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Optimising air quality co-benefits in a hydrogen economy: a case for hydrogen-specific standards for NO_x emissions

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A global transition to hydrogen fuel offers major opportunities to decarbonise a range of different energy-intensive sectors from large-scale electricity generation through to heating in homes. Hydrogen can be deployed as an energy source in two distinct ways, in electrochemical fuel cells and *via* combustion. Combustion seems likely to be a major pathway given that it requires only incremental technological change. The use of hydrogen is not however without side-effects and the widely claimed benefit that only water is released as a by-product is only accurate when it is used in fuel cells. The burning of hydrogen can lead to the thermal formation of nitrogen oxides (NO_x – the sum of NO + NO_2) *via* a mechanism that also applies to the combustion of fossil fuels. NO_2 is a key air pollutant that is harmful in its own right and is a precursor to other pollutants of concern such as fine particulate matter and ozone. Minimising NO_x as a by-product from hydrogen boilers and engines is possible through control of combustion conditions, but this can lead to reduced power output and performance. After-treatment and removal of NO_x is possible, but this increases cost and complexity in appliances. Combustion applications therefore require optimisation and potentially lower hydrogen-specific emissions standards if the greatest air quality benefits are to derive from a growth in hydrogen use.

Environmental significance

New more demanding hydrogen-specific NO_x emissions standards are required for a range of appliance sectors to ensure that low carbon infrastructure associated with the adoption of hydrogen also delivers a step-change in air quality. Placing hydrogen power within existing air quality regulatory frameworks (for example for Ecodesign Directive or EURO vehicle standards) may see NO_x emissions, efficiency and cost optimised in a way that leads to hydrogen appliances matching current fossil fuel emissions performance, but potentially not improving on them. This would be a major missed opportunity to further reduce NO_x emissions and improve air quality as a co-benefit of net zero commitments and low carbon investment.

Introduction

The use of hydrogen as a replacement for fossil fuels is a significant component of many national decarbonisation strategies and is a major technical component of most Net Zero plans (*e.g.* UK Committee on Climate Change, 2019; A European Green Deal, 2020).[1,2] A very rapid increase in the use of hydrogen as a fuel is anticipated in the 2020s and the associated industries given significant political support (UK Prime Minister Boris Johnson, Financial Times 18th Nov 2020).[3] Two key routes exist to use hydrogen fuel, *via* electrochemical fuel cells (which produce dc electricity directly) or combustion either in thermal boilers or engines. An assumption, at least in a UK context, is that much of this future hydrogen will be combusted – at industrial scale, in domestic settings and in some off-road internal combustion engine applications.

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Hydrogen offers many very valuable features as a fuel; it is energy-dense when in its liquified form, has a wide flammability range, and can be manufactured *via* a variety of routes. In some applications it can be substituted relatively straightforwardly as an alternative fuel, displacing natural gas, gasoline, kerosene and diesel in combustion applications.[4] Hydrogen has potential to fill certain energy supply requirements that may not be easily met using battery electric storage, for example for gas boilers in the home, as a fuel for heavy-duty/long-distance vehicles, off-road machinery, back-up capacity for grid electrical supply and as a fuel for global shipping and aviation.[5] The pre-eminent argument in favour of the use of hydrogen as a fuel is that the *major* waste product formed from its use is water vapour.

Several different routes to manufacturing hydrogen exist with differing degrees of environmental impact.[6–8] So-called grey hydrogen is from the synthetic cracking of carbon–hydrogen bonds in fossil fuels, blue hydrogen which captures carbon emissions as those fossil fuels are cracked, or green hydrogen made from electrolysis of water using renewable electricity. Other routes to hydrogen include the use of excess nuclear-generated electricity and oxygen-free pyrolysis. The full lifecycle and greenhouse gas impacts arising from each process are complex and can require significant emissions management including long-term carbon capture and storage. Van Renseen[9] recently reviewed the role that hydrogen may play in net zero plans for Europe, but as is common, the emphasis was on identifying supply-side environmental and economic challenges, rather than downstream consequences of use.

