMO35 is an abbreviation of 'sum of means over 35', an indicator used in health impact assessments of ozone by, among others, the European Environmental Agency and UK Dept. for Environment, Food & Rural Affairs.

EU exceedances of SOMO35 are common, particularly in southern Europe. Exceedances of the UK standards are also common. According to [9], ozone concentrations regularly exceed limits, with rural sites showing considerably more exceedances than urban ones. For 2016, there were 253 UK exceedances (where ozone exceeded 100 $\mu g/m^3$ (50 ppbv) for 8-h more than 10 times). Usually the maximum UK concentrations are up to around 200–300 $\mu g/m^3$ (100–150 ppbv). This is considerable lower than maxima reached in infamous 'ozone pots' such as Los Angeles or Mexico city, where concentrations are known to reach peaks of 1000 $\mu g/m^3$ (500 ppbv).

1.2.3 Ozone's priority in regulation: Relatively high

Among air-quality pollutants, ozone can be ranked as the second-highest priority among European regulators, behind the first priority, particle matter (PM). This has two reasons. One is that the perceived exposure risk of ozone is second only to that of PM. That ranking comes from the European Environment Agency [10], which reports that 95–98% of Europeans in cities were exposed in 2010–12 to excessive ozone concentrations.

The other reason is that, within the transport sector, PM and ozone are seen as the main pieces of 'unfinished business'. Thanks to widespread introduction of tailpipe catalysts, hydrocarbons and CO emissions have been reduced massively, and SOx has been cut greatly by reformulating petrol and diesel. Now it is PM's and ozone's turns to be reduced. For ozone, that job is not straightforward, because it is mostly created indirectly, from other primary pollutants.

1.3 The counterintuitiveness of ozone formation

Ground-level ozone is created by nature, but the primary cause of exceedances is ozone created by human activity. The atmospheric chemistry of ozone – its creation and destruction – is complex, and it leads to two conditions that can be very surprising. One is that certain reductions in ozone precursors can lead to increases in ozone concentrations. The other is that ozone concentrations over time tend to be higher in rural than in urban areas.

1.3.1 Ozone from nature

Ozone formed entirely from natural processes creates a 'background concentration' at ground level that, depending on location and climatic conditions, ranges from 20 to 50 ppbv. Most of this background ozone is created in the ozone layer and then transported down to ground-level by gravity (it is heavier than air) and weather, especially by thunderstorms. Small amounts are also created by conversion of ground-level oxygen to ozone by lightning bolts [11].

1.3.2 Ozone from (mostly) man-made precursors

Most ozone exceedances are caused by photochemical ozone creation (POC). The process is complex [12], but can be boiled down to four main reactions.

It starts with nitrogen dioxide reacting with photons from sunlight to form nitrogen oxide and a single atom of oxygen. This oxygen atom then reacts with molecular oxygen, abundantly present in air, to form ozone.

$$NO_2 + sunlight \rightarrow NO + O$$
$$O + O_2 \rightarrow O_3$$

And it doesn't stop there. Nitrogen oxide then reacts with the ozone to complete the loop, to convert the ozone back into nitrogen dioxide and molecular oxygen.

$$NO + O_3 \rightarrow NO_2 + O_2$$

If this were the extent of it, there would be no net formation of ozone. However, in the presence of hydrocarbons (which have been partially oxidised by oxygen and water vapour), they compete with the ozone to oxidise the NO, allowing ozone to remain undestroyed.[2]

$$NO + \textit{oxidised hydrocarbon} \rightarrow NO_2 + \textit{oxidised hydrocarbon}$$

Other reactions occur, including carbon monoxide and other side products, which in turn kick off other equilibria. Chemical representations of POC typically include 12–15 eqs. [2] [12], but the four presented above are the key ones.

Ultimately, the output of ozone depends on: 1) the relative concentrations of NOx (NO and NO_2) to hydrocarbons; and 2) sunlight. If both main reactants are present in more-of-less stoichiometric quantities, then ozone output takes off. If not, the output is either 'NOx-limited' or 'hydrocarbon-limited'. According to a report for the United States Environmental Protection Agency,[3] at hydrocarbon:NOx weight ratios of <4:1, the area is hydrocarbon limited. At ratios of >15:1, the area is NOx limited. The in-between ratios of 4–15:1 are stoichiometric.

