AIRBORNE PARTICULATE MATTER – CHALLENGES FOR ABATEMENT

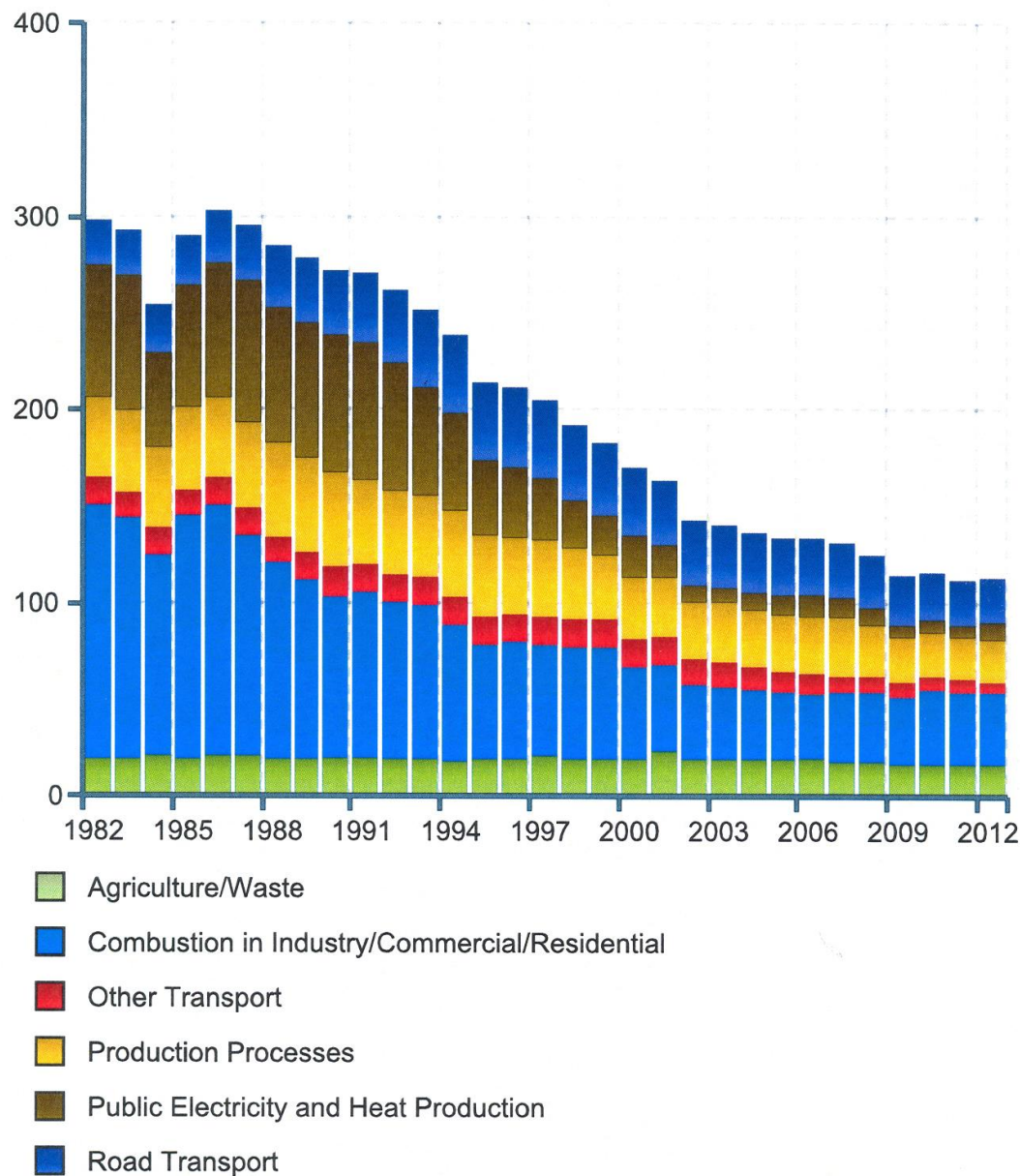## Roy M. Harrison

University of Birmingham

and

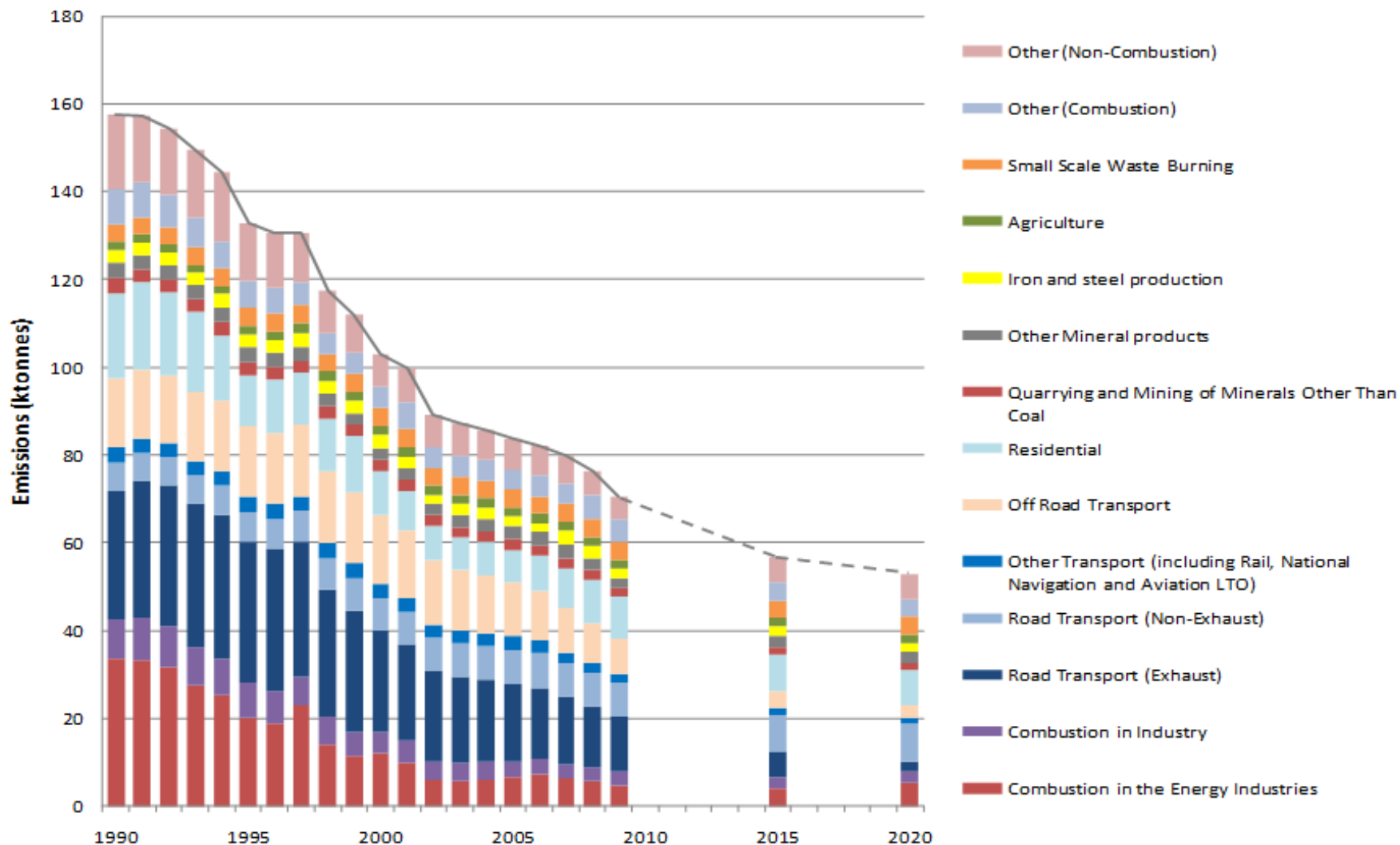## UK National Centre for Atmospheric Science

CONTENT

- Sources of particulate matter
- Receptor modelling of particulate matter
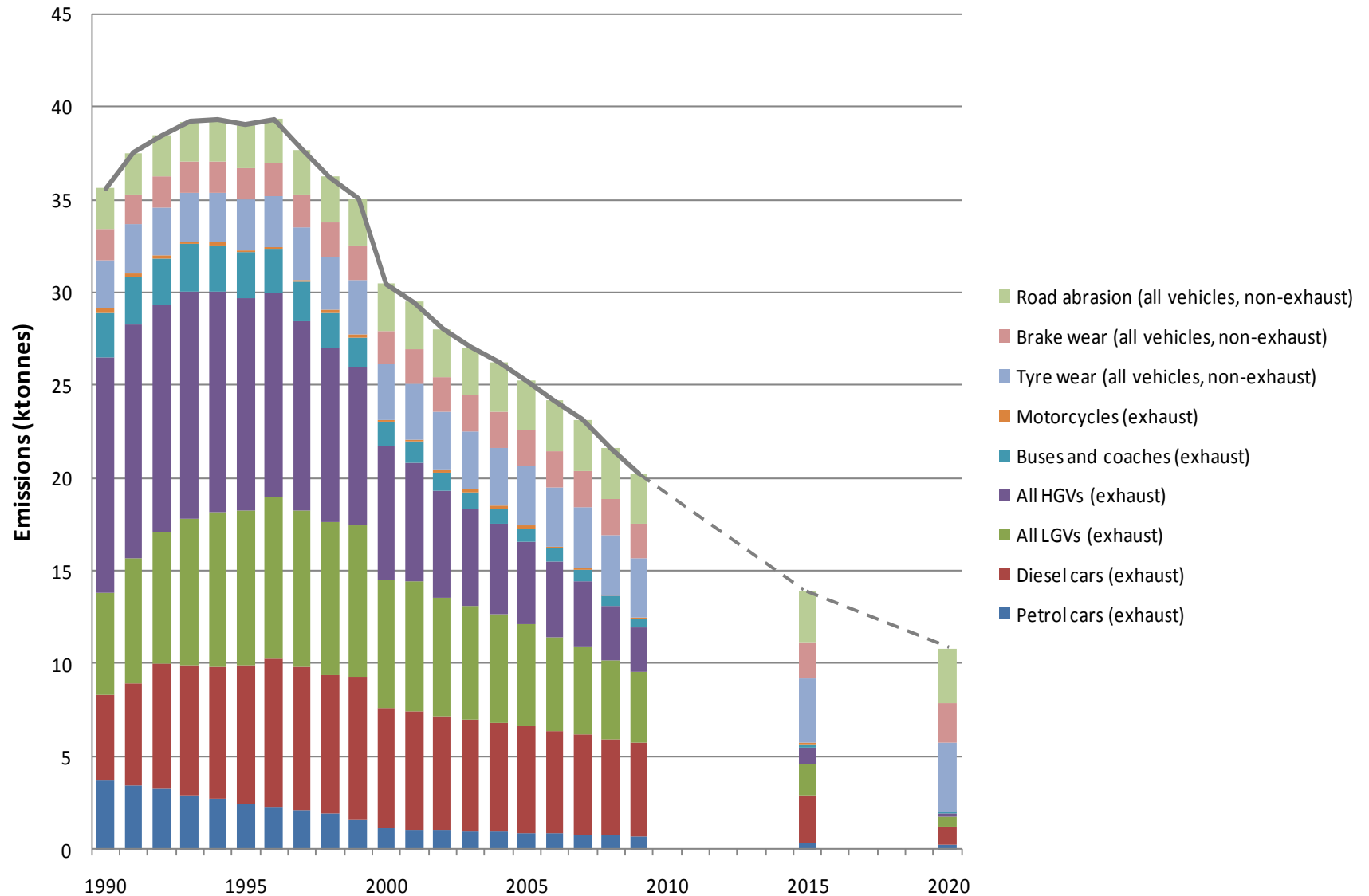- Abatement issues
- Exposure reduction

