Ambient particulate matter: sources and apportioning

Both current and predicted levels of PM emissions are subject to great uncertainty; hence the development of meaningful air quality strategies is complicated.

INTRODUCTION

The quality of the air we breathe has become an increasingly important and emotive issue over the past few years. To date, the regulation of air quality has focused mainly on outdoor air (even though for particular pollutants, indoor concentrations may be greater and exposures are often more significant—on average we may spend up to 90 per cent of our lives indoors). Ambient air quality standards have been established for many different pollutants, including standards for particulate matter in various countries.

Unlike many air pollutants, ambient particulate matter is not well characterized; it encompasses a range of different sizes and has variable chemical composition depending on where, when and how it is sampled. This variation depends both on the nature of the original sources and on subsequent physical and chemical transformations. At a particular location, the quantity and composition may vary throughout the day, and will be subject to seasonal variation, impacts of the weather and proximity to local sources.

So what is ambient particulate matter? It is a complex mixture of varying composition emitted from a variety of sources, formed in the atmosphere by gas-to-particle conversion, or re-suspended by the action of the wind or mechanical processes.

Why are we interested? Over recent years, a large number of epidemiological studies have suggested an association between exposure to ambient particulate matter at current concentrations and adverse health effects. The establishment of national air quality standards in many countries has increased pressure to improve air quality and reduce the concentration of ambient particulate matter. It is essential to understand the relative contributions of the different sources to total particulate matter so that cost-effective abatement strategies can be devised to reduce emissions and, hence, improve air quality. With this in mind, we need to understand not just the concentrations of particulate matter in the ambient air, but the contribution made by different sources to the total.

AMBIENT PARTICULATE/AEROSOL

Ambient particulate matter is a complex mix of liquid and solid particles existing in dynamic equilibrium with gases in the surrounding air. It arises from a wide range of sources, both natural and related to human activity (anthropogenic), and can be divided into three main categories (APEG[1], 1999): primary particles (emitted to atmosphere directly from source); secondary particles (formed in the atmosphere from gas to particle conversion) and re-suspended particles (by wind or mechanical action). Particulate matter originating from different sources may have specific size ranges and chemical characteristics (useful for identifying sources i.e. 'fingerprinting'),

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but these characteristics tend to be lost as particles from different sources mix and 'age' in the atmosphere. These ageing processes include both physical interactions and chemical transformations, such as coagulation, adsorption of gaseous pollutants and oxidation.

Routine measurements of the concentration of ambient particulate matter are taken at fixed-point measuring sites using either gravimetric filter methods or electronic equipment such as the TEOM (tapering element oscillating micro-balance). These measurement techniques focus on size-selected particles (usually PM_{10}, although measurements of $PM_{2.5}$ are now becoming more common). Historically other parameters such as total suspended particulate (TSP) and black smoke (BS) have been used. The various methods collect and measure different types of particulate matter and it is difficult to calibrate these methods against each other or the 'true' ambient concentrations. As PM from different sources also varies, the choice of collection method will influence the assessment of the predominant contributing sources (i.e. the main sources of black smoke, PM_{10} and $PM_{2.5}$ will be different). It is also recognized that underestimation of total mass (in particular, secondary particulate contribution) may result from evaporative losses of more volatile particulate matter, such as ammonium nitrate, during collection (Hering and Cass, 1999).

ASSESSMENT OF SOURCE CONTRIBUTIONS

Taking these issues into consideration, it is impossible to assess the contribution of individual sources to total ambient particulate matter from simple air quality monitoring, or indeed to derive the contribution of secondary, re-suspended or transboundary particulate without more detailed chemical analysis. Historically, source contributions to total primary emissions have been developed from information on emission factors estimated for particular processes or technologies. From the emission factor, for example, for a furnace of particular design, knowledge of fuel throughput allows the contribution to total emissions to be estimated. These contributions can then be aggregated to develop an overall source inventory. CORINAIR (CORe INventory AIR) is probably the most extensive source inventory available for European gaseous emissions to air. However, emissions of primary particulate matter are not included in CORINAIR. The most widely cited inventory of European primary particulate matter emissions has been developed by the Dutch scientific research organization, TNO, and includes information on PM_{10}, $PM_{2.5}$ and $PM_{0.1}$ emissions (TNO, 1997). A key issue for source inventories is verification of the estimates by reference to real world measurements. Validation of the inventory for the different PM size fractions is currently hampered by the lack of reliable data on ambient particulate matter characterization and source attribution.

