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Introduction

The first trade-off between *utility* and *amenity* probably arose with fire. The utility was manifold: cooking, heating and illumination; and also artwork, as facilitated by a mysterious black pigment: ancient hands, pressed against cave walls, and exposed to strongly sooting flames, have left their silhouettes for modernity. The luminosity of the fire, and to some extent the heat, stemmed from the black pigment – namely, *soot*. Yet the amenity was also degraded, as the soot particles, when inhaled, were prejudicial to health; and it was fire, perhaps, that engendered the first respiratory diseases. Such ailments still plague developing countries, where households continue to rely on a variety of stoves for cooking and heating, ventilation for which is poor or virtually nonexistent (Koshland and Fischer, 2002).[1]

Smoke became more inimical once population densities increased to those of the first civilisations, and the problem greatly accelerated once coal began to replace wood as the principal fuel (Brimblecombe, 2001). The great forests of England were swiftly dwindling before a rapidly expanding population: fuel was needed for energy, and land for agriculture. The competition, between utility and amenity, is written into the legislative record: Edward I (reigned 1272–1307) issued a royal proclamation forbidding coal-burning in London; Edward II (reigned 1307–1327) ordered the torture of persons fouling the air with coal smoke; Richard II (reigned 1377–1399), more humanely, chose to control coal-burning via taxation; Henry V (reigned 1413–1422) established a commission to regulate the entry of coal into London (Wilson R., 1996); and Elizabeth I (reigned 1558–1603) legislated against coal-burning whenever Parliament was sitting.

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[1] Some misguided souls in the developed world express a preference for the ‘naturalness’ of open fires. This position is untenable even from the environmental standpoint that such people profess to adopt. Open fires are inefficient and largely uncontrolled; they consequently release into the atmosphere large quantities of soot and sundry organic compounds – with genuine toxicological risks. Contrastingly, the combustion inside today’s engines is controlled to great precision; the pollution is excessive because motor vehicles are present at high density, not because they are particularly polluting on a unit basis.

Particulate Emissions from Vehicles P. Eastwood

In 1648, coughing Londoners petitioned the government to prohibit the importation of coal from Newcastle, citing the injurious effects of the smoke; but they were unsuccessful. And in 1661, the famous diarist John Evelyn wrote:

> It is this horrid smoake, which obscures our churches and makes our palaces look old, which fouls our clothes and corrupts the waters so that the very rain and refreshing dews which fall in the several seasons precipitate this impure vapour, which with its black and tenacious quality, spots and contaminates whatever is exposed to it. (Cited by Wilson R., 1996).

What provided the greatest impetus of all was the Industrial Revolution: by 1819, the problem had become so conspicuous that Parliament appointed a committee to investigate how to make steam engines and furnaces less prejudicial to public health. It was concluded that smoke could be effectively controlled . . . but no action was taken. As a question of social policy, addressing smoke was often viewed as prejudicial to the nation's economy, and public good was held more important than private need. There also seems to have been a subtle transition from regulation as 'nuisance' to regulation as 'negligence' (Farrell and Keating, 2000). A technical question was whether to prevent the formation of smoke in the first place; whether the smoke, once formed, should be collected prior to emission; or whether the smoke, once emitted, should be more effectively dispersed, under the time-honoured cynicism 'the solution to pollution is dilution'. The construction of ever-higher chimney stacks, as the Industrial Revolution advanced, bears testimony to the dispersion principle (and in the modern era, power stations send their pollution above the inversion layer).

Smoke emissions from locomotives were finally regulated by the Railway Clauses Act of 1845, and from factory furnaces by the Town Improvement Clauses Act of 1847. From 1875, English law regulated smoke from factory chimneys under clauses in the Public Health Acts; London was singled out for more severe restrictions. This legislation was subsequently modified by the Smoke Abatement Act of 1926, which invested local authorities with powers to enforce the provision of equipment in new buildings as might prevent smoke. (Domestic grates were not, however, included.) Several towns established 'smokeless zones', wherein all smoke emissions were prohibited (Fishenden, 1964).