PM 10 (Particulate Matter < 10µm) (kilotonne)
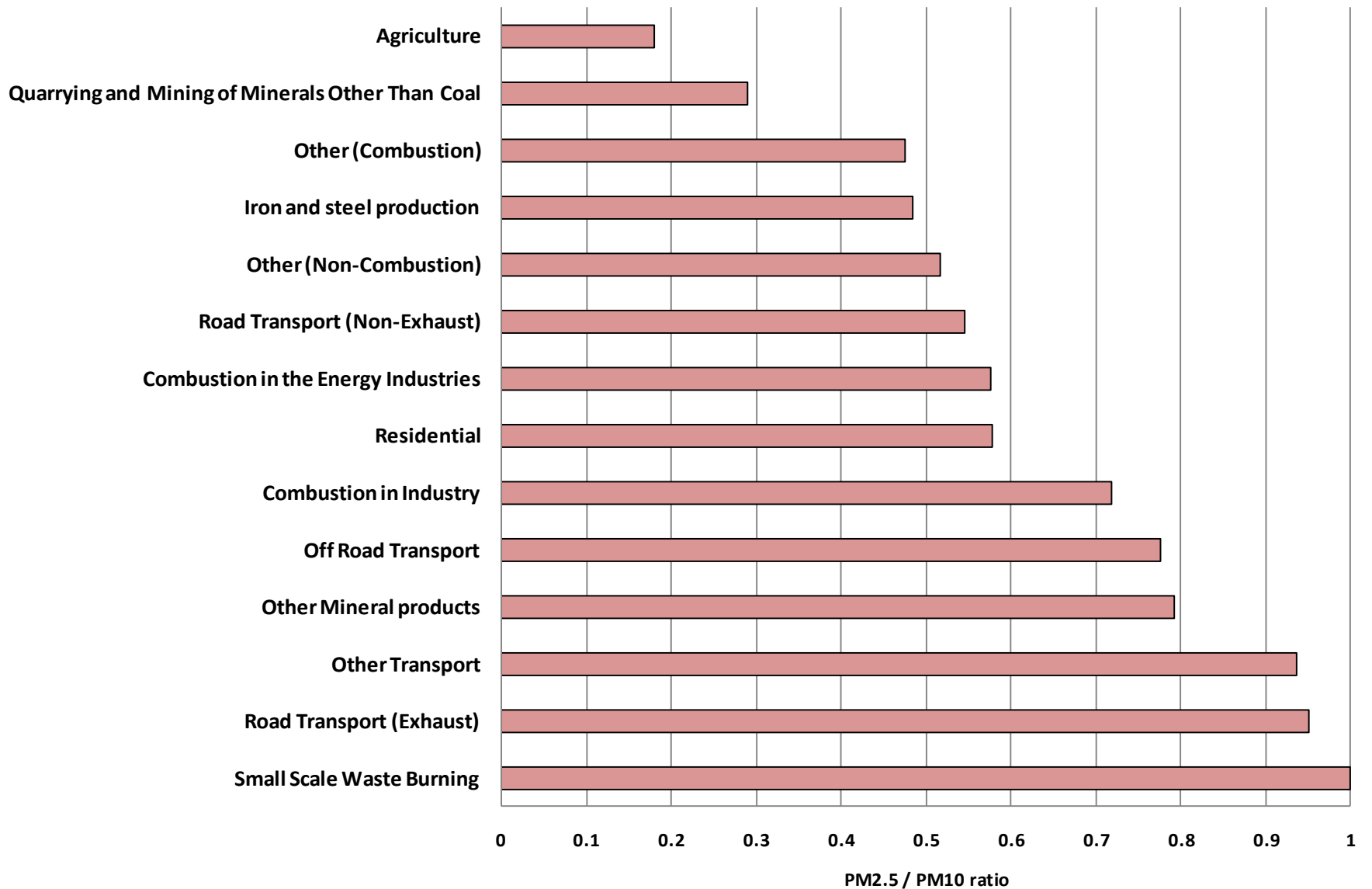
Legend:
- Agriculture/Waste
- Combustion in Industry/Commercial/Residential
- Other Transport
- Production Processes
- Public Electricity and Heat Production
- Road Transport

Total PM$_{2.5}$ emissions (kt), 1990-2009, 2015 and 2020

Total UK PM$_{2.5}$ Emissions from Road Sources (1990-2009, 2015 and 2020)

Ratios of PM2.5 to PM10 by Emission Sources in the UK

Emission Source	PM2.5 / PM10 ratio
Agriculture	~0.18
Quarrying and Mining of Minerals Other Than Coal	~0.29
Other (Combustion)	~0.47
Iron and steel production	~0.48
Other (Non-Combustion)	~0.51
Road Transport (Non-Exhaust)	~0.55
Combustion in the Energy Industries	~0.57
Residential	~0.58
Combustion in Industry	~0.72
Off Road Transport	~0.77
Other Mineral products	~0.79
Other Transport	~0.93
Road Transport (Exhaust)	~0.95
Small Scale Waste Burning	~1.0

PM2.5 / PM10 ratio

RECEPTOR MODELLING

- Use of air quality data to infer the sources responsible for measured pollution levels (opposite of dispersion modelling!)

- Receptor modelling of airborne particles depends upon an assumption of mass conservation

$$C_i \; = \; \sum^{j} f_{i,j} \, g_j$$

where C_i = airborne concentration of component, i

$f_{i,j}$ = mass fraction of component i in particles from source, j

g_j = mass of particles from source j in an air sample

- Analysis of many air samples for multiple chemical components is necessary

TYPES OF RECEPTOR MODELLING OF PARTICULATE MATTER

There are two main types

- **Chemical Mass Balance**
    - Requires only one air sample, although better results are obtained with more
    - Requires knowledge of chemical composition of particles from each source ($f_{i,j}$)
    - Varies g_j for all chemical components to obtain best fit to mass conservation equation

- **Multivariate Statistical**
    - Principal Component Analysis widely used, but Positive Matrix Factorization (PMF) has advantages and is more frequently utilised
    - Requires no advance knowledge of source chemical composition
    - Requires many separate samples, and identifies temporal correlations of components (e.g. Na and Cl in sea salt) in a multidimensional space.

RECEPTOR MODELLING WITH CMB MODEL

- Uses organic molecular markers and trace elements to apportion the carbonaceous component of $PM_{2.5}$

- Source apportionment of the entire $PM_{2.5}$ is conducted using the Pragmatic Mass Closure Model

- Results have been processed for winter air samples collected at LNK and HAR

CHEMICAL MASS BALANCE STUDY USING MOLECULAR MARKERS

- **PM$_{2.5}$ samples were collected and analysed for**

  - ➤ *n*-alkanes from C$_{24}$ – C$_{36}$
  - ➤ 9 specific hopanes
  - ➤ 13 PAH
  - ➤ 14 carboxylic acids
  - ➤ levoglucosan
  - ➤ cholesterol
  - ➤ inorganic marker elements (Si, Al)

CMB MODEL RESULTS

- **Model used to apportion sources of organic carbon to:**

  - ➢ **road traffic**
  - ➢ **vegetative detritus**
  - ➢ **dust and soil**
  - ➢ **wood smoke**
  - ➢ **coal combustion**
  - ➢ **natural gas combustion**

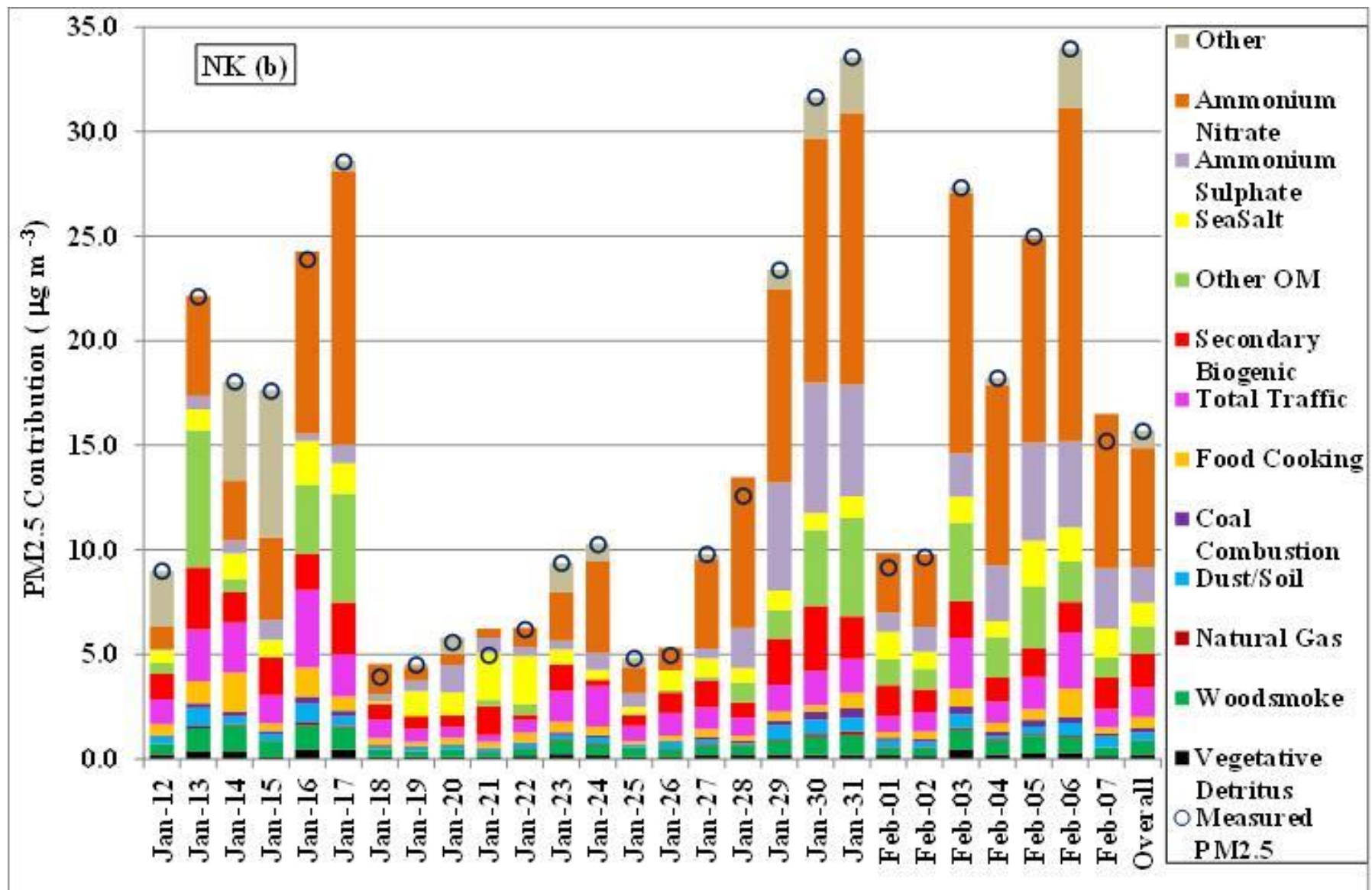# NK Site During ClearfLo (1)