This photochemical creation of ozone can be entirely natural. Nitrogen dioxide is created by natural fires, by lightning bolts and by reactions in the soil. Hydrocarbons are emitted by vegetation, especially trees. In sunny conditions, some forests can easily reach ozone levels of 50 ppbv without a significant man-made contribution [11]. Usually, however, ozone exceedances are due to man-made emissions of NOx and hydrocarbons.

1.3.3 Two surprises

Ozone's 'NOx-limited' or 'hydrocarbon-limited' creation leads to two, related phenomena that run counter to conventional views of pollution.

One surprise is that **reductions in urban NO emissions can lead to increases in ozone concentrations**. In cities such as London, where NOx concentrations have historically been high, cuts in NOx emissions have seen an increase in ozone levels. This stems from a decline in the ozone destruction reaction, sometimes called 'NOx scavenging', whereby nitrogen oxide converts ozone to oxygen [13]

$$NO + O_3 \rightarrow NO_2 + O_2$$

This phenomenon was first confirmed by scientists studying London [14]. Widely confirmed since then, it often is called the 'weekend effect' – because when traffic falls markedly at the weekend, ozone concentrations often rise. This effect is sensitive to hydrocarbon levels. More hydrocarbons will provide more competition for the NO, raising ozone levels even further (the situation is hydrocarbon-limited).

The other surprise is that **ozone concentrations over time tend to be higher in rural than in urban areas**. This is a corollary of the first surprise, which can be seen clearly in UK ozone maps and monitoring reports. Rural areas, especially forested ones, have plentiful hydrocarbons but limited NOx, creating what could be called 'hydrocarbon scavenging' [13]. Places such as the Scottish Highlands, northwestern Wales and southern Cornwall rarely have 'ozone events', where concentrations rise to alarming levels, but over time their average concentrations tend to be the highest in the UK.

2 Ozone precursors in the UK (and northern Europe)

Ozone precursors are mostly man-made and emitted by a variety of economic sectors, with road transport as one of the leading sectors [15]. To show the typical situation in northern Europe, this section profiles ozone-precursor emissions in the United Kingdom – which is a reasonable proxy for the region.

In the UK, ozone precursors are mostly man-made, with some natural emissions as well. Transport is a significant source of ozone precursors today, but less predominant than it was. Diesel road-transport is the leading source of NOx, whilst solvents/aerosols are the leading source of hydrocarbons. Diesel emissions of hydrocarbons might be greater than conventionally assumed.

2.1 Natural (biogenic) emissions

On a global basis, NOx, non-methane hydrocarbons (NMHCs) and methane all have significant sources of natural emissions [2]. Biogenic hydrocarbons greatly outweigh their man-made emissions. NOx and methane are about equally split between biogenic and anthropogenic. Only CO is exclusively man-made. In the UK, however, man-made emissions dominate. Hydrocarbon emissions are 90–95% manmade [16]. Biogenic NOx is no more than 2–3% at the very maximum. Biogenic methane is about 5% of its total emissions [17].

[2] Hydrocarbons emitted to air are naturally oxidised to hydroxyl radicals and other oxidised hydrocarbons.

[3] http://capita.wustl.edu/nescaum/reports/pams94/nepams4.html

2.2 Man-made NOx

Since 1970, annual NOx emissions in the UK have dropped by about two-thirds, from 2677 kilotonnes to just over 1000 kilotonnes. Automotive catalysts were a main cause, as were emission controls on power stations and industrial plants. Passenger cars was the most volatile segment: from 1970 to 1990 it doubled its contribution of NOx from 15% to 30%, and from around 1987 to 1997 it was the leading source. As car catalysts became ubiquitous, its share dropped steadily, down to around 16% today (Fig. 1).

Transport has all along been the predominant source of NOx. This includes 'other transport' – aviation, rail and waterborne shipping – cars, and heavy duty vehicles. In 2014 these accounted for just over half of all NOx emissions (Table 1).

2.3 Man-made hydrocarbons

Since 1970, annual hydrocarbon (HC) emissions in the UK have dropped by nearly two-thirds, from 2030 kilotonnes to 803 kt (Fig. 2). Automotive catalysts were an important cause, so were controls placed on combustors and industrial processes, and so was the substitution of VOC solvents with non- or low-VOC ones. Sector rankings have changed over time. Transport and stationary combustion, which in 1970 contributed nearly half of HCs, now account for only 15%. Transport has made the deepest dive, falling from nearly 600 kt/year to just over 50 kt, under 10% of the total. Solvents have fallen in absolute terms, but risen in relative ones from 29% to around 40%[4].