A 2018 UK Government commissioned review of the atmospheric impacts of hydrogen for heating[10] identified some small possible atmospheric disbenefits through fugitive emissions. These could lead to increased stratospheric water content[11] and climate impacts *via* perturbation of global methane and ozone cycles.[12] The review did not identify hydrogen combustion by-products as being a disbenefit or impact of using hydrogen for heating. Most research assessments of hydrogen impacts on the atmosphere have focused on global issues arising from leakage into the air from the distribution network rather than point of use effects.[13–15] Other studies have quantified air quality benefits assuming its use was only *via* fuel cells and not combustion.[16] The possible NO_x air quality impacts of hydrogen use were however highlighted in a UK assessment of net zero pathways.[17]

This paper aims to raise a key policy issue associated with hydrogen† combustion – the emission of the air pollutant NO_x as a *minor* waste by-product when hydrogen is burned, and the potential mitigations.

What happens when hydrogen is burned?

The combustion of hydrogen is one of the simplest chemical reactions, something most people are aware of from school chemistry. In a pure atmosphere of oxygen, combustion proceeds *via*:

$$2H_2 + O_2 \rightarrow 2H_2O$$

With the exception of a small number of specialist high-energy applications, such as rocket engines, the vast majority of combustion of hydrogen takes place in the presence of air. When hydrogen is burned, the mixture involved is more accurately some combination of $H_2 + O_2 + N_2$. Hydrogen burns with a very hot flame and the temperatures generated in that flame can be sufficiently high that it splits normally stable molecules apart and this leads to the reactions:

$$N_2 + O \rightarrow NO + N$$

$$N + O_2 \rightarrow NO + O$$

$$N + OH \rightarrow NO + H$$

The combined formation process is referred to as 'thermal NO', or the 'Zel'dovich mechanism'[18] and occurs in all fuel – air mixed flames hotter than around 1300 °C. Below around 750 °C virtually no NO_x is formed. Thermal NO is also produced by gas boilers that burn methane, or internal combustion engines burning gasoline or diesel[19,20] [note: fossil fuel combustion can also lead to the formation of NO from other routes, 'prompt NO_x' *via* reaction of $CH + N_2 \rightarrow HCN + N \rightarrow NO$, and 'fuel NO_x' which derives from traces of organic nitrogen in some liquid fuels. Hydrogen combustion is not substantially impacted by these two effects].

Combustion of hydrogen has the potential therefore to generate NO (nitrogen oxide) as a minor waste by-product. NO is a critical air pollution emission which reacts rapidly in the atmosphere to form nitrogen dioxide (NO_2). NO_2 is a globally regulated air pollutant that is harmful to health, and which in turn contributes to the formation of photochemical ozone pollution and fine particulate matter ($PM_{2.5}$). The contribution of NO_x to the formation of ozone and wider atmospheric oxidant levels also has climatic effects. In the mid and upper troposphere ozone is a short-lived climate forcer which leads to warming, however the presence of NO_x can also generate higher concentrations of hydroxyl radicals (OH), which shorten the lifetime of methane *via* atmospheric oxidation. The IPCC 5th Assessment Report from 2013 considered it uncertain whether NO_x emissions led overall to a warming or a cooling effect.[21]

The potential formation of NO_x as a by-product of combustion of hydrogen is rarely discussed when the *pros* and *cons* of its use as a future climate-friendly fuel are debated. There exists some risk however that the adoption of hydrogen as a combustion fuel, if not well-managed, could lead to some unintended impacts arising from the co-emission of harmful NO_x. If hydrogen was commonly used as a fuel for homes, and effects not mitigated, those NO_x emissions would be concentrated in cities with high population densities, which in turn are home to often more disadvantaged communities.

Combustion is not the only way in which hydrogen can be used as a fuel. It can be supplied to electrochemical fuel cells to directly generate electricity, and this approach does not generate NO as a waste by-product.[22] In principle the energy efficiency of a fuel cell is higher than an internal combustion engine (the former is limited by the Gibbs free energy, the latter by the Carnot efficiency). Without doubt fuel cells will form an important component of any future hydrogen economy, and their point-of-use impacts on air quality are unequivocally positive since this does not produce NO_x as a by-product. However, fuel cell technologies have a more limited application history than combustion and require a greater degree of technological transformation than the modification of existing combustion approaches to accommodate hydrogen.