NK Site During ClearfLo (2)
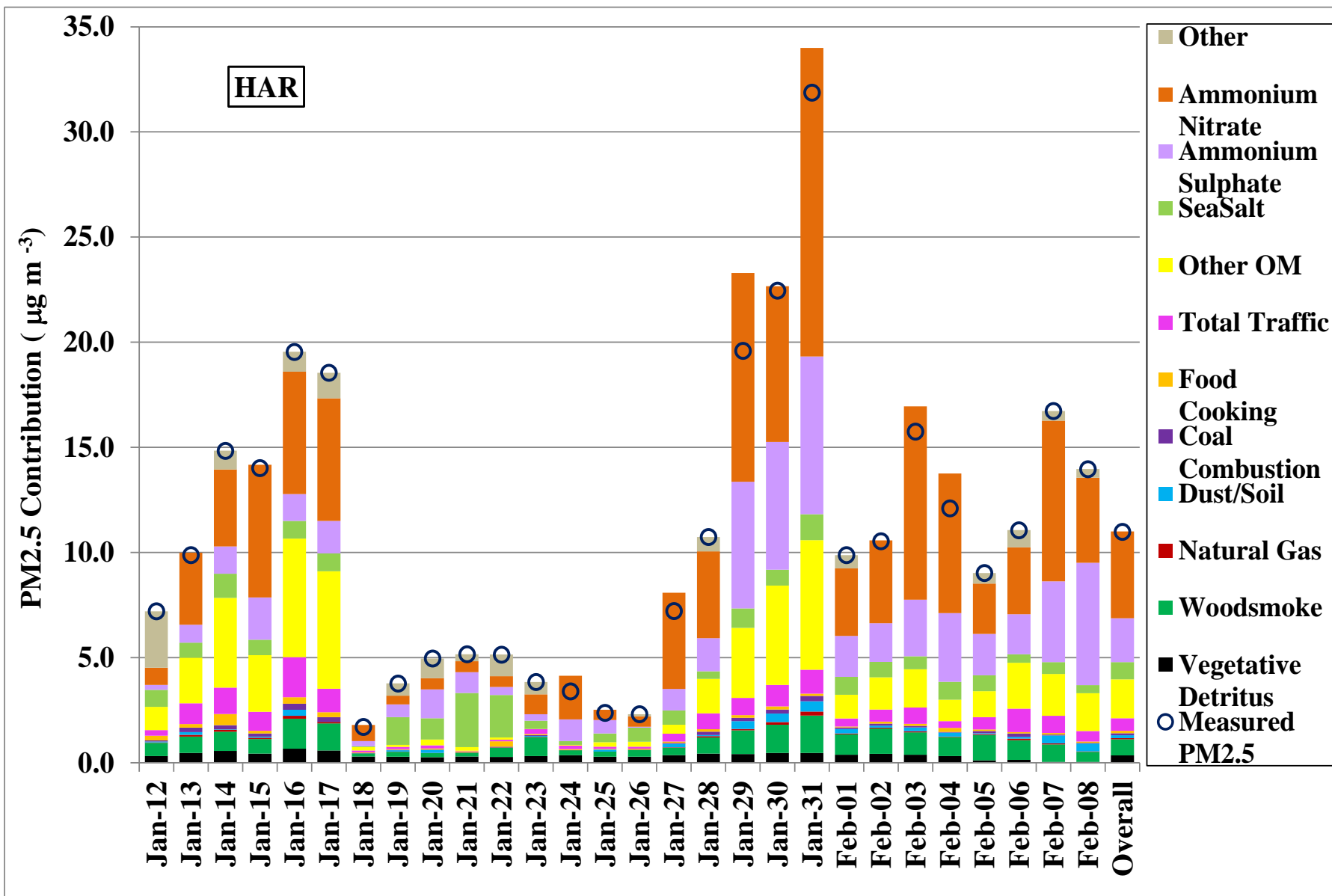
Daily OC Source Contributions at NK and HAR
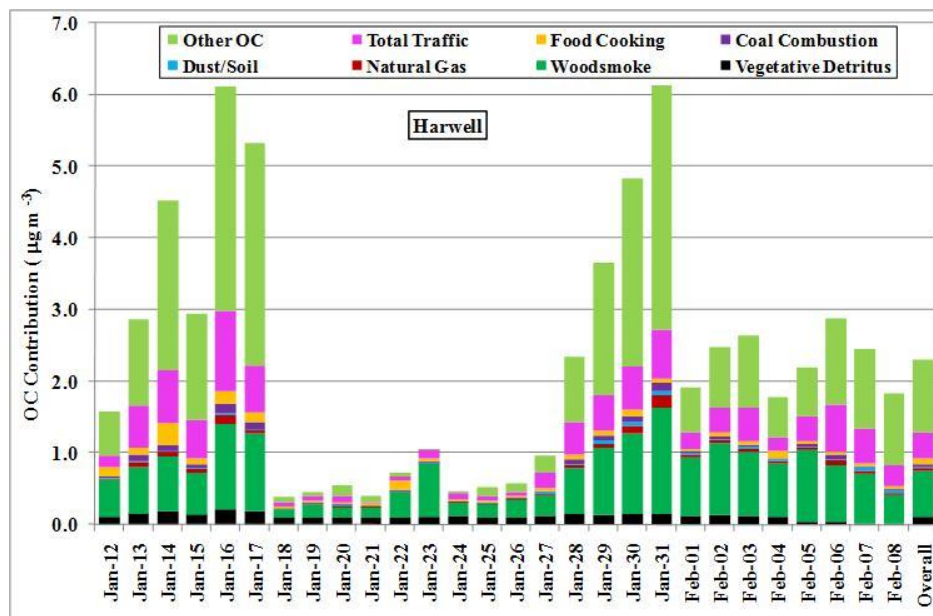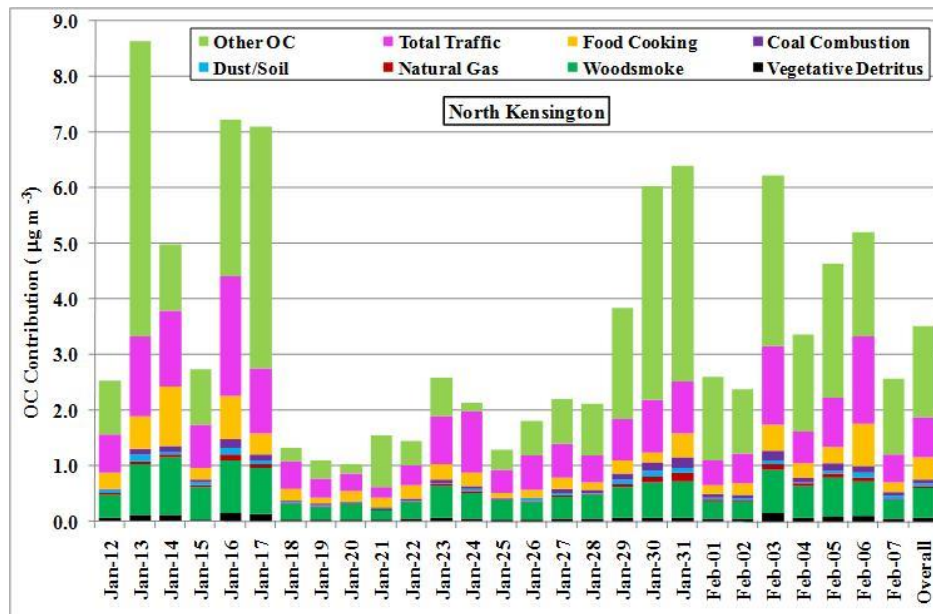
Daily OC Source Contribution Estimates with Secondary Biogenic Components at NK
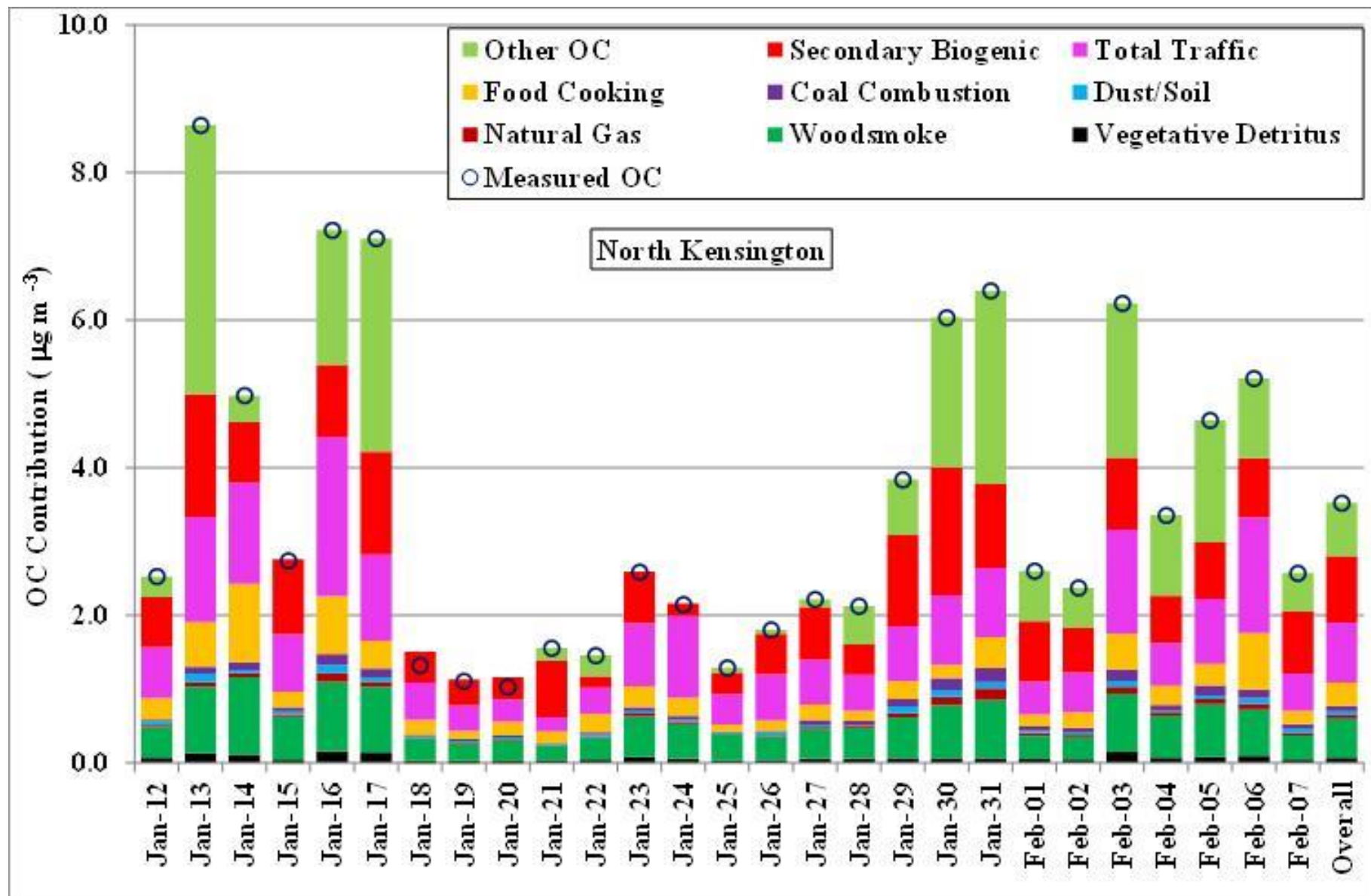
Daily PM$_{2.5}$ Source Contribution Estimates with Secondary Biogenic Components at NK