Today, solvents are the largest contributor (Table 2). Transport ranks behind livestock, food and beverages and natural sources.

2.4 Diesel hydrocarbons might be worse than previously thought

Emissions reported above are based mostly on calculations and inferences, rather than on actual measurements of either emissions or of actual concentrations in the atmosphere. Hydrocarbon emissions from diesel might be seriously underestimated. In an article entitled *'Diesel-related hydrocarbons can dominate gas phase reactive carbon in megacities'*, [18] find that diesel's contribution to urban ozone formation has been underestimated by half. Hydrocarbons from diesel account for 50 + % of all HCs in London's atmosphere, contributing up to 50% of ozone production potential in London.

Why have traditional estimates not captured these diesel hydrocarbons? This is a suitable topic for further research. The author's hypothesis is that longer-chain hydrocarbons, in the range of C_{12} to C_{22}, are not picked up by conventional tailpipe measurements, as is noted by a major testing organisation, the Southwest Research Institute (SwRI). By contrast, the C_1 to C_{12} hydrocarbons emitted by gasoline and LPG tailpipes are picked up. This measurement bias skews the modelled results away from reality.

[4] Sector definitions are not exactly the same in the figure as in the subsequent table, so figures are somewhat different, but still representative.

3 Method: Ozone comparison of road transport fuels

The importance of ozone as a pollutant, and the importance of road transport in causing it, raises an interesting question: how do road transport fuels compare on ozone formation? That question is addressed in the following analysis, which is broken into three parts. First is an inventory of passenger car emissions, by fuel type: gasoline, diesel, liquefied petroleum gas (LPG) and electricity. Second is a comparison of the inventories. Third is a comparative impact assessment of those inventories.

Hybrids were not analysed as such, as these are a combination of two or even three of the above fuels. However, as an anonymous reviewer of this paper has noted, hybridisation adds weight to the vehicle, and hybrid technologies are not standardised, so hybrid results would not be a simple addition of results for electricity and a hydrocarbon fuel. Fuel cell vehicles have not been included, due to a lack of available, relevant data. Both hybrids and fuel cells should be considered in future analyses of this type, if they continue to gain significance in the market.

3.1 Emission inventory: The compilation

The basis of the inventory is a Ford Focus. The inventory covers vehicle tailpipe and fuel production, on a per-kilometre basis. For the vehicle tailpipe inventory, emissions data for 624 Euro-6 cars (nearly identical, except for fuel type) were sourced from Germany's Federal Motor Transport Authority (Kraftfahrtbundesamt, or KBA).

3.1.1 Basis

The basis of the inventory is the Ford Focus. The Focus was chosen, because it is:

- Common, one of Europe's and the world's best-selling,
- Available in all the fuel types studied (except CNG), and
- Well-tested, with plentiful available emissions data.

The only other brand that clearly meets these criteria is the Volkswagen Golf. This is unsuitable, because Volkswagen notoriously has manipulated its emissions testing [15]. The

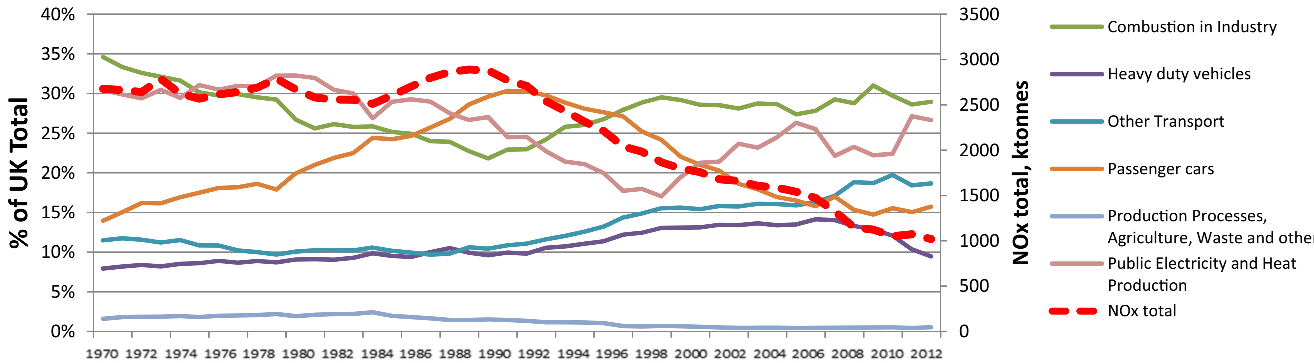

Fig. 1 NOx emissions by sector, UK, 1970–2013. Six sectors in percent, left-hand Y axis. NOx total in kilotonnes, right-hand Y axis. Sector definitions are not exactly the same as those in the subsequent table, so figures are somewhat different, but still representative. (Source: NAEI)

Fiat Panda also might be possible, and it has a CNG version, but there is no electric version.