In compiling the European PM inventory, TNO acknowledged the limited availability of information on emission factors and their variability, especially in relation to $PM_{0.1}$; TNO describe reliable data on $PM_{0.1}$ emissions as 'scarce'. Even for PM_{10} emission factors, there is generally no information available on what emissions control technologies were in place when the factors were estimated for each type of plant. The significance of uncertainty about the source contributions to total ambient small particles becomes increasingly important as the health effects focus moves to smaller and smaller particulate matter. For the UK, 25 per cent of PM_{10} and 60 per cent of $PM_{0.1}$ emissions are reported to come from road transport (APEG, 1999); these estimates were developed from information from USA data (US EPA, 1995) and the TNO inventory. However, a recent source apportionment study in the US has suggested that 80–90 per cent of primary particulate emissions are fugitive dusts, with only a 3–9 per cent contribution to total PM_{10} from automotive exhaust emissions (Chow *et al*., 1999). Similarly, a study in Australia has suggested that automotive sources account for only 13 per cent of PM_{10} and 6 per cent of primary $PM_{2.5}$ aerosol mass (Chan *et al.,* 1999).

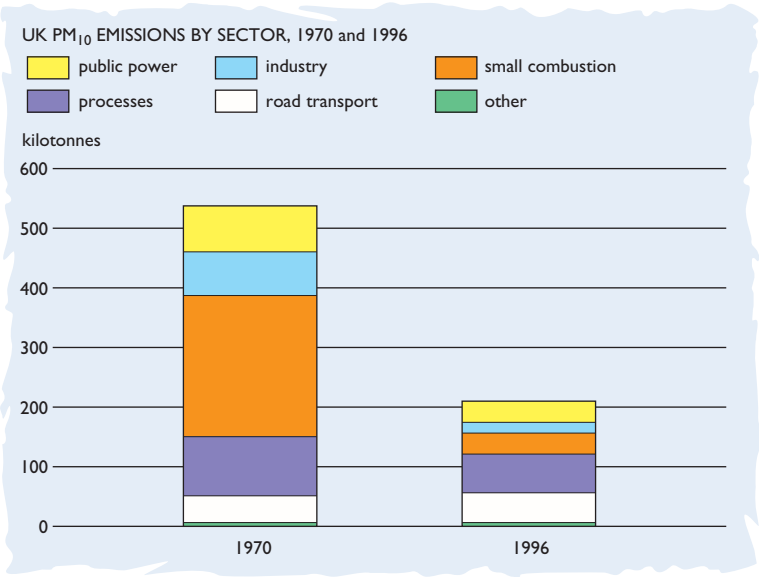

Figure 1
Total PM_{10} emissions and source contributions have changed markedly from 1970–96.

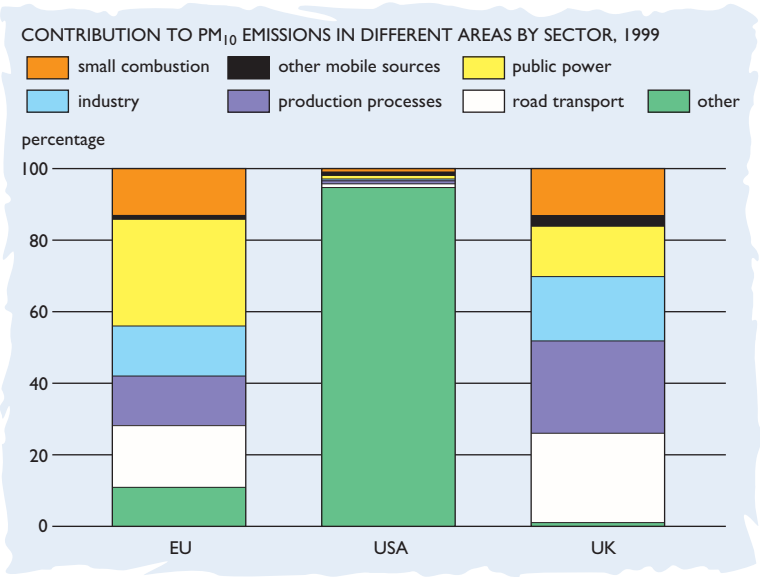

Figure 2
Percentage contributions to PM_{10} by different sectors vary considerably between Europe and the USA.

Figure 1 shows how both total PM_{10} emissions and the source contributions have changed during the period 1970–96 in the UK (APEG, 1999). Figure 2 gives a wider overview of the percentage contributions of different sectors to total primary PM_{10} emissions in different regions, based on existing inventories (from Holman, 1999).