• How hydrogen is used is critical in determining whether any air quality trade-offs could occur. Burning hydrogen in gas boilers or internal combustion engines carries with it the potential for NO_x emissions as a by-product. Using hydrogen in

† This paper refers to hydrogen fuel throughout, but the issues raised are equally applicable to ammonia, NH_3 which is a possible 'carrier' vector for hydrogen that may be used as a fuel in its own right. The most likely route to use for ammonia is large scale combustion, with shipping identified as a prime application.

fuel cells does not have this downside and so has in-built advantages from an air quality perspective.

Managing the risks

Hydrogen has been used as a fuel for hundreds of years, indeed the very first internal combustion engine designed by De Rivaz in 1806 ran on a hydrogen/oxygen mixture. Since that time a huge wealth of research and practical experience has been published that has identified the factors that influence the emissions of NO_x when hydrogen is combusted and importantly how it can be managed.[23,24] There is a particularly detailed literature on vehicle internal combustion engines where hydrogen has been used commercially since the 1970s, although it has never grown to become a mainstream fuel.[25] In more recent years in the context of passenger cars hydrogen use has shifted to fuel cell powertrains (*e.g.* Hyundai *Nexo*, Toyota *Mirai*) avoiding altogether issues of co-emissions of NO_x. The past literature on vehicle emissions is however highly relevant when considering the possible impacts of stationary engine applications such as back-up grid power which relies heavily on large diesel plants for primary power (so-called diesel farms).

The original work of Zel'dovich showed that the formation of thermal NO during combustion was closely tied to combustion temperature. Simplistically the hotter the flame, the more NO is produced. The temperature of combustion can however be managed through a number of mechanisms, most notably through changing the mixture of fuel to air (the equivalence ratio), by cooling the flame through the addition of other gases, or the design of the burner, for example premixing fuel and air.

When the amount of fuel is restricted relative to the amount of air, so-called *lean burn* conditions, combustion temperature is reduced and so are NO emissions.[26] In internal combustion engines the flame temperature in the cylinder can also be reduced by feeding some cooled exhaust gases back into the chamber, referred to as Exhaust Gas Recirculation (EGR),[27] a strategy applied in all modern diesel engines to also reduce NO_x.

There are however some important operational trade-offs. Reducing thermal NO emissions through use of lean-burn conditions can also lead to a reduction in energy output, measured either as the mechanical power from an internal combustion engine or heat released from a boiler. A designer of a hydrogen boiler or hydrogen-powered engine will therefore be faced with a dilemma. Maximise the fuel efficiency, power output and energetic performance of the appliance, but with increased NO emissions, or minimise the NO emissions and accept a reduced level of performance (an example optimisation in a spark ignition engine is described in Jabbr *et al.*).[28] These trade-offs can be summarised in an illustrative schematic for an internal combustion engine use of H_2 in Fig. 1. This figure shows a stylised behaviour of NO_x emissions, temperature and efficiency, drawn from previously published real-world test-bed data – *e.g.* ref. 31 for a hydrogen internal combustion engine. Similar curves of NO_x emissions and power output can be found for an aircraft gas turbine engine in ref. 24.