Compared to other ways of developing an inventory, this choice of a single brand brings a number of advantages in data quality:

- *Commercial relevance* – the Focus is a real car, not a mathematical model such as that used by the European Commission's Joint Research Centre in its emissions analyses [19]. It is among the top-five sellers in the world: 2015 sales were over 800,000.[5] In the past 14 years (the average lifetime of a European car) some 5 million have been sold in Europe, which makes up 2% of the 250 million European passenger cars on the road [20]. During the same period, nearly 3 million were sold in the United States.[6] Precise data are not available, but many hundreds of thousands have been sold in other regions, too.
- *Inherent fairness of comparison* – some models of the Ford Focus come as essentially the same car, just with different fuel types. So this is a comparison of 'apples to apples', for which independent, standardised test data are available.
- *Abundance of available data* – because the Focus sells so well in Europe, a good sample of them are emissions-tested for regulatory approvals. As of March 2016, Germany's Federal Motor Transport Authority had tested 624 Euro 6 models of the Focus that were available in three different fuels. Thousands of earlier versions have been tested previously.
- *Excellent representativeness* – the Focus is a real car, not an estimation or an aggregation of real cars. Temporally, data are available for cars manufactured recently that meet current Euro 6 standards. Technologically, the data represent one of the world's most common cars from one of world's top-ten automakers. Geographically, it is a major seller in Europe and all over the world.
- *Avoidance of over-aggregation* – many studies of automotive emissions define inventories by using emission models such a TREMOVE and COPERT. For many policy analyses these are surely appropriate, but for fuel comparisons such as this they are not. The problem is that such models over-aggregate cars of a fuel type, mostly ignoring important differences such as weight, power rating and performance.

3.1.2 Scope and method

The inventory covers two scopes of emissions: vehicle tailpipe; and fuel production.

These scopes are similar to tank-to-wheel (TTW) and well-to-tank (WTT), except that in this inventory TTW does not include non-combustion emissions (from brakes or tyres) and WTT does not include car manufacturing. These unincluded aspects should be roughly equivalent, given that the cars are

Table 1 NOx emissions by sector, UK, 2014 (Source: NAEI)

Sector	Share of total	
Diesel road transport	21%	
of which		
Trucks/Buses diesel		*11%*
Cars diesel		*10%*
Power generation	17%	
Shipping	16%	
Aircraft	14%	
Gas combustion	3%	
O&G upstream	3%	
Petrol road transport	2%	
of which		
Cars petrol		*2%*
Trucks/Buses petrol		*0%*
Other	24%	
SUM	100%	

[5] https://www.statista.com/statistics/239229/most-sold-car-models-worldwide/
[6] http://www.goodcarbadcar.net/2011/01/ford-focus-sales-figures.html

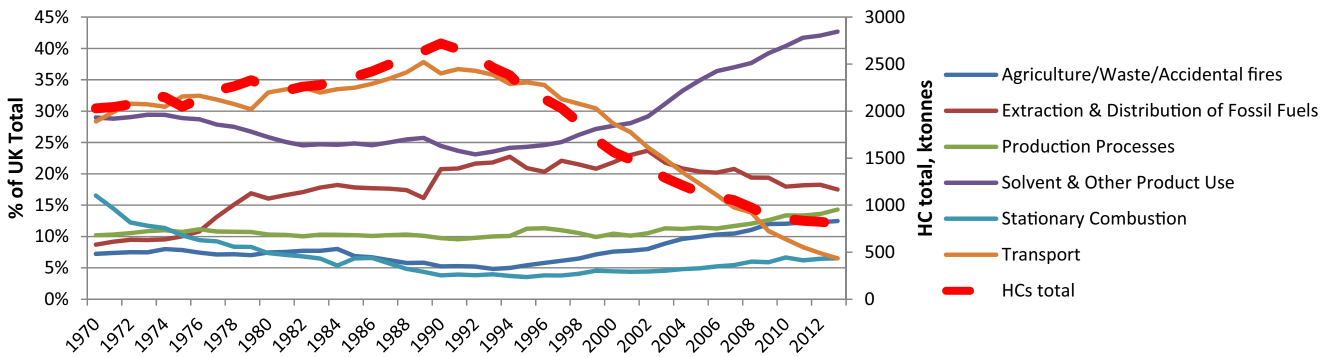

Fig. 2 HC emissions by sector, UK, 1970–2013. Six sectors in percent, left-hand Y axis. HC total in kilotonnes, right-hand Y axis. Sector definitions are not exactly the same as those in the subsequent table, so figures are somewhat different, but still representative. (Source: NAEI)

essentially the same except for their fuel types. The brake and tyre emissions might be slightly higher in the electric car, because it is heavier.

