Regional Scale Modelling of Particulate Matter in the UK: Source Attribution & Future Trends

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Background

- Fine particles (PM_{10} or finer) have known health effects on cardiovascular and respiratory systems
- Focus for air quality standards nationally and internationally
- Effects can be driven by short term (e.g. episodes) or long term (chronic) exposure
- UK Air Quality standards now differ across the country. Annual mean target concentrations for 2010:

  - England (except London), Wales and Northern Ireland 20 $\mu g\ m^3$
  - London 23 $\mu g\ m^3$
  - Scotland 18 $\mu g\ m^3$

Introduction to PM_{10}

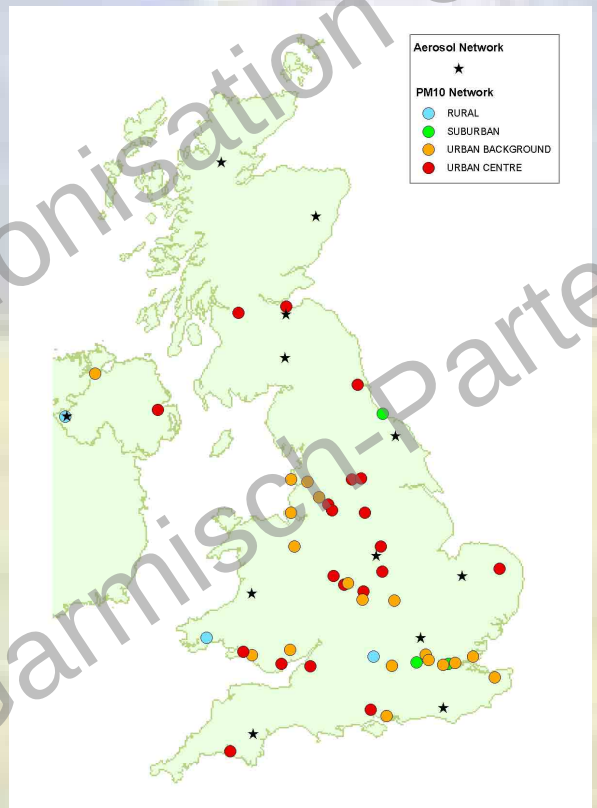
- PM_{10} comprises a range of materials:

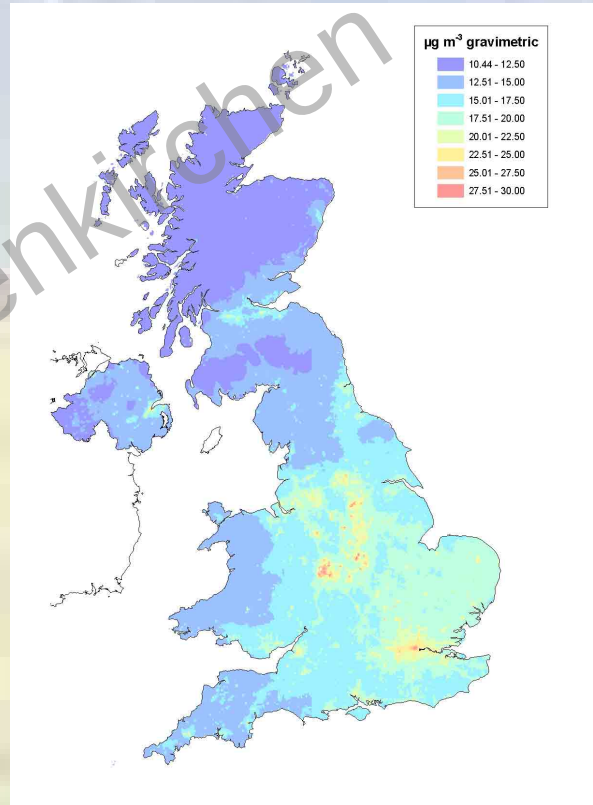
  - primary particles (e.g. black carbon from combustion, road traffic, metals)
  - secondary particles (inorganic and organic)
  - soils, dust, sea salt (coarse i.e. > 2.5 $\mu g\ m^3$)
  - biological particles

- Increasing interest in the fine fraction ($PM_{2.5}$) which appears to have a distinctive composition from PM_{10} and may have greatest health effects

PM_{10} in the UK

- Netcen estimates of PM_{10} for the UK are based on:
  - measured PM_{10} values
  - secondary inorganic aerosol from monitoring sites
  - a constant coarse particle contribution (8.8 μg m^3)
  - roadside increment*





* excluded on the above map

What is Missing?