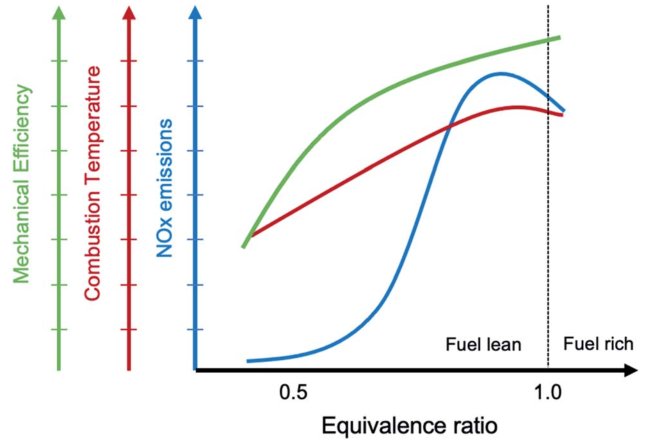

Fig. 1 The variation of mechanical efficiency, combustion temperature and NO_x emissions as a function of the equivalence ratio. The equivalence ratio is a measure of the amount of fuel relative to the amount of air. A ratio of 1 means that the amount of oxygen supplied in the air exactly matches the amount of fuel available for all the fuel to be burned with no excess. 'Fuel lean' means there is more oxygen available than there is fuel to burn, and 'fuel rich' more fuel than oxygen to completely combust it. Illustrative performance drawn from a recent real-world internal combustion engine, see ref. 31.

This NO_x – performance dilemma is not unique to hydrogen and similar decisions have been made for decades particularly with combustion conditions in diesel engines. History suggests that the air quality dimension of the trade-off is not always the leading priority, even if regulation is in place as mitigation to limit NO_x emissions.

- When used in thermal boilers or engines hydrogen generates NO_x in the same way as traditional fossil fuels like natural gas or diesel. Technical mitigation of NO_x emissions is possible but this can lead to reduced performance and increased capital and operating costs. It creates a dilemma over whether to prioritise optimal energy efficiency or air quality relevant emissions.

After-treatment of hydrogen combustion

For internal combustion engines and large-scale combustion in power stations an array of effective technologies exist for the after-treatment of exhaust gases to reduce NO_x emissions once combustion has taken place. For vehicles these include the three-way catalytic convertor (TWC), selective catalytic reduction (SCR) and lean NO_x traps (LNT) all typically applied in combination with the previously described strategies associated with fuel mix and exhaust gas recirculation. All these can be applied to hydrogen engines and hydrogen combustion. After-treatment mitigation of NO_x emissions from hydrogen is therefore possible in many end-use combustion applications using exactly the same technologies that are applied today for fossil fuels, but these do inevitably add cost and complexity [as an example, the International Council on Clean Transportation estimate the manufacturer technology costs of meeting European Stage V NO_x emission standards for a non-road 2.6 L engine are 1696 USD per unit;[29] the cost passed to the purchaser can be factor of 2.5 higher than this]. More generally there is economy of scale to exhaust after-treatment: industrial-scale hydrogen combustion is likely to be more cost effectively mitigated with after-treatment technologies for NO_x than domestic-scale usage.

A notable current exception to NO_x mitigation using after-treatment is gas combustion in small domestic heating boilers. The economics and design of home boilers does not lend itself to after-treatment in a straightforward way, beyond some simple flue gas recirculation or burner design modifications. The precise evolution of domestic heating with gas remains uncertain although in the UK 6th Carbon Budget it is envisaged that there will be use of hybrid boilers than can accommodate methane/hydrogen blends. Irrespective of whether the fuel is methane, hydrogen or some blend of the two, the temperatures at which home boilers operate are sufficient for NO_x formation and there is a balance to be struck between energy efficiency, thermal performance and management of NO_x emissions. The very fast speed at which hydrogen flames travel makes controlling flame propagation (avoiding flash back) in the burner a particular engineering challenge. A review by Frazer-Nash Consultancy[30] highlighted a number of uncertainties associated with the replacement use of hydrogen in domestic boilers concluding:

"In the absence of testing it is difficult to determine whether the burner temperatures will be higher or lower with hydrogen than natural gas and therefore it is difficult to predict the implications for NO_x."

With the anticipated electrification of the passenger vehicle fleet, and potentially some fraction of the LGV and public transport fleet also, urban NO_x emissions are predicted to decrease significantly in the coming decades. Widespread use of hydrogen in conventional gas boilers would likely mean that exposure to urban NO_2 will become dependent on the effectiveness of that appliance sector in controlling emissions.‡ It is possible that boilers can be engineered in such a way that NO_x can be well-managed, but this will require extensive testing of performance prior to deployment and potentially industry-wide and hydrogen-specific (or hydrogen – methane blend) emissions standards. A more stringent type-approval standard for NO_x emissions from hydrogen boilers would be one mechanism to ensure that the vast national investment incurred in switching energy source to a lower carbon alternative also delivered air quality benefits to urban populations.