Daily PM$_{2.5}$ Source Contribution Estimates at HAR

SELECTED MEAN CONTRIBUTION TO PM$_{2.5}$ MASS, μg m^{-3} (%)

|	NORTH KENSINGTON	HARWELL
COOKING	0.69 (4%)	0.13 (1%)
WOODSMOKE	0.64 (4%)	0.76 (7%)
TRAFFIC EXHAUST	1.26 (8%)	0.61 (6%)
SULPHATES AND NITRATES	8.0 (51%)	6.2 (56%)
PM$_{2.5}$ MASS	15.7	11.0

SPECIFIC SOURCES: WOODSMOKE

- Concentrations are relatively uniform across southern England.

- Although comprehensive evidence is lacking, it seems probable that emissions are increasing.

- Emissions inventories have a major problem in quantification of residential wood burning.
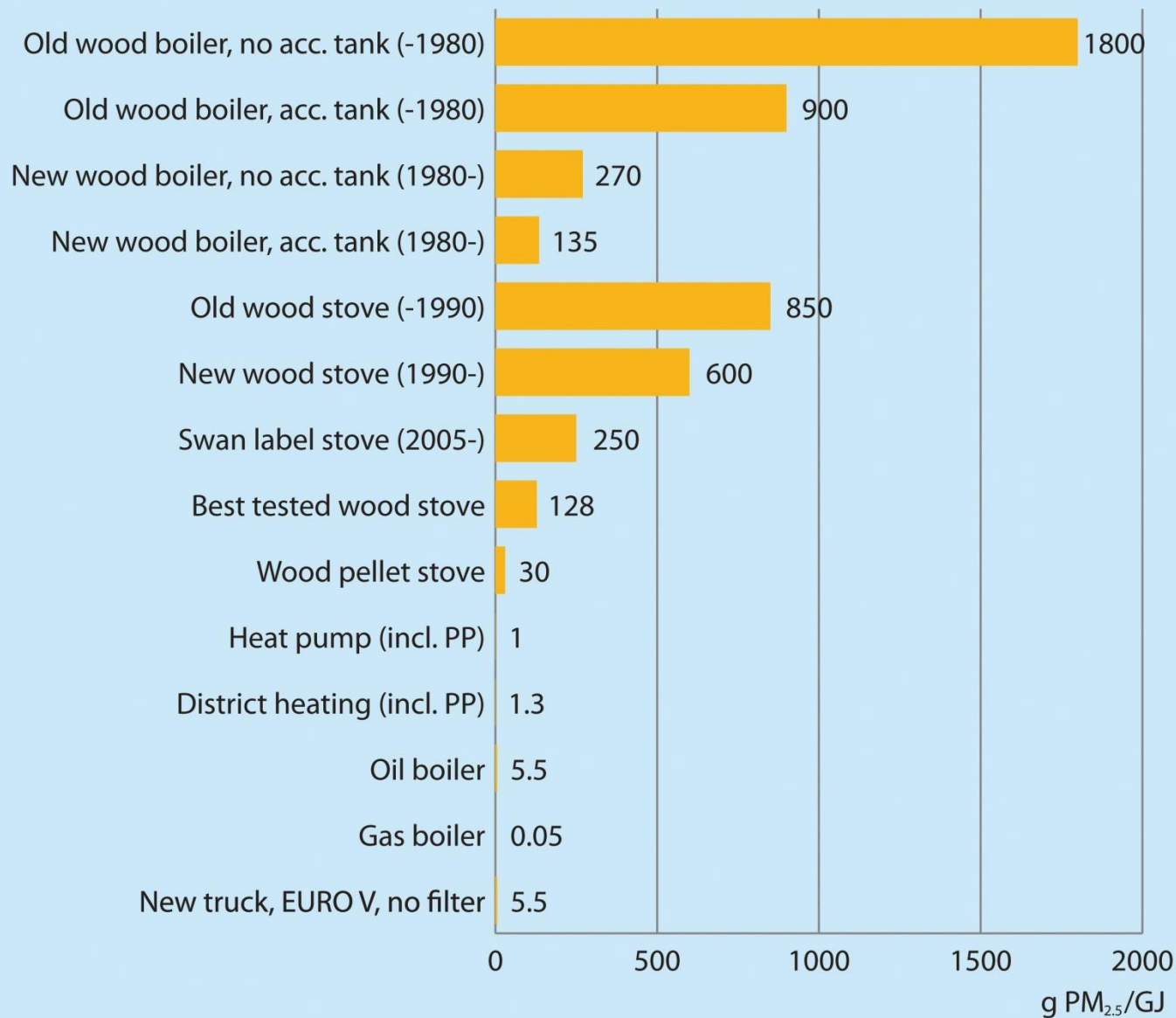
PM$_{2.5}$ emission levels including condensates (Norwegian standard NS 3058-2) from boilers and stoves compared to other heat sources. In comparison, the emission level of a truck without filter (EURO V) is included.

Annual mean concentration of benzo(a)pyrene (ng/m³), 2011 (EEA, 2013a)
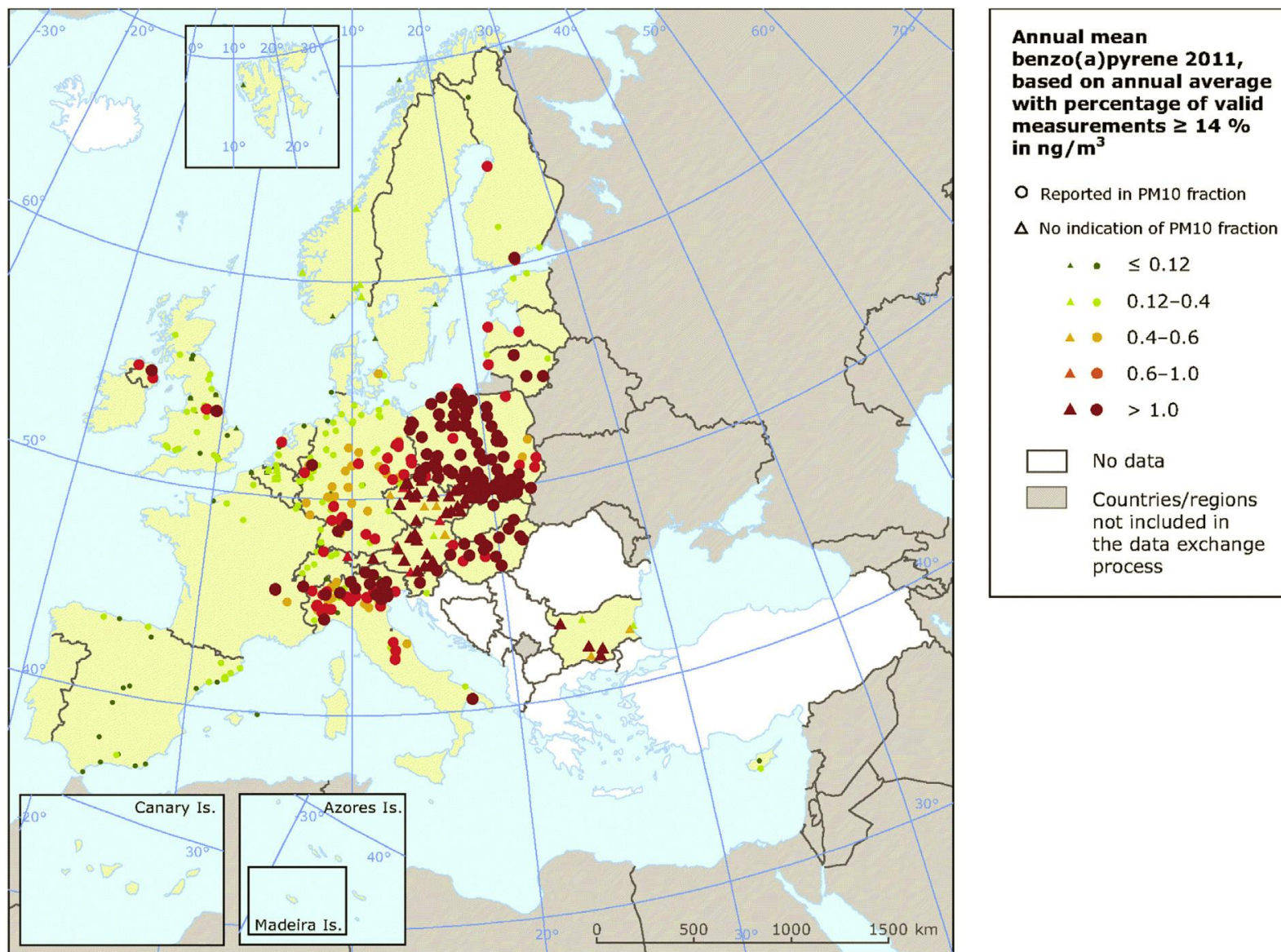
AIR QUALITY POLLUTANTS

Polycyclic aromatic hydrocarbons

❑ The EU air quality target (1 ng m^{-3} of benzo(a)pyrene) equates to a lifetime risk of 1 in 10^4 which is 10 to 100-fold higher than that normally considered tolerable in regulatory toxicology

SPECIFIC SOURCES: COOKING AEROSOL

- Studies using Aerosol Mass Spectrometers (AMS) have highlighted this source.