Emissions are normalised (given a functional unit) on a per-kilometre basis.

3.1.3 Data sources

For vehicle tailpipe emissions, the data source is that compiled by Germany's Federal Motor Transport Authority, the KBA. The current KBA database contains emissions data for approximately 400,000 cars and vans, tested from 2003 through to April 2016. It is believed to cover most every car model available in the EU during this time. The KBA database has independent, NEDC test data on 624 Euro-6 Ford Focuses that are available in gasoline, diesel or LPG. These 624 tests results are the core of the inventory.

Table 2 HC emissions by sector, UK, 2014 (Source: NAEI)

Sector	Share of total	
Solvents/Aerosols	38%	
O&G fuel supply	14%	
of which		
O&G upstream		7%
Gas leakage		3%
Refining/bulk dist		2%
Petrol stations		2%
Livestock	11%	
Food/bev	10%	
Natural sources	10%	
Wood heat, domestic	3%	
Offroad machinery	2%	
Road transport, petrol	2%	
Road transport, diesel	1%	
Other	9%	
SUM	91%	

Electric cars of course have no on-board combustion, so tailpipe emissions are not reported by the KBA. For the electric version of the Focus, operating electricity consumption is based on a model developed by [21] that includes all consumptions and efficiencies. The computed efficiency is almost identical to the figure used in a similar study conducted in the UK [22]. The consumption also includes a 'battery-production' footprint of 15 g CO_2/km. Battery production is especially energy and carbon intense [23]. The 15 g estimate comes from a model developed for [24][7] based on [25], which has been modified to run on UK electricity.

Production emissions for electricity are taken from the ecoinvent V3 database: 'Electricity, low voltage {GB}| market for | Alloc Def, U'.

For fuel-production of gasoline, diesel and LPG, two sources are used. One is ecoinvent. The modules used are:

- Petrol, unleaded, at regional storage/RER U (of project Ecoinvent unit processes)| Alloc Def, U (of project Ecoinvent 3 - allocation, default - unit)
- Diesel, low-sulfur {Europe without Switzerland}| market for | Alloc Def, U (of project Ecoinvent 3 - allocation, default - unit)
- Liquefied petroleum gas {RoW}| market for | Alloc Def, U (of project Ecoinvent 3 - allocation, default - unit)

These fuel-production modules from ecoinvent are disputed to some extent (see, for instance, [26]), because of the way those emissions are allocated among products. They are used nonetheless, because no better datasets are available.

The ecoinvent module for LPG production is based solely on refinery production of LPG. Yet according to statistics compiled by market-researchers Argus and the World LP Gas Association, actual LPG tonnage is produced 40% in refineries and 60% from associated oil and gas. So an emission inventory

[7] The module in ecoinvent is named: 'Transport, passenger car, electric {GLO}| processing | Alloc Def, U'.

Table 3 Vehicle tailpipe emissions, Ford Focus, Euro 6, in gasoline, diesel and LPG bi-fuel

	Vehicle dimensions			Pollutant emissions[a]							
Units	*Power* kW	*Displ.* CC	*Weight* kg	*CO* mg/km	*THC* mg/km	*CH_4* mg/km	*NMHC* mg/km	*NOx* mg/km	*PM* mg/km	*PN* Number of particles	*CO_2combi* g/km
Gasoline-LPG Euro 6 limit				1000	100	32	68	60	5	6×10^{11}	NA
Diesel Euro 6 limit				500	HC + Nox 170	NA	NA	80	DI only 5	6×10^{11}	NA
Fuel											
Gasoline	91	1′596	1′826	526.9	53.5	5.5	47.9	26.5	0.0000	0.000	141.5
Diesel	88	1′499	1′900	343.3	35.6	NA	NA	60.2	0.5381	1.127×10^{11}	104.1
LPG	88	1′596	1830	651.3	46.2	4.2	42.0	19.3	0.0000	0.000	123.0

THC = total hydrocarbons. NMHC = non-methane hydrocarbons. CO_2 combi is the carbon dioxide emission for rural and urban driving combined

[a] Particles (PM and PN) and carbon dioxide are not ozone precursors. KBA reports these emissions, which are likely to be of interest to some readers

for associated LPG production was generated, based on [27], and a 60/40 weighted average of the two was used.