It is clear from these data that the relative contributions to ambient primary particulate matter vary enormously between Europe as a whole and the UK, and even more so between Europe and the USA. The US inventory indicates that most PM_{10} emissions come from 'other' sources, which include fugitive dusts from paved and unpaved roads, agriculture and forestry, whilst in the UK these contributions are very small. Certainly, some of the variations between regions reflect real differences in the relative importance of particular sources. However, they do also serve to prompt questions about the robustness of the methodologies employed in different locations to arrive at the estimates. Methodologies must be consistent if comparisons between inventories are undertaken and aggregated inventories are to be meaningful.

On a worldwide basis, estimates of the overall contribution of natural, primary and secondary contributions to ambient particulate matter have been published (IPCC, 1996); the total anthropogenic contribution is estimated to be approximately 11 per cent (see Table 1).

A weakness of all of these inventories is their focus on primary emissions, whilst it is clear that secondary and re-suspended particulate makes a significant contribution to total ambient PM mass. For $PM_{2.5}$, Holman suggests that the secondary particulate contribution may be four times greater than the primary (Holman, 1999). Similarly, the inventories do not account for the contribution made by transboundary long-range transport of particulate matter, which is increasingly recognized as representing a significant proportion of the total particulate matter load under appropriate meteorological conditions (APEG, 1999).

In order to be useful, emission factors must be updated to keep abreast of developments in combustion technology and abatement methods. The positive impact of developing legislation on emissions is easily underestimated when inventories are not updated. This results in increasing uncertainty in emissions estimates (and air quality predictions) for future scenarios, the further we look ahead. Incomplete or inaccurate information on emissions source contributions based on outdated emission factors risks the establishment of the wrong priorities for legislation.

As an alternative to this form of 'bottom-up' analysis, several groups of investigators have reported on extensive chemical characterization of particulate matter produced by different sources, which they have used to define profiles or 'fingerprints' for particular types of particulate matter in the ambient mix. From this form of detailed analysis, it has been possible to begin to investigate real world source-receptor relationships for different sorts of ambient particulate matter (e.g. Cass, 1998; Kleeman and Cass, 1999; Spindler *et al.,* 1999). Such approaches are especially valuable in estimating the primary source contributions of fugitive dust sources, such as wind-blown dust, re-suspended road dust and sea spray, for which emission factors are not meaningful.

Sources of worldwide particulate matter

Source	*Type*	*Emissions (Mt/y)*	*Percentage of total emissions (%)*
Natural			
Primary	Soil dust (mineral)	1500	44
	Sea salt	1300	38
	Volcanic dust	33	1
	Biological debris	50	1
Secondary	Sulphates from natural precursors (as ammonium salt)	102	3
	Organic matter from biogenic VOC	55	2
	Nitrates from NO_x	22	1
	Total (Natural)	*3060*	*89*
Anthropogenic			
Primary	Industrial dust, etc.	100	3
	Soot (elemental carbon) from all fossil fuels	8	<1
	Soot from biomass combustion	5	<1
Secondary	Sulphates from SO_x (as ammonium salt)	140	4
	Biomass burning	80	2
	Nitrates from NO_x	36	1
	Total (Anthropogenic)	*370*	*11*
TOTAL		3430	100

Table 1
Total anthropogenic contribution to ambient PM worldwide is estimated to be 11 per cent. (Source: IPCC, 1996)

Emissions inventories are not available for all countries and, therefore, there is an incomplete representation of the overall picture. Different emissions inventories often employ different source categories, which makes inter-comparison and aggregation very difficult. They are often limited in their spatial coverage, and extrapolation to larger domains requires validation of the underlying assumptions. However, the available inventories do allow a 'first order' estimate of the source contributions of the major sectors to the overall primary emission.

SUMMARY

Limited availability of particulate matter emissions data (in particular for the smaller size fractions), means that, for Europe as a whole, current estimates are subject to significant uncertainty and error. Consequently, the basis for forward projection of PM emissions is subject to great uncertainty.

The contributions made by automotive and different stationary emission sources (primary PM) and the significance of secondary and transboundary PM are currently uncertain. Knowledge of the source contributions to ambient PM in the size ranges of particular interest from a health perspective ($PM_{2.5}$, $PM_{0.1}$ and smaller) is essential, but currently weakest.

The development of meaningful and cost-effective strategies to improve air quality by reducing emissions of particulate matter (and precursors of secondary PM) is currently hampered by this uncertainty. As a consequence, there is a real risk that costly legislation will be enacted that will fail to produce the anticipated benefits.

Note: The full list of references cited in this article may be found on page 26 of this PDF version of the Review.