These measures were insufficient. In the 1950s, the duration of sunshine in cities such as Leeds, Sheffield and Manchester, during the winter months (when domestic hearths saw most use), was less than half that in the outlying districts. Pea-soup smogs, or just 'pea-soupers', sometimes lasting for several days, plagued many cities, and (quite apart from respiratory ailments) were responsible for numerous road accidents. As recently as 1950 it was reported that, in Glasgow, three tonnes of soot fell to earth per acre per year (cited by Wilson R., 1996).

The incident which undoubtedly provided the greatest case for further legislation was the notorious London smog of December 4–9, 1952, which arose through a freak combination of low wind speed, temperature inversion and dry weather; local levels of atmospheric particulate peaked at $7\,\mathrm{mg/m^3}$ (Maynard, 2001) – ten times the amount normally seen to day, even in the most heavily polluted of cities. The smog is reputed to have killed 4000 people:[2] the association between mortality, smoke and sulphur dioxide (SO_2) is pointedly illustrated by Figure 1.1 (cited by Wilson R., 1996). During this event, deaths attributed to bronchitis and pneumonia increased eightfold and threefold, respectively; there were also increases in deaths attributed to other respiratory or cardiac diseases (Higgins I.T.T.,

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[2] This incident is wearisomely related in virtually every book on air pollution, amongst which the present one is no exception. Around 4000 deaths is the figure usually quoted. Recent research reported in the *Daily Telegraph*, Dec. 14, 2002, suggests the smog-related deaths to have been confused with those of a flu epidemic, and that the real figure could be as high as 12 000. The newspaper article credits this work to D. Davis and M. Bell, but the present author has been unable to locate any further information.

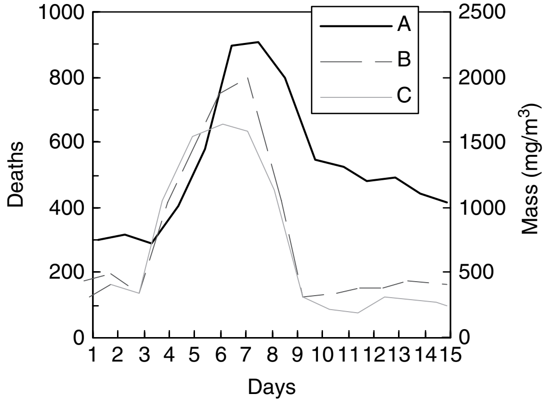

Figure 1.1 The London smog of December, 1952, and its fatal effects: A, number of deaths; B, mass concentration of SO_2; C, mass concentration of smoke. The graph is taken from Wilson R. (1996) (original reference Beaver (1953)). (The concentrations are averages over several sites).

1971). But post-mortems failed to reveal anything specifically particle-related; the common feature was that persons with cardio-respiratory diseases suffered exacerbations of their symptoms, and that some died as a result (Maynard, 2001). Morbidity rates increased also, as seen in various statistics for hospitalisations and sickness-benefit payments.

Suitably chastened, Parliament introduced the Clean Air Act of 1956, making the emission of dark smoke an offence. But mortality and morbidity in London, for certain respiratory diseases, remained until the early 1960s fairly well correlated with smoke and SO_2. After this period the proscription of smoke seems finally to have taken effect, inasmuch as mass concentrations fell from 300 to 60 $\mu g/m^3$; and there is some evidence of a concomitant improvement in public health (studies cited by Higgins I.T.T., 1971).

To be sure, the reason for the demise of pea-soup smogs was widespread conversion of domestic heating appliances to natural gas (Farrell and Keating, 2000). (The dieselisation of railways and small-scale power generation was another factor.) Smoke is a key symptom of poor air–fuel mixing: an inability to supply sufficient oxygen to the primary combustion zone is the eternal problem with solid fuels. And in the domestic arena, coal was often burned in a haphazard and uncontrolled way. Natural gas can be mixed with air automatically and far more precisely; hence the cleanliness of the combustion becomes far less dependent on the competence, or diligence, of the user.

Natural gas possesses two other advantages – it is low in sulphur and incombustible ash (principally metals). In the presence of moisture, and the catalytic effects of incombustible metals emitted as ‘fly ash’, fuel sulphur invariably finishes up in the atmosphere as *sulphuric acid*. The combination of soot, ash and acid was what formed the infamous smogs. This drives home forcefully the importance of *secondary* pollution: not directly emitted, but formed subsequently through ongoing atmospheric reactions. Secondary pollution greatly obfuscates the protection of ambient air quality: it can only be controlled indirectly, via the primary pollution, and a thorough knowledge of atmospheric processes.