3.1.4 Emission inventories of the fuels

The inventories are presented by scope: vehicle tailpipe and fuel production. It is important that they are separated. The vehicle tailpipe inventory is representative for Europe, and indicative for other regions as well. By contrast, the fuel production inventory for electricity is specific only to the UK. The tailpipe inventories (Table 3) are averages for all the vehicles tested by KBA. The cars' power ratings, displacements and weights are nearly equal. All their tailpipes are compliant with Euro 6 limits.

KBA does not report all details of the tested vehicles, so it is difficult to say conclusively what technologies are used in the engines and emission-control systems. Nonetheless, from examining product literature published by Ford, it appears that: the gasoline and LPG versions use port-injection, not direct-injection; and the diesel has a particle filter but does not use selective catalytic reduction ('Adblue') to reduce NOx.

The fuel production inventory for a battery-electric Focus is the inventory of power generation and transmission, normalised to a kilometre of driving (Table 4). Strictly speaking, the car does not exceed Euro 6 limits, because these regulate tailpipes. Nonetheless, seen from well-to-wheel, the car is well above limits on particles, NOx and methane. For fuel-production of gasoline, diesel and LPG, emissions are sometimes significant in relation to those in the tailpipe (Table 3).

3.1.5 Speciation of NOx

It is widely recognised that vehicle NOx emissions of petrol/LPG and diesel cars are significantly different in their NO and NO2 components [28]. Recent estimates from [29] have been applied here (Table 5).

Well-to-tank speciation of NOx is less obvious. Based on examination of ecoinvent models of fuel and electricity supply, the split between industrial combustion and transport is estimated to be 90%/10%. To split the NOx in each of those phases, a literature reference is used for industrial combustion and the diesel vehicle tailpipe speciation is used for transport.

Table 4 Production emissions, Ford Focus, battery-electric, gasoline, diesel and LPG

Pollutant[a]	Units	Battery-electric car[b]	Gasoline	Diesel	LPG	Euro 6 limit
CO_2	g/km	128	22	26	19	NA
PM 10	mg/km	32	9	10	9	5
NOx	mg/km	238	69	69	62	60–80
Hydrocarbons	mg/km	1	42	40	35	90–100
Methane	mg/km	343	70	77	68	32
CO	mg/km	58	28	34	27	500–1000

[a] Particles (PM and PN) and carbon dioxide are not ozone precursors. KBA reports these emissions, which are likely to be of interest to some readers

[b] Assuming UK grid electricity

Table 5 Speciation of NOx emissions

Life-cycle phase	Gasoline	Diesel	LPG	Source
Vehicle tailpipe	3% NO_2	30% NO_2	3% NO_2	[29]
Well-to-tank: industrial combustion	92.5% NO			[30]
Well-to-tank: transport	30% NO_2			[29]

3.2 Inventory comparison of ozone-precursor emissions

From the above-described compilation comes an inventory comparison of the four fuel types (Table 6). NMHCs are not speciated here, because there are so many species, they would dominate the inventory. Carbon dioxide and particles are not ozone-relevant, but they are presented anyway, because they are of interest to many readers.

Table 6 Comparison of Ford Focus emission inventories – gasoline, diesel, LPG and electricity

		Ford Focus Fuel Type			
Pollutant	Unit	Gasoline	Diesel	LPG	Electricity
Tailpipe					
CO2	g/km	141	104	123	0
PM10	mg/km	0	0.538	0	0
NOx	mg/km	27	60	19	0
of which					
NO	mg/km	26	42	19	0
NO_2	mg/km	0.8	18	0.6	0
NMHCs	mg/km	48	36	42	0
Methane	mg/km	6	0	4	0
CO	mg/km	527	343	651	0
Fuel Production					
CO2	g/km	22	26	19	128
PM10	mg/km	9	10	9	32
NOx	mg/km	69	69	62	238
of which					
NO	mg/km	67	48	60	220
NO_2	mg/km	2	21	2	18
NMHCs	mg/km	42	40	35	1
Methane	mg/km	70	77	68	343
CO	mg/km	28	34	27	58
Tailpipe + Fuel Production					
CO2	g/km	163	130	142	128
PM10	mg/km	9	10	9	32
NOx	mg/km	96	129	81	238
of which					
NO	mg/km	93	90	79	220
NO_2	mg/km	3	39	2	18
NMHCs	mg/km	90	76	77	1
Methane	mg/km	75	77	73	343
CO	mg/km	555	377	678	58

4 Results: Impact assessment

Comparison of the inventories does not yield a conclusive finding, other than that the tailpipe emissions of the electric car are much lower than those of the others. (And this was obvious without an explicit comparison.) So an impact assessment (IA) is worthwhile.