- There are indications that the AMS data over-estimate the mass of cooking aerosol.

- Nonetheless, this is a source which cannot be ignored.

Median Diurnal Profiles of the Factors from the Three Campaigns
(from J.D. Allan et al., ACP, 10, 647-668, 2010)
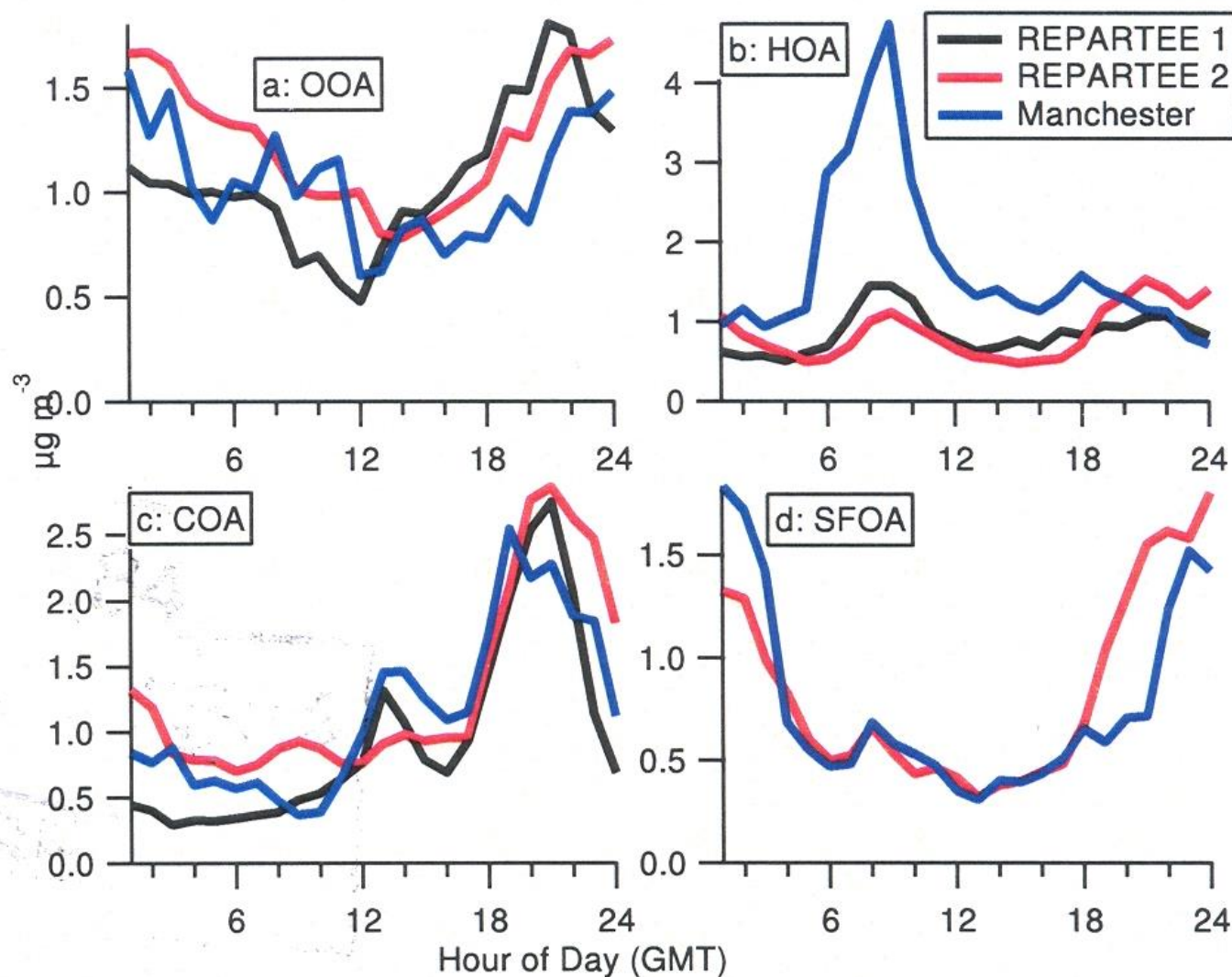
SPECIFIC SOURCES: NON-EXHAUST EMISSIONS FROM ROAD TRAFFIC

- Emission inventories include tyre wear, brake wear and road surface wear. They do not include particle resuspension.

- Currently, non-exhaust emissions of PM_{10} are of a similar magnitude to exhaust emissions. By 2020, non-exhaust emissions will be strongly dominant.

- This source contributes similar masses of particles to the fine ($PM_{2.5}$) and coarse ($PM_{2.5-10}$) fractions.

- There are no current measures in place, or planned, to control emissions from this source.

ELEMENTAL DATA AS TRACERS OF NON-EXHAUST EMISSIONS

Examine:

- Relationship between metals to identify those with a common source

- Consider typical chemical origins of metals

- Fe, Cu, Sb and Ba characteristic of brake dust

- Al, Si, Ca, Ti are typically crustal and likely to arise from soil or resuspension

- Size distributions are indicative of source

MASS RECONSTRUCTION

- **Assumes**

  Brake dust = Ba x 91

  Tyre dust = Zn x 50

  Resuspension = Si x 3.6

- **Gives contributions to mass of 0.9 – 11.5 μm particles**

  Brake dust = 55.3 ± 7.9%

  Tyre dust = 10.7 ± 2.3%

  Resuspension = 38.1 ± 7%

Estimation of the Contribution of Brake Dust, Tire Wear and Resuspension to Nonexhaust Traffic Particles Derived from Atmospheric Measurements, R.M. Harrison, A. Jones, J. Gietl, J. Yin and D. Green, Environ. Sci. Technol., 46, 6523-6529 (2012).

# Mass reconstruction - difference (all available data)
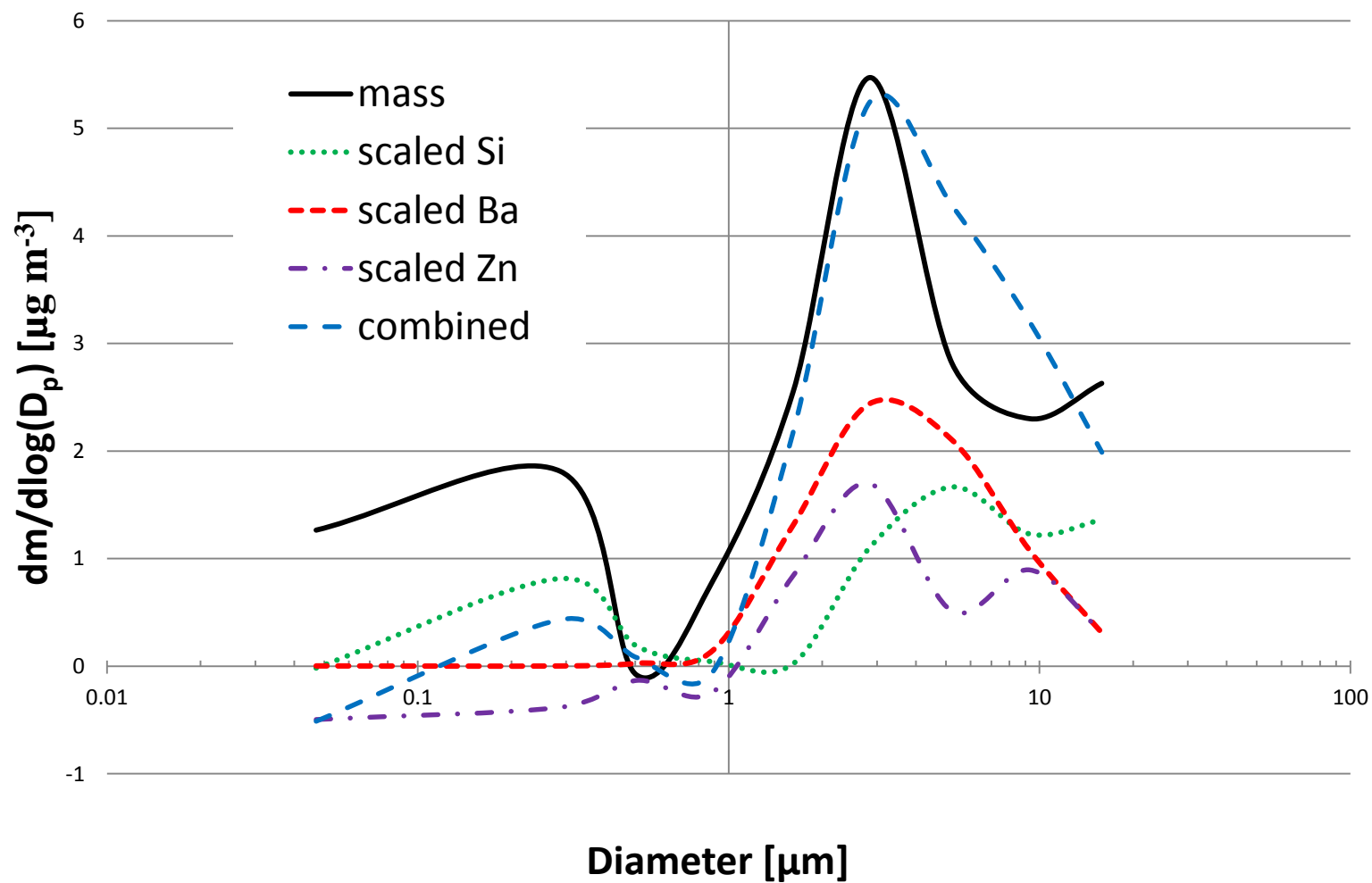
SPECIFIC SOURCES: SECONDARY PARTICLES

- Sulphates and nitrates arise from the oxidation of sulphur dioxide and nitrogen dioxide respectively, and the precursor-secondary pollutant relationships appear to be strongly non-linear.

- Abatement of ammonia (largely arising from agriculture) would be an effective way of slowing the oxidation of sulphur dioxide and hence reducing the formation of sulphate particles. It would also reduce nitrate particle formation.