• Widespread adoption of hydrogen as a fuel for domestic heating could be a major step towards decarbonising homes. Burning hydrogen would however lead to some NO_x emissions, although this is a major area of uncertainty due to limited experimental data. In a future with a largely electrified transport fleet, home combustion of hydrogen could become the dominant source of NO_2 in cities. At present after-treatment technologies are not used to reduce domestic boiler emissions but this is not to rule out future innovation. Hydrogen use in fuel cells for domestic heating would avoid this potentially negative impact.

‡ Placing domestic gas boiler NO_x emissions in context A typical 30 kW h domestic gas boiler is currently limited to NO_x emissions of 56 mg kW h^{-1} by the EU Ecodesign Directive. In wintertime using such a boiler for ~4 hours per day would lead to ~5.7 g NO_x emitted. In comparison a EURO 6 diesel car is limited to an emission of 80 mg km^{-1}. The daily wintertime use of a home gas boiler therefore makes an air quality contribution for NO_x, broader similar in impact to a modern diesel car journey of ~70 km.

Hydrogen combustion and the regulatory environment

For virtually all hydrogen combustion applications there is an existing emissions control framework, for example the EURO vehicle exhaust standards or EU Ecodesign Directive requirement for home appliances. These are summarised in Table 1 from a largely European perspective, although some controls are global in nature, such as for aviation and shipping. The adoption of hydrogen as a widespread combustion fuel would, as a minimum expectation, fall within these existing type-approval and emission limit standards. Any NO_x emitted from hydrogen combustion processes would also fall within the reporting scope (and agreed national limits) of existing international obligations on air pollution emissions, such as the UNECE Convention on Long-Range Transboundary Air Pollution, and EU National Emissions Ceiling Directive (NECD). Meeting 2030 NECD emission targets for NO_x appears challenging for many European countries, with at least 16 countries needing to reduce emissions by more than 30% compared to 2018.

Simplistically, a baseline outcome would be that hydrogen-fuelled appliances, no matter what the end-use, would perform *no worse* for NO_x emissions than a contemporary fossil fuel counterpart. Given the significant political investment and technical effort that is typically involved in developing and agreeing new standards, it might be anticipated that applying existing emissions standards would be the path of least resistance.

Adopting that approach would however be one of low-level ambition and crucially not result in the most substantial NO_x air quality co-benefits from the decarbonisation investment. It is possible that in some applications that hydrogen use might lead automatically to lower NO_x emissions whilst delivering like-for-like performance by other metrics. However, given that NO_x emissions are to a large degree 'tuneable' through engineering choices, increasing thermal or mechanical performance at the expense of NO_x emissions, right up to the limit of the existing regulatory standard would be a 'human nature' response. As has been seen previously for NO_x and diesel passenger cars even the existence of a standard is not always sufficient, and delivery of benefits also depends on the robustness of the type approval and testing regimes.

• The future use of hydrogen as a mainstream fuel requires a parallel set of policies to define more ambitious limits on NO_x emissions from a range of hydrogen-fuelled systems. In the absence of new hydrogen-specific limits on NO_x emissions, equivalence with existing natural gas, diesel and gasoline performance would be the de facto performance benchmark. This would be a lost opportunity to reduce NO_x and improve

Table 1 Possible hydrogen combustion uses, control framework and possible emission impacts relative to fossil fuel Business As Usual (BAU)