First, potential IA methods are considered and chosen. Second, the methods are applied to the inventories and results are compared.

4.1 Review and selection of impact-assessment methods

A survey was conducted of spatial/temporal-independent IA methods [31] [32], and from this survey, a candidate list of methods was assembled (Table 7). These methods use a variety of mid-points and end-points for impact, and they assess damage in various kinds of units. Some differentiate the impact of NO_2 to that of NO, some differentiate the impacts of hydrocarbons by species, and most include the impacts of carbon monoxide and methane.

A number of methods fell out of consideration promptly, because they were missing detail in general or in relation to ozone formation. Others were rejected, because their units are recursive, i.e. expressed in NOx or NMVOCs, which are the same units as the inventories. Others were rejected, because their factors for NOx components and NMVOCs are identical, which ignore the complexity of ozone formation (Section 1.3). The LIME method was knocked out, because it is specific to Japan, which is peripheral to this study. The short list therefore came down to CML, IMPACT 2002+ and TRACI.

CML is problematic, because its factor for NO is strongly negative (−42.7) and its factor for NO2 is weakly positive (2.8). Although CML has been applied in studies of the same type, e.g. [33, 34], these factors generate a perverse results: unless the NO_2/NO ratio exceeds 15.3 (i.e. 42.7 divided by 2.8), NOx emissions will be judged to decrease ozone. Because this ratio is rarely or never exceeded in normal combustion, the CML finding will be that emitting as much NOx as possible is best. IMPACT 2002+ is also problematic, because it rates NO as more toxic than NO_2, which is strongly counter to prevailing opinion [10].

After elimination of the incomplete or unsuitable methods, the remaining methods are TRACI and ReCiPe endpoint, so these were chosen for application to the inventory. (Although ReCiPe endpoint has identical factors for several pollutants (Table 8), this method is recommended by [32, 35].)

Table 7 Candidate list of impact assessment methods for ozone

IA Method	Point	Damage unit	NOx differentiation	HCs	CO	CH_4	Notes
BEES	Mid	g NOx eq 'smog'	NOx and NO2, w equal factors. HCs given a NOxe factor.	Various, but with equal factors	Yes	Yes	Using NOx as the unit is recursive.
CML baseline	Mid	kg ethylene	Yes, NO and NO2	Many, but no NMVOCs	Yes	Yes	Negative value of NO.
CML non-baseline	Mid	kg ethylene	NOx not included, only HCs	Many species	Yes	Yes	4 background levels: 2 low NOx, high NOx and very high NOx
De Leew	Mid	Index TOFP[a]	No	NMVOC only	Yes	Yes	Easy to apply (De Leeuw, 2002)
Eco Scarcity 2013	End	UBP (environmental loading points)	NOx only				
EcoSense Web							Last updated in 2011. Unavailable.
EDIP	End	Person.ppm.h	No	Many, + NMVOC	Yes	Yes	
EPD 2013	Mid	kg ethylene	Yes, NO and NO2	Many	Yes	Yes	This is a copy of CML baseline for ozone, i.e. redundant.
EPS 2000	End	Ozone factors not explicit	No				Impact is morbidity, life expectancy. NO and NO2 are equal impact.
ILCD 2011	Mid	kg NMVOC	NOx, NO and NO2 have equal value = 1	Many, + NMVOC	Yes	Yes	NO and NO2 are equal impact.
IMPACT 2002+	Mid	kg ethylene and kg PM2.5	Yes, for NO, NO2 and NOx	Many, + NMVOC	Resp inorg	Resp org	Factor seems strange – NO is higher impact than NO2 and NOx
LIME	Mid End	kg ethylene or DALY (disability adjusted life year)					For Japan atmosphere and weather
MEEuP							Not really impact assessment
Passant	Mid	kg ethylene	NOx not included	Many, with speciation profiles by source	No	No	Excellent detail, but limited scope
ReCiPe endpoint	End	DALY (disability adjusted life year)	NOx and NO2, but they have same value. Also both = to NMVOC.	Many, + NMVOC	Yes	Yes	Recommended by JRC
ReCiPe midpoint	Mid	kg NMVOC	NOx and NO2, but they have same value. Also both = to NMVOC.	Many, + NMVOC	Yes	Yes	Using NMVOC as the unit is recursive.
TRACI 2.1	Mid	kg O3	NO2 and NOx, so NO could be done by inference.	Huge speciation, also 'VOCs'	Yes	Yes	
USEtox		CTuh, comparative toxic units	NOx, NO and NO2 not included				Not applicable