We have followed the history of smoke, from the first beginnings to the modern era; and from primeval man, to the grimy image, etched into our cultural psyche, of the nineteenth-century industrial city. But how does this relate to particles emitted by today’s internal combustion engines? This question does not, usually, concern aesthetics, as exhaust plumes are less visible, indeed approaching invisibility, just as their offending dirt and filth are evanescent. Nor is the danger nowadays a slow asphyxiation by acrid smoke, because particle concentrations in the ambient atmosphere are much lower than in the instances described – by two orders of magnitude. It should be emphasised that the pollution in the aforementioned London smog episode provided evidence of the risks to public health – evidence that was unambiguous and unmistakable. The situation today is not nearly so clear-cut; and considerably greater efforts are needed to avoid poorly focused or ineffective legislation (Hall *et al.*, 1998).

1.1 Air Traffic

Emissions legislation, directed at aircraft, began to appear at around the same time as for motor vehicles, i.e. in the 1960s (Kittredge and NcNutt, 1971). This is not exactly a coincidence: there is to some extent a parallel between the two types of transport: similar pollutants are emitted (carbon monoxide, oxides of nitrogen, particulate and hydrocarbons), and levels of traffic increased rapidly at about the same time. But in aviation, the internal combustion engine was not the culprit. The need to control emissions, especially those of smoke, first arose towards the end of the 1950s, when the gas turbine began to replace the internal combustion engine as the preferred power plant. This was not only a problem with civilian airliners: the conspicuousness of the trails was particularly undesirable with military aircraft (Fiorello, 1968).

In fact, the earliest jets did not smoke appreciably; the problem seems to have been instigated by increases in combustion pressure and heat release rates, undertaken to improve performance (Shayeson, 1967). Smoke emitted by aircraft was particularly noticeable during take-off and on the final approach to landing. The sight and sound of jet aircraft, especially when on full thrust, further focused attention; and an aircraft's smoke trail is inherently visible against an empty sky, unlike on the ground, where neighbouring buildings render assistance in hiding the evidence (Parker, 1971).

In gas turbines, soot is formed in fuel-rich regions of the combustor; it can be burned up prior to emission, but this action is impeded by overly rapid quenching. To combat soot, therefore, the engine manufacturers were forced to redesign their combustors, as, unlike with motor vehicles, aftertreatment, that is, an exhaust gas clean-up, is impractical (Nelson, 1974). It was, apparently, possible to control the soot by directing air into the primary combustion zone, without, moreover, unacceptable increases in NO_x (Bristol, 1971). This soot–NO_x trade-off–since *both* pollutants must be reduced–is a constraint that continues up to the present time (Gupta A.K., 1997). The same trade-off is inherent in the diesel engine (to which we shall shortly turn): this is the principal reason why diesel soot has proven so intractable.

1.2 Motor Vehicles

Smoke has always been emitted by motor vehicles; and its sheer visibility, competing with the malodorous emission, ensures immediacy in the eyes, and noses, of the public. Yet in the days of Otto and Diesel, gasoline engines were denounced just as much as diesel engines. There is an interesting paper published by the newly formed US Society of Automobile Engineers [*sic*], from before the First World War, which castigates automobiles of 'the early days' [*sic*] as 'ill-smelling affairs' (Howe, 1910). It was at this time that New York City's Department of Health extended an old smoke ordinance to include motor vehicles: 'No person shall cause, suffer or allow dense smoke to be discharged from any building, vessel, stationary or locomotive engine or motor vehicle, place or premises within the City of New York.' Intriguingly, the ordinance gave no definition for a smoke density that *was* acceptable, even though it did, apparently, make a culprit liable to police arrest. (Perhaps the authorities realised just how difficult meaningful measurements of smoke are to conduct.) The author of the paper recognises the close association between smoke and tampering, and calls for manufacturers to prohibit maladjustment of carburettors such that black smoke is emitted. Apparently, it was not unknown for exhaust gas at this time to contain carbon monoxide to the tune of 12 % [*sic*].