Application	After treatment possible?	Regulatory framework	Better than BAU for NO_x?
Large scale combustion for electricity generation	Extensive	Directive 2001/80/EC limitation of emissions of certain pollutants into the air from large combustion plants	Likely lower NO_x than comparable natural gas fired power generation
Back-up grid power, diesel farm replacement	Yes	Directive 2015/2193 limitation of emissions of certain pollutants from medium combustion plant	Unclear, may aim to only match existing diesel NO_x emissions performance Fuel cells may be highly competitive
Non-road mobile machinery (NRMM)	Yes	Directive 2016/1628 emission limits and type-approval for internal combustion engines for non-road mobile machinery (Stage V).	Unclear, may aim to only match existing diesel NO_x emissions performance Fuel cells may be highly competitive
HGVs	Yes	Regulation no 595/2009 type-approval of motor vehicles and engines with respect to emissions from heavy duty vehicles (EURO VI)	Unclear, may aim to only match existing diesel NO_x emissions performance Fuel cells may be highly competitive
Passenger cars	Yes	Directive 2017/1151 supplementing regulation (EC) No 715/2007 on type-approval of motor vehicles with respect to emissions from light passenger and commercial vehicles (EURO 5 and EURO 6)	Likely to outperform diesel equivalents, possibly similar to gasoline equivalents Unlikely to see widespread uptake other than *via* fuel cells
Commercial heating boilers	Possible	Directive 2009/125/EC Ecodesign requirements for energy-related products	Potentially lower NO_x than natural gas
Domestic boilers	Unlikely	Directive 2009/125/EC Ecodesign requirements for energy-related products	Unclear
Aircraft turbine	Unlikely	ICAO regulatory limits for engine NO_x emissions CAEP/8	Unclear
Maritime shipping	Yes	MARPOL 73/78 and subsequent amendments	Likely to outperform heavy fuel oil equivalents, with major sulphur co-benefits

air quality at the same time as reducing greenhouse gas emissions.

Conclusions

The adoption of hydrogen as a net zero fuel is still in its infancy and many significant environmental challenges lie ahead, not only how it is to be sustainably produced and distributed, but also how it is used. The latter issue has received significantly less attention than the former. The simplicity and relatively modest technical adaptations needed to burn hydrogen as a direct fossil fuel replacement means that this route will almost certainly be followed in many industries, alongside other approaches such as fuel cells. Use of hydrogen combustion appliances within existing frameworks for air quality emissions control will certainly mean that NO_x emissions will not get worse with widespread adoption, but it may also mean they do not improve either. It is possible that other air pollutant emissions arising from hydrogen are better than fossil fuel equivalents, for example of carbon monoxide or directly emitted particular matter, and these benefits would need including in cost-benefit analyses. However, taking NO_x as a specific pollutant of concern, to reduce emissions beyond business-as-usual may require the development of new emissions standards for hydrogen fuel combustion appliances that go beyond the limits in place for fossil fuels.

There is the potential for perverse and unintended outcomes if not well managed. As an example, a temporary expansion in standby electrical generation capacity is anticipated as a transitionary measure needed on pathways to net zero, supplying peak power whilst additional low carbon energy infrastructure is installed. Back-up power provision is currently served in some locations by diesel farms, which may plausibly switch to hydrogen fuel instead as a lower carbon alternative. The optimal net zero supply-side energy input would be green hydrogen generated from surplus renewable energy. At point-of-use however the choice of hydrogen combustion as the energy vector could lead to air quality disbenefits from what was originally a zero-pollution energy source.

There is a high degree of technical feasibility for delivering low NO_x from hydrogen combustion, accepting that some compromises may have to be made around cost, efficiency and performance. New and more ambitious limits on NO_x from hydrogen may stimulate innovation in after-treatment technology and controls, particularly in the domestic boiler sector. Hydrogen or hybrid gas boilers are not however the only technical option available for heating homes. If NO_x is accepted as an undesirable outcome and minimised through regulation, a consequence may be a tipping of the economic balance in favour of alternative low carbon approaches such as better home insulation, solar heating and air/ground source heat pumps.

The complexity of hydrogen adoption is not under-estimated and may be complicated further because regulatory responsibilities for individual hydrogen end-use may lie at a national level across multiple different government departments, spanning energy, transport, environment, and for some sectors are managed internationally. However, the optimal time to develop those new standards is *before* a net zero hydrogen roll-out has become substantially advanced and major investments in new manufacturing and infrastructure become locked-in.

Conflicts of interest

There are no conflicts to declare.

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