[a] Trophospheric ozone formation potential

Table 8 Impact assessment methods and factors applied in this study

	Impact assessment method	
	ReCiPe endpoint	*TRACI*
Units	DALYs/kg	kg O_3/kg
Pollutants		
NO	3.9E-08	7.945
NO_2	3.9E-08	16.845
NOx	3.9E-08	24.790
CO	1.78E-09	0.056
CH_4	3.95E-10	0.014
VOCs	NA	3.595
NMHCs	3.9E-08	NA

With both methods, the non-methane hydrocarbons (or VOCs, volatile organic componds, as TRACI labels them) are not speciated. They are considered as an unspeciated mass. They are not speciated, because the emissions reported by [36] cannot be mapped successfully against either method: too many species reported by Passant are not present in TRACI or ReCiPe.

4.2 Comparison of results

Impact assessments were compiled on a tailpipe-only and on a fuel-production-plus-tailpipe basis (Table 9). For tailpipe emissions only, diesel creates a significantly greater ozone impact than the other liquid fuels, and gasoline is slightly higher than LPG. If fuel production and tailpipe emissions are considered jointly, then the electric car has the highest ozone impact, and the liquid fuels keep the same ranking as for tailpipe only.

5 Conclusions and recommendations for further research

Ozone is a significant environmental problem, to which road transport contributes significantly. Changing the fuel/energy type of passenger cars changes their emissions of ozone precursors, so prescriptive fuel/energy policies could be useful in combating ozone. Governments could encourage some fuels/energies and discourage others, in order to reduce ozone levels.

Based on the results shown above, a priority ranking of the main types, from best to worst in the United Kingdom, is: LPG, gasoline, diesel and battery electric. For electric, this ranking will vary in other regions, depending on the emissions of the power-generation grid. For the liquid fuels, the rankings are valid for Europe and North America in general. At a national level, reducing emissions of both NOx and hydrocarbons should be a priority. At the local or regional level, policy detail might differ, particularly depending on degree of urbanisation and levels of natural emissions.

Regulators should recognise that reductions in ozone-precursor emissions can have a bifurcated effect. Reductions in urban NO emissions can lead to periodic spikes in ozone concentration, because 'NOx scavenging' is weakened. Eliminating these ozone spikes will require reductions in hydrocarbon emissions in addition to those of NOx. Also, they should recognise that reducing emissions in urban areas will lead to a knock-on effect of reductions in rural areas, which have the highest average concentrations of ozone.

These are just two of ozone-formation's complexities, which are greater than those of most of the other criteria pollutants. This complexity is only partially incorporated in existing impact assessment methods. Work to incorporate them further is recommended. An initial step would be to map the speciated hydrocarbon emissions as defined by [36] against the factors and units used in TRACI or ReCiPe endpoint, and to introduce more differentiated factors into ReCiPe.

As national energy mixes continue to evolve, so too will sources of ozone precursors. This is also an area suitable for further investigation. It can be speculated that solar and wind energy will likely be low on ozone impact, but biomass might be higher on ozone impact than conventional fossil fuels. Also, whilst electrified automobiles eliminate tailpipe emissions, they do not necessarily reduce overall emissions. Indeed, as this work shows, they can increase such emissions.

Table 9 Ozone impact – gasoline, diesel, LPG and battery electric – Ford Focus

IA Method	Units	Fuel type			
Tailpipe only					
		Gasoline	*Diesel*	*LPG*	
TRACI	mg O_3/km	419	787	346	
ReCiPe endpoint	µDALYs/10,000 km	38	44	36	
Fuel production + Tailpipe					
		Gasoline	*Diesel*	*LPG*	*Electric*
TRACI	mg O_3/km	1′145	1′664	984	2′058
ReCiPe endpoint	µDALYs/10,000 km	48	57	44	95

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