- Most secondary organic aerosol derives from biogenic precursors, and is therefore unlikely to be subject to abatement measures.
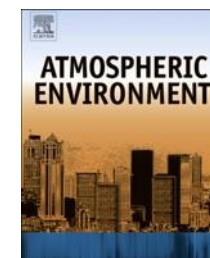
The effect of varying primary emissions on the concentrations of inorganic aerosols predicted by the enhanced UK Photochemical Trajectory Model

Roy M. Harrison[*], Alan M. Jones, David C.S. Beddows, Richard G. Derwent [1]

National Centre for Atmospheric Science, Division of Environment Health & Risk Management, School of Geography, Earth & Environmental Sciences, University of Birmingham, Edgbaston, Birmingham B15 2TT, United Kingdom

Predicted sulphate as a function of SO_2 at Harwell (SO_2 emissions varied in UK and Europe)
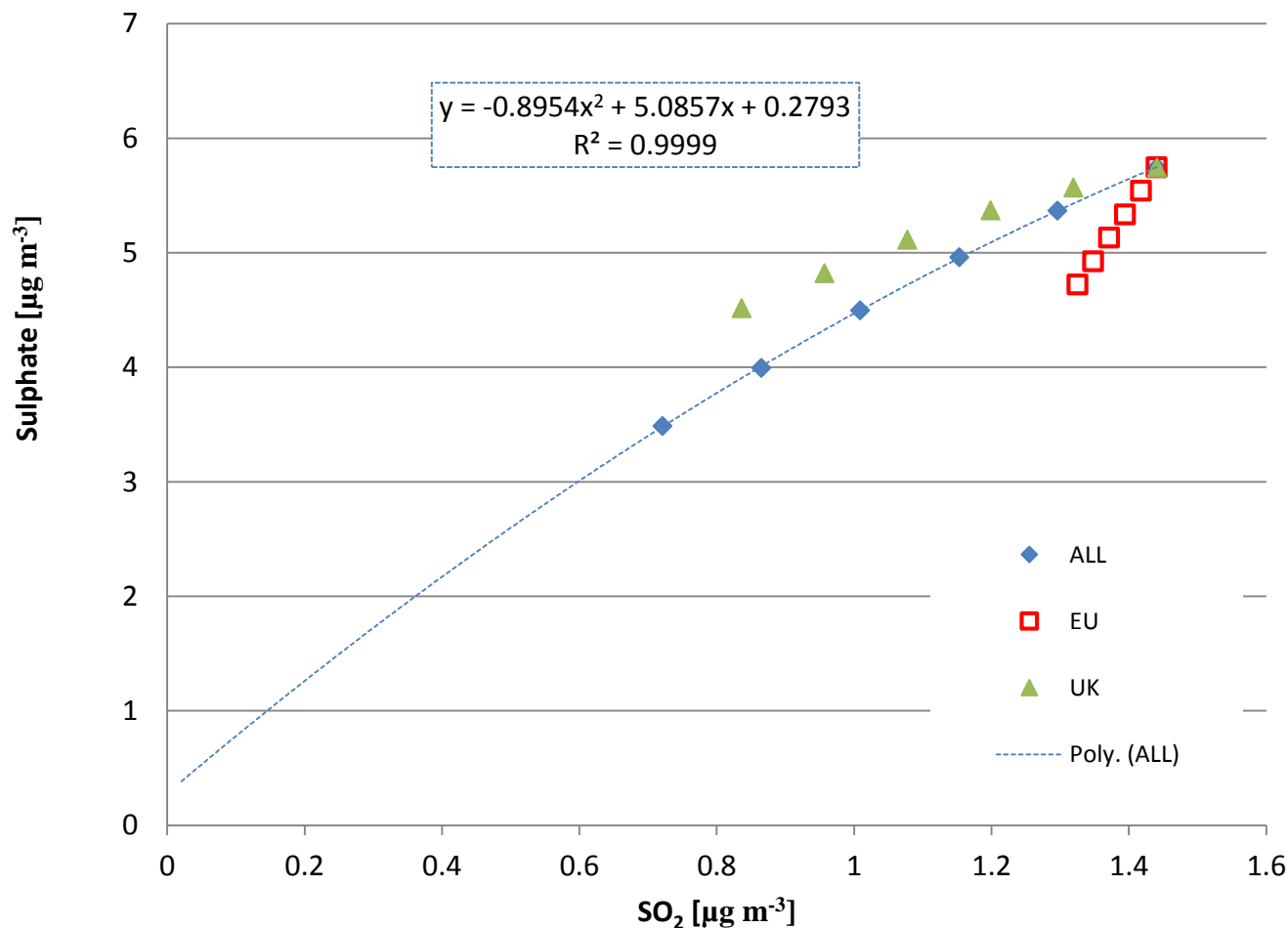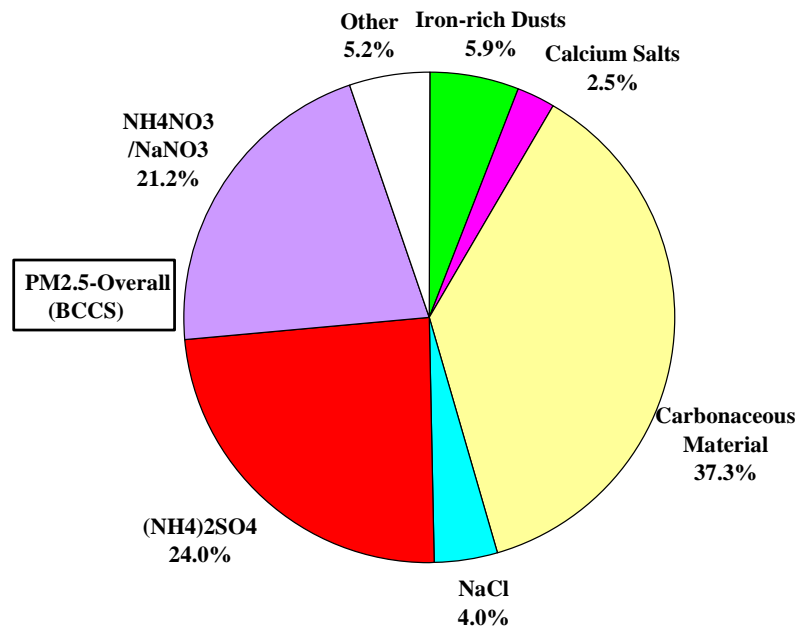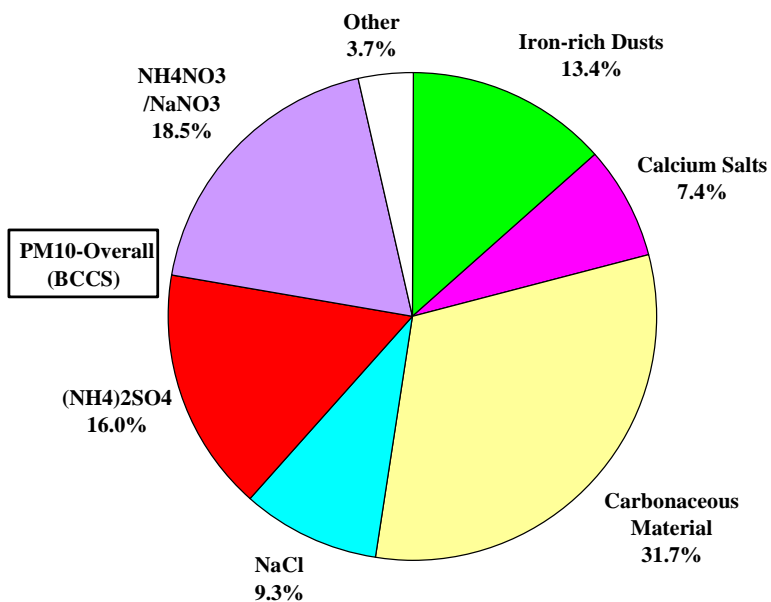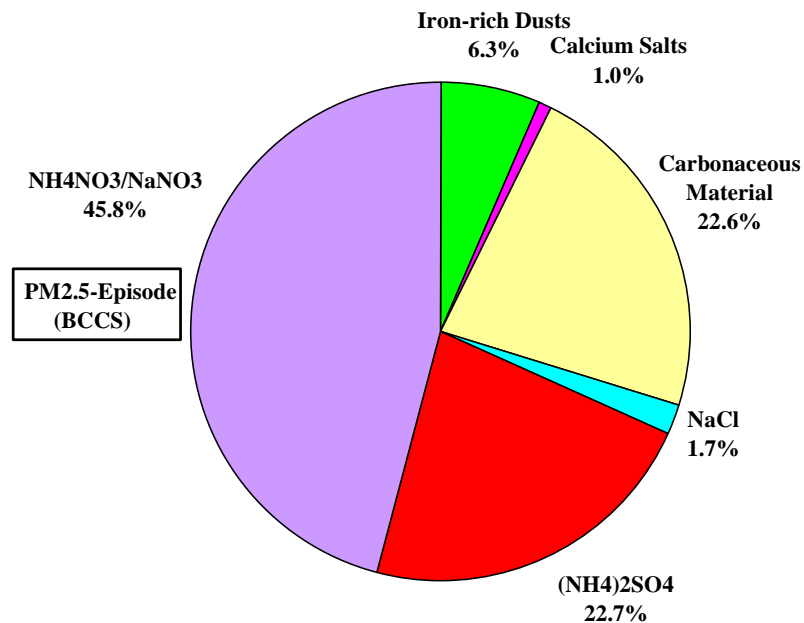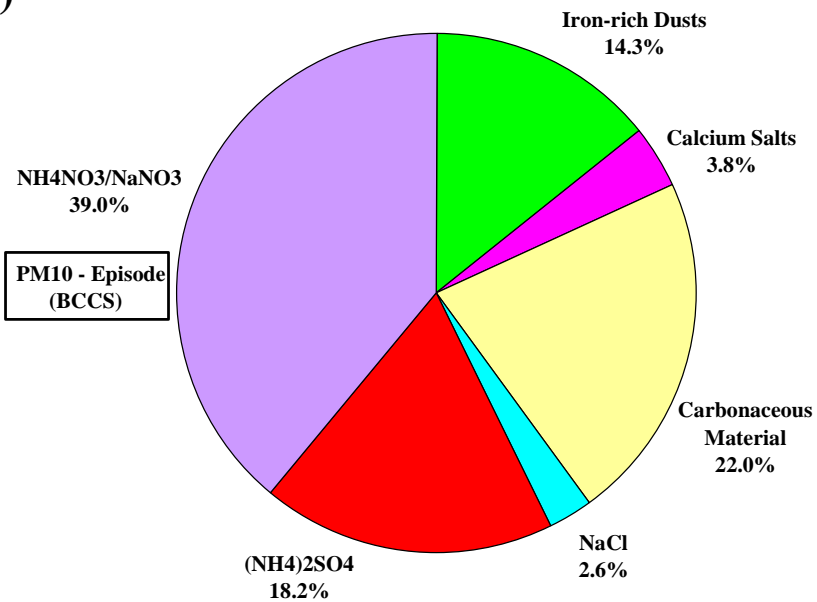
Days with Concentrations of PM$_{10}$ > 50 µg m^{-3}