The introduction of leaded gasoline in the 1920s gave rise to a new type of particulate emission. Tetraethyl lead was added to gasoline as an antiknock agent; and, to prevent the formation of lead deposits in the engine, ethylene dichloride and dibromide, as scavenging agents, were also added. These additives were emitted as particles of lead bromochloride, bromide and chloride. Because they were invisible, lead particles were arguably more insidious than smoke. But no changes were made until motor vehicle ownership reached modern-day proportions.

The eventual withdrawal of leaded gasoline happened for two cogent reasons. First, elevated levels of lead were discovered in the blood of persons exposed to traffic emissions. This raised a host of health issues (Russell Jones, 1987), a chief one being the suspected impairment of child development. Second, a pressing need had arisen to tackle other emissions, and catalytic converters were the only feasible technology with which to do this. Catalysts are swiftly poisoned by lead, and no aftertreatment sufficiently tolerant to this metal has ever been found.

The USA in the 1970s saw the introduction of the first catalytic converters; they were for oxidation purposes only (unlike today's three-way catalysts). Perversely, this led to another type of particulate emission: droplets of sulphuric acid. Reading the literature with the benefit of hindsight, this problem appears to have come as something of a surprise to the automotive industry; however, the appearance of sulphuric acid, after burning fuel which contains trace amounts of sulphur, and passing the exhaust gas over an oxidising catalyst, will not come as a surprise to any chemist. Again, this problem was not really solved as such; rather, technology simply moved in another direction. The introduction of three-way catalysts in the 1980s forced the adoption of stoichiometric, rather than lean air–fuel ratios; and the oxidising conditions in the exhaust necessary for the formation of sulphuric acid were lost.

Thus far we have discussed particles that are, in a sense, interlopers, inasmuch as they arise by side reactions in the combustion process. Not all particles are produced this way. Unburned fuel, if emitted in sufficiently large quantities, forms liquid droplets in the exhaust plume, and these droplets are perceived as *white* smoke. For engines that have been designed successfully, calibrated competently and maintained assiduously, this type of smoke is not an issue. Emissions legislation now places strict limits on the quantity of unburned hydrocarbons that may be emitted; and to obtain white smoke, these limits must be exceeded by an order of magnitude, so that this phenomenon is a little academic. White smoke may still be a problem, however, during cold starts, especially at low ambient temperatures. Similarly, *blue* smoke, if caused by the escape of lubricating oil, is indicative of improper or inadequate maintenance, and seldom otherwise seen.

Finally, there is the long-standing issue of *black* smoke, chiefly discharged by diesel engines, and whose characteristic blackness arises from the element carbon. Black smoke appears to be an unfortunate and ineluctable consequence of diesel combustion, if only because it has frustrated the attempts of generations of engineers to eliminate it. Solely from an engine performance perspective, black smoke reflects the efficiency, or perhaps more appropriately, the inefficiency of the combustion, because it represents lost energy. But that is not quite the point: with today's diesel engines, the carbon that is usually emitted as soot, if it were to be successfully burned, would make little difference to the overall combustion efficiency, whereas if a diesel engine at full load were to emit only 0.5 % of the fuel as black smoke, the result would be completely unacceptable from an emissions perspective. The combustion efficiency, taking into account unburned hydrocarbons, carbon monoxide and soot, is usually better than 98 %, so that, from an energy conversion perspective, the combustion is virtually complete (Heywood, 1988, p. 509). The implications for public health are a completely different issue.

1.3 The Legislative Framework

Up to now, ‘smoke’ and ‘particulate’ have been employed fairly loosely and interchangeably. From now on this practice will be insufficient, and both terms must be defined much more closely. Strictly speaking, ‘smoke’ denotes an *aerosol*, i.e. a suspension of particles in a gas, whereas ‘particulate’ describes a collection of these particles, say, on a filter.[3] But, passing from these semantic quibbles, a more important difference, of a technical (and indeed environmental) nature, arises: when there is smoke, the presence of particles may be safely inferred; but when there are particles, these are not necessarily manifested as smoke. *Smoke, then, is the visible corollary of particulate*. Thus, ‘particulate’ is a considerably *broader* and indeed *all-encompassing* term: it incorporates smoke, and much else besides. So, we can now state this distinction explicitly, as follows:

- *Particulate.* All material which deposits on a filter.
- *Smoke.* All material which attenuates a beam of light.