- **As in earlier work, the component showing the greatest enhancement in concentration on high pollution days is nitrate in both PM$_{10}$ and PM$_{2.5}$**
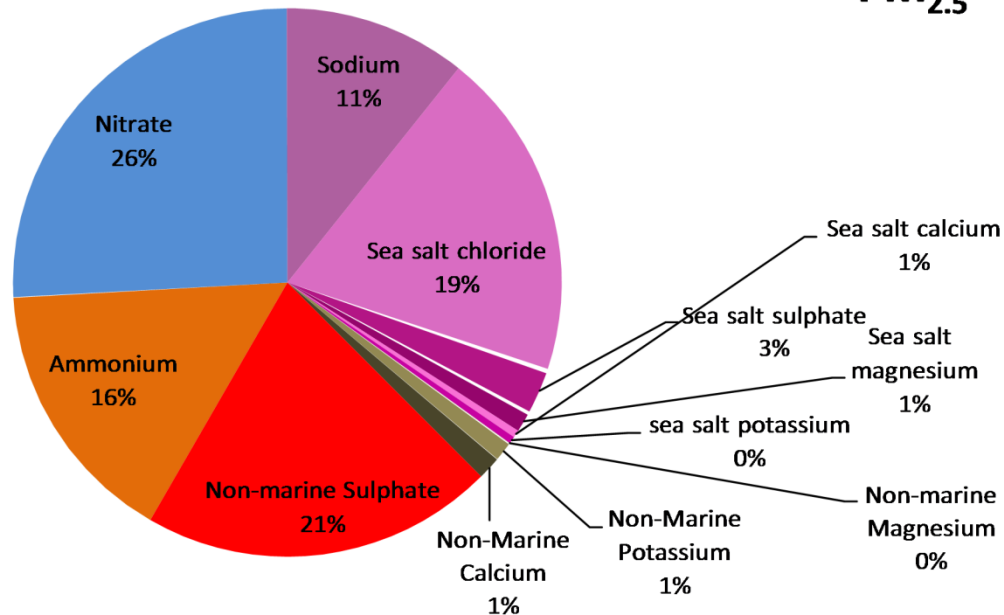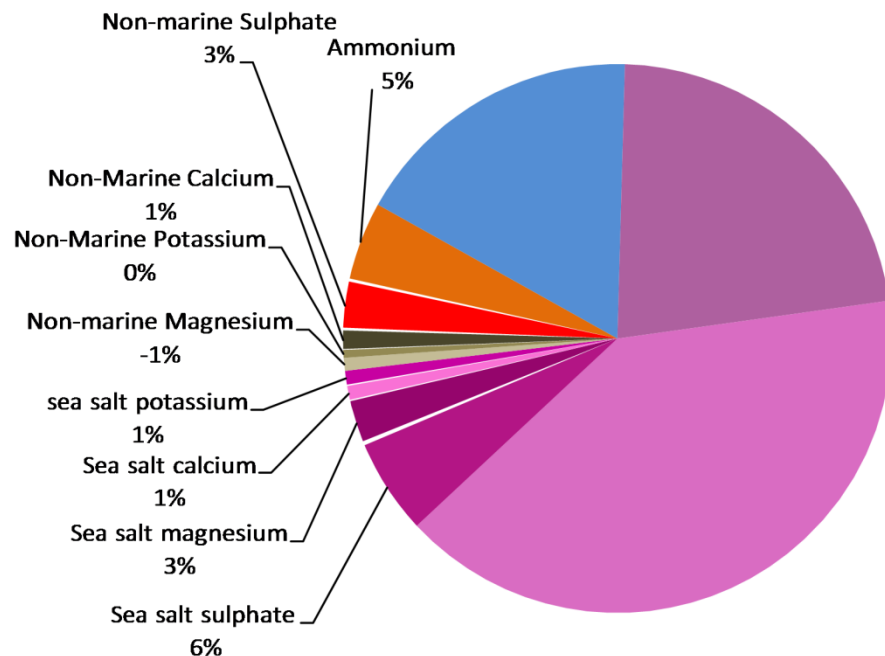
(a)

PM10-Overall (BCCS)
- Iron-rich Dusts 13.4%
- Calcium Salts 7.4%
- Carbonaceous Material 31.7%
- NaCl 9.3%
- (NH4)2SO4 16.0%
- NH4NO3/NaNO3 18.5%
- Other 3.7%

PM2.5-Overall (BCCS)
- Other 5.2%
- Iron-rich Dusts 5.9%
- Calcium Salts 2.5%
- Carbonaceous Material 37.3%
- NaCl 4.0%
- (NH4)2SO4 24.0%
- NH4NO3/NaNO3 21.2%

(b)

PM10 - Episode (BCCS)
- Iron-rich Dusts 14.3%
- Calcium Salts 3.8%
- Carbonaceous Material 22.0%
- NaCl 2.6%
- (NH4)2SO4 18.2%
- NH4NO3/NaNO3 39.0%

PM2.5-Episode (BCCS)
- Iron-rich Dusts 6.3%
- Calcium Salts 1.0%
- Carbonaceous Material 22.6%
- NaCl 1.7%
- (NH4)2SO4 22.7%
- NH4NO3/NaNO3 45.8%

AUCHENCORTH MOSS, 2007-2012
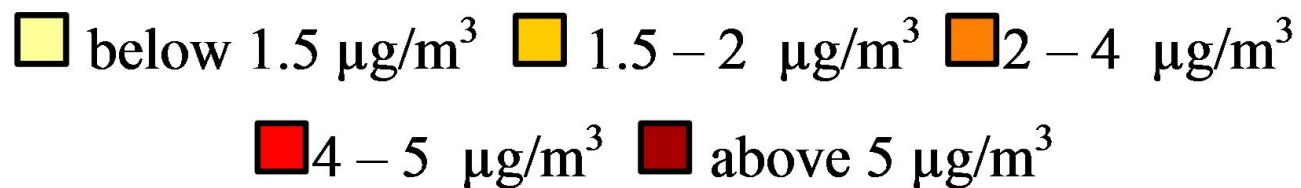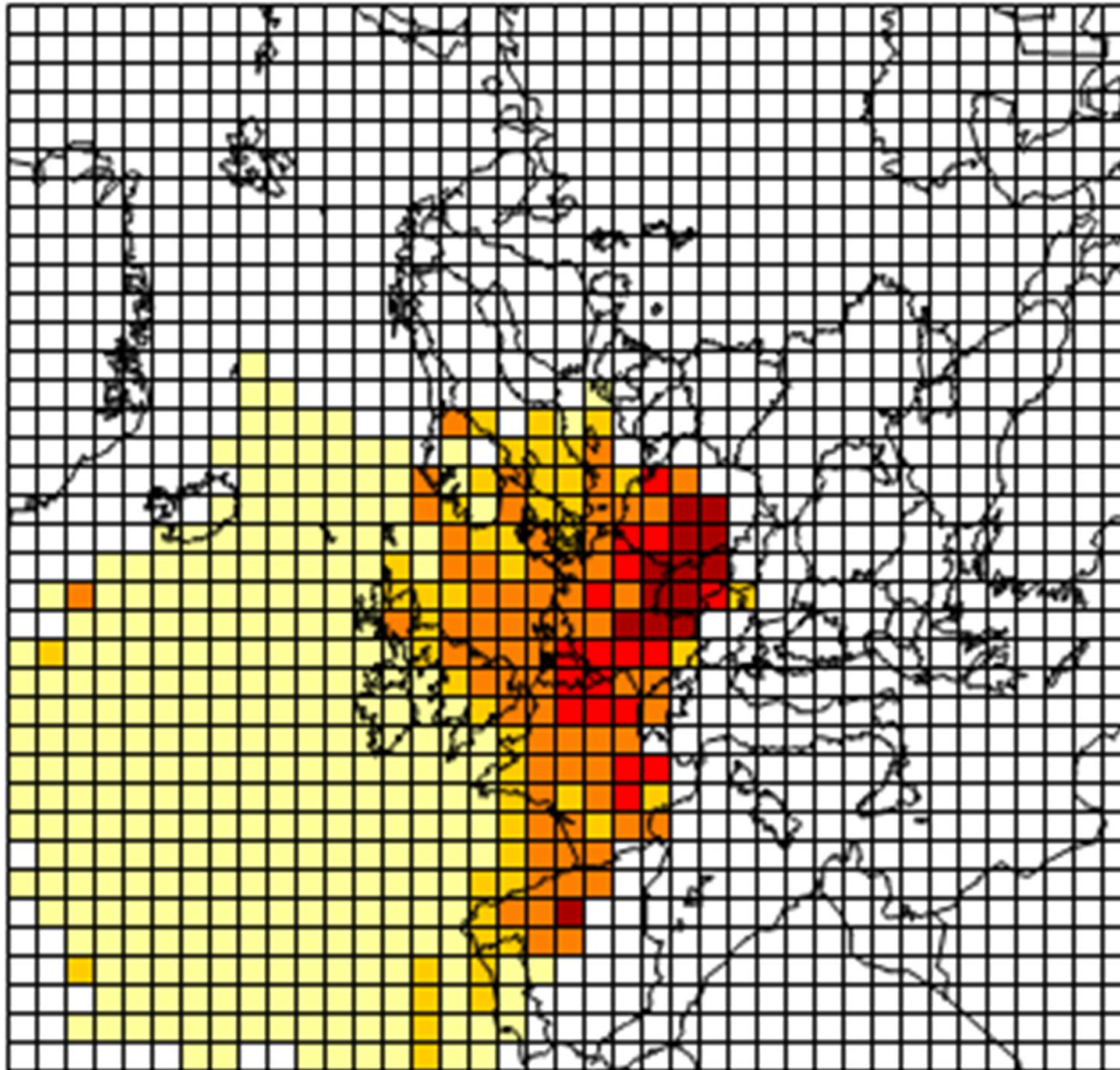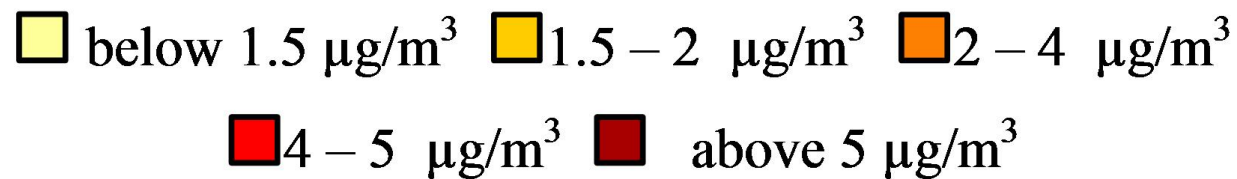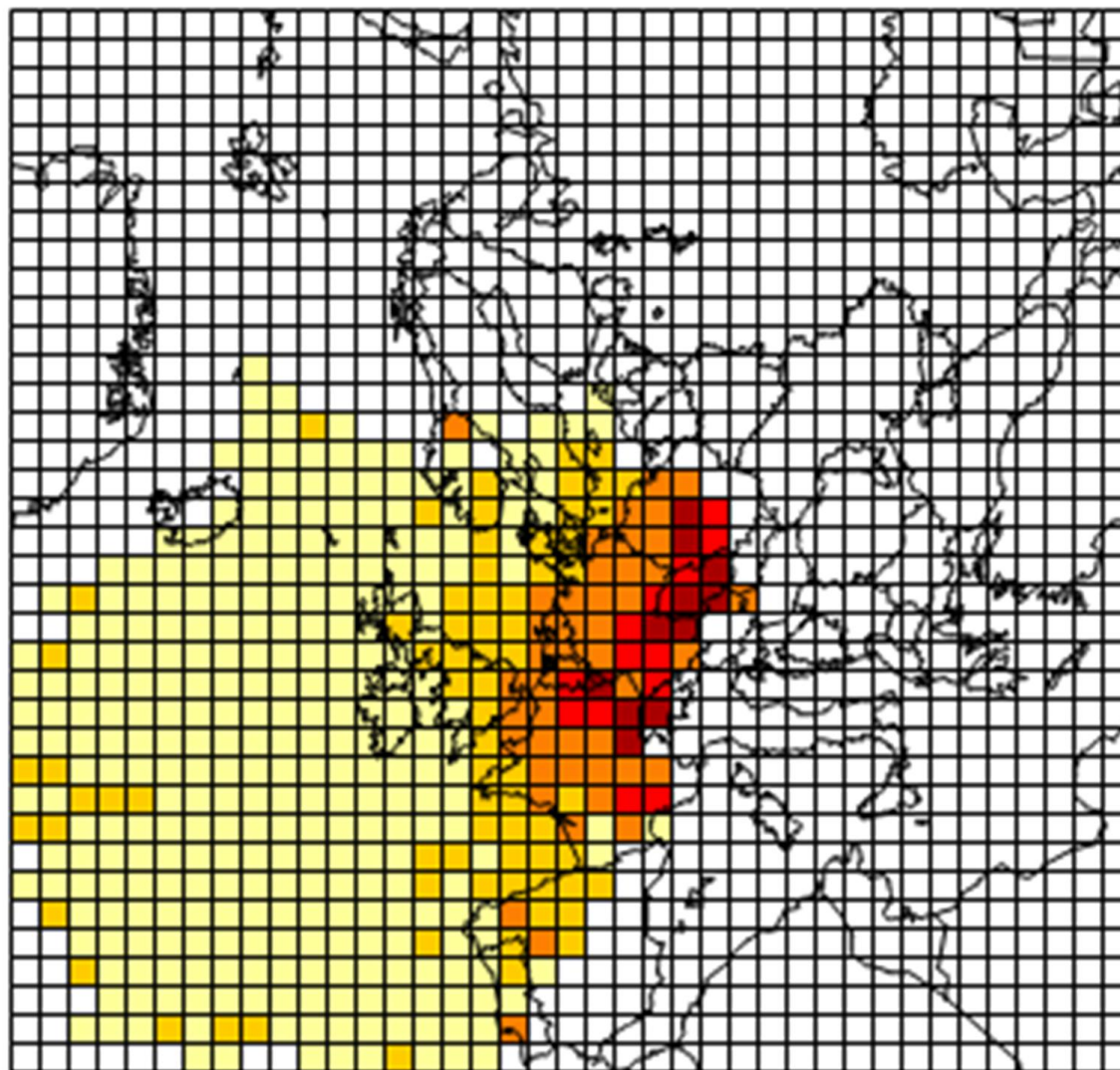

PM$_{2.5}$

PM$_{coarse}$

Average composition by mass of the water soluble inorganic aerosol fraction measured by the MARGA from January 2007 to December 2012 in both the PM$_{2.5}$ and PM$_{coarse}$. (from Twigg et al., ACPD, 3703-3743, 2015).
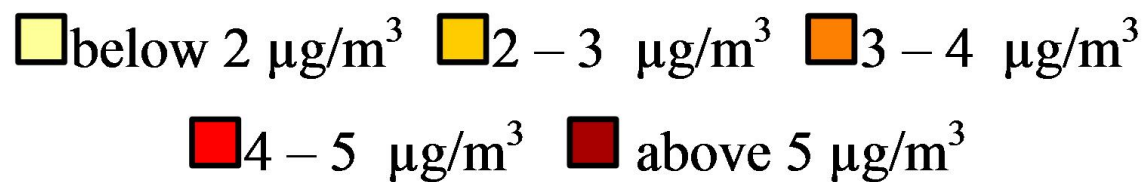
NITRATE AT HARWELL

- ☐ below 1.5 μg/m^3
- ☐ 1.5 – 2 μg/m^3
- ☐ 2 – 4 μg/m^3
- ☐ 4 – 5 μg/m^3
- ☐ above 5 μg/m^3

SULPHATE AT HARWELL

below 1.5 µg/m³ 1.5 – 2 µg/m³ 2 – 4 µg/m³

4 – 5 µg/m³   above 5 µg/m³

PARTICULATE ORGANIC CARBON AT HARWELL

below 2 µg/m³   2 – 3 µg/m³   3 – 4 µg/m³

4 – 5 µg/m³   above 5 µg/m³
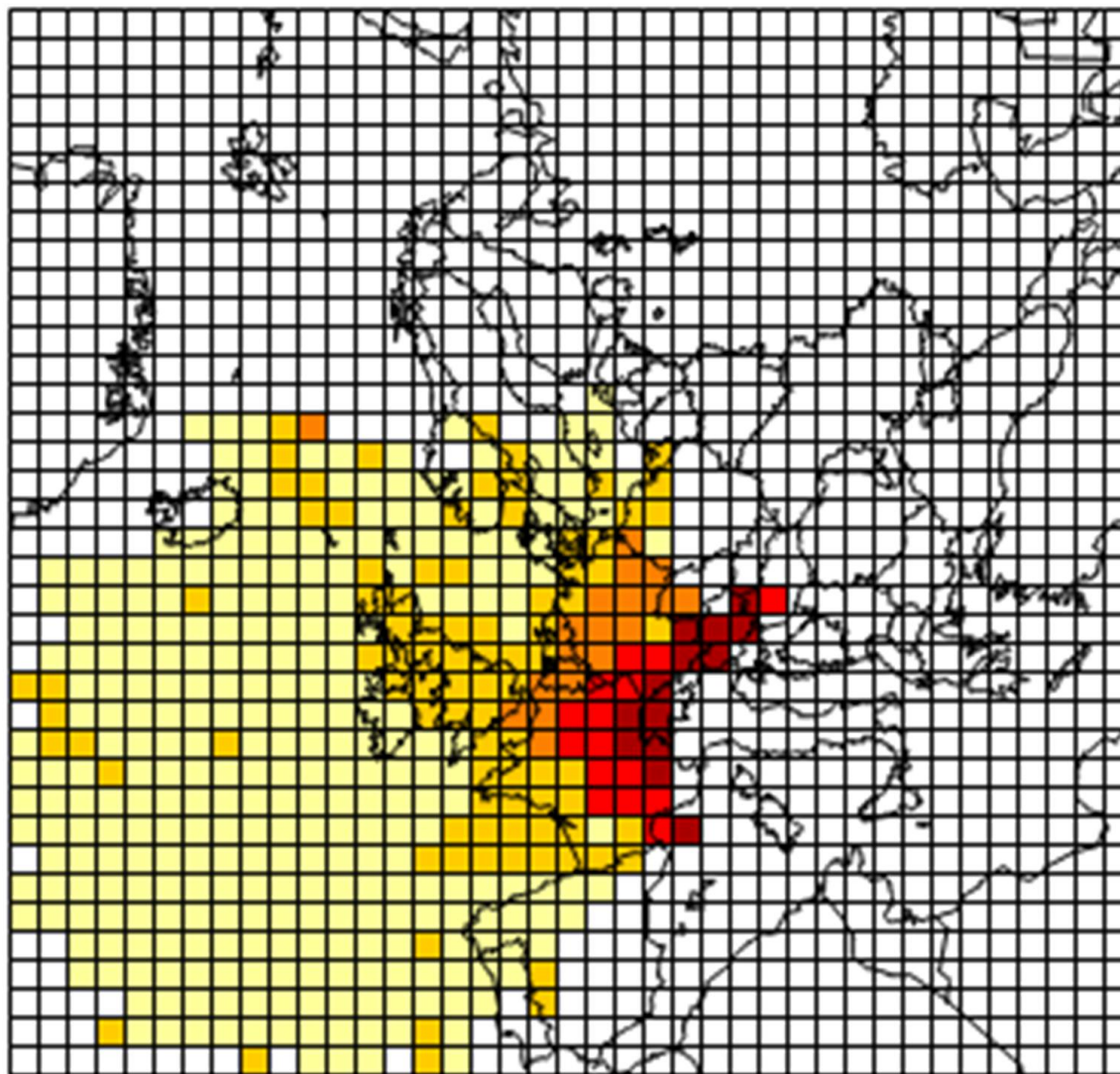
EXPOSURE REDUCTION

- The EU approach to policy on $PM_{2.5}$ has adopted the exposure reduction concept alongside traditional Limit Values.

- Exposure reduction implies maximising the function:

$$\sum^{i} \Delta PM_i \cdot N_i$$

- where Δ PM_i is the reduction in PM concentration in grid square i, and N_i is the population of grid square i.

- This implies that the most cost-effective policies **may** be those that focus upon reduction of urban concentrations, because of the large values of N_i in cities.

CONCLUSIONS

- Knowledge is improving of the sources contributing to $PM_{2.5}$ and PM_{10} in UK air.

- Sources such as wood smoke and cooking aerosol are significant and may lead to problems with abatement policies.

- Secondary particles are dominant and provide especially difficult mitigation issues due to non-linearity (sulphate and nitrate) and biogenic sources (organic aerosol).

- There are few local policy levers which reduce PM concentrations appreciably.

THANK YOU