These two definitions – both of which, incidentally, exclude condensed water – should not be confused: they are often directionally consistent, but this consistency is not by any means an essential prerequisite: in fact, the mass of particulate discharged by a smoking vehicle might actually be less than for a nonsmoking vehicle (Knapp *et al.*, 2003). This is because particle masses and particle interactions with light are entirely different measures – consistent only when the particles under investigation (size and composition) are also consistent.

In the USA, smoke from heavy-duty diesels was first legislated on in 1970 as ‘opacity’, that is, the fraction of light successfully traversing the exhaust stream. The development of particulate-control legislation is complicated (e.g. Cucchi and Hublin, 1989; Walsh and Bradow, 1991; Walsh, 1993; Charmley, 2004), but, simplifying somewhat, the first diesel particulate standards in the world were established in the USA in 1980, and related to passenger cars and light-duty trucks; heavy-duty engines and trucks were subsequently covered in 1985. In Europe, particulate emissions from diesel engines were first controlled in 1989, via EC Directives 88/436, 91/441 and 91/542 (Hall *et al.*, 1998).

Particulate is now strictly regulated in most countries, and the mass emitted by both light-duty (g/km) and heavy-duty (g/kW h) diesels has decreased since the 1970s, by more than two orders of magnitude. It is not improper to observe that the debate between industry and legislatures about these regulations has been contentious at times (Merrion, 2003). While the enactment of this legislation has undoubtedly helped to shape, and propel forward, the technology of emission control, the diesel engine has, at times, seemed threatened with extinction (Pethers, 1998); indeed, the downward trend in statutory requirements is set to continue unabated.

Current legislative practice in most countries extends, in a belt-and-braces manner, to smoke *and* particulate. But since smoke is far more conveniently measured, it tends to be used as an in-use compliance test, i.e. for *individual* vehicles during the course of their lives – perhaps those pulled over by the authorities for roadside checks. In the UK it is an offence for any vehicle to emit smoke at levels that impair visibility for other drivers; clearly, therefore, this is not so much to protect air quality as to ensure road safety. Smoke is assessed by a roadside or garage test, in which the clutch is disengaged and the engine rapidly accelerated up to the governor run-out (on passenger cars, usually between 4000 and 5000 rpm). This is the so-called ‘free acceleration’ test. It is quick and convenient, but says nothing about the smoke emitted during *real* driving, i.e. with the engine operated over a

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[3] Exact speakers of English prefer to stay with this strict definition; but in this text, ‘particulate’ is applied more loosely insofar as it denotes also the suspended particles *prior* to collection.

genuine duty cycle. The free acceleration test is the sum total of what a vehicle is likely to experience after it has been sold, according to the current regulations.

The regulations to be satisfied *before* sale relate to a vehicle *model*, and are known as 'type approval' or 'homologation'. These regulations are much stricter, and the manufacturer must demonstrate satisfactory levels of smoke *and* particulate. On an engine dynamometer, the smoke is assessed in two ways: first, for a progressive series of steady states along the full load curve (otherwise known as the torque limit), and second, for a free acceleration test as already described. Particulate emissions from an actual vehicle (i.e. the full power train) are assessed on a chassis dynamometer.[4] The vehicle is run over a (transient) drive cycle, during which the exhaust gas is fed into a 'dilution tunnel', mixed with air, and then filtered. This procedure is designed, among other things, to approximate the dilution of real exhaust plumes in the ambient. Current legislation mandates a dilution such that the exhaust gas temperature at the filter is less than or equal to 52°C:[5] this condition is pivotal, as it decides the transfer of material from the gas phase into the particulate phase. Once the drive cycle is completed, the filter is removed, conditioned to a certain temperature and humidity, and then, to determine the mass of particulate it has retained, simply weighed.

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[4] This description focuses on light-duty vehicles. Legislative practice for heavy-duty vehicles is different; engine dynamometer rather than chassis dynamometer tests are preferred. Some countries also only use steady-state (cruise) tests. Test protocols vary considerably from country to country, and are continually evolving; the information here is necessarily generalised.

[5] This is the long-standing specification, the laxity of which in view of today's stringent emission control legislation is well recognised. At the time of writing, tighter specifications such as 47 ± 5°C are under discussion (Wu *et al.*, 2007).