- Secondary organic aerosol (SOA) is not included in netcen estimates of PM_{10}

- Is also missing from the EMEP model
  - incomplete understanding of formation processes
  - little observed data against which to validate model estimates

- SOA may account for a significant proportion of PM_{10} mass concentration in summer episodes

Increased significance in the future as anthropogenic emissions decline?

Do trees pollute the atmosphere?

Tim Radford
Thursday May 13, 2004

The Guardian

Yes, just as president Ronald Reagan said in 1981. "Trees cause more pollution than automobiles do," he opined. A little later, environmental scientists ruefully confirmed he was partially right. In hot weather, trees release volatile organic hydrocarbons including terpenes and isoprenes - two molecules linked to photochemical smog. In very hot weather, the production of these begins to accelerate.

America's Great Smoky Mountains are supposed to take their name from the photochemical smog released by millions of hectares of hardwoods.

Modelling PM_{10} for the UK

- Need to understand which sources contribute to:
  - develop policy to meet target concentrations for PM_{10} and $PM_{2.5}$
  - improve our assessment of the impact of abatement measures (on different size fractions and chemical components)

- Models can be used to estimate primary, secondary inorganic (SIA) and secondary organic aerosol (SOA) from different source categories

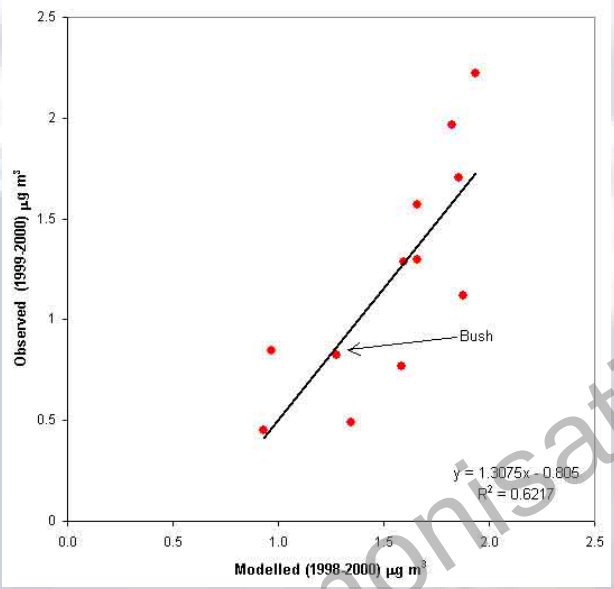
- Modelled concentrations can be compared with site measurements, netcen estimates and other model output (e.g. EMEP, NAME)

Modelling PM_{10} for the UK

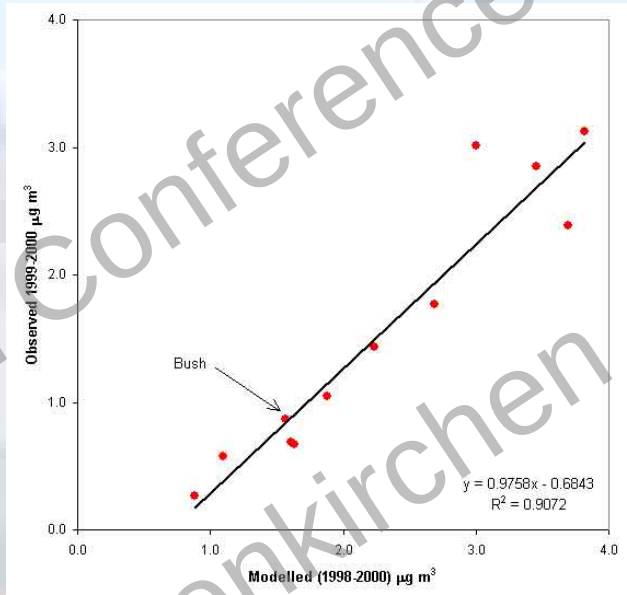
- **HARM** used to derive primary PM_{10} and SIA (SO_4, NO_3, NH_4, Cl)
- **ELMO** used to derive SOA through the photo-oxidation of terpenes (represented by α-pinene)
- Aerosol water added to primary component ($0.29*(SO_4+NO_3)$)
- Coarse component (8.8 μg m^3 gravimetric) added to the sum of primary PM_{10} + SIA + SOA
- Validation against:
  - nitric acid and aerosol network (12 sites, rural)
  - EC/OC campaign data (1 site, rural)
  - PM_{10} network (39 sites, mainly urban)
  - netcen annual mean background estimate (UK at 1km resolution)

HARM (1998-2000) vs. Observed SIA (1999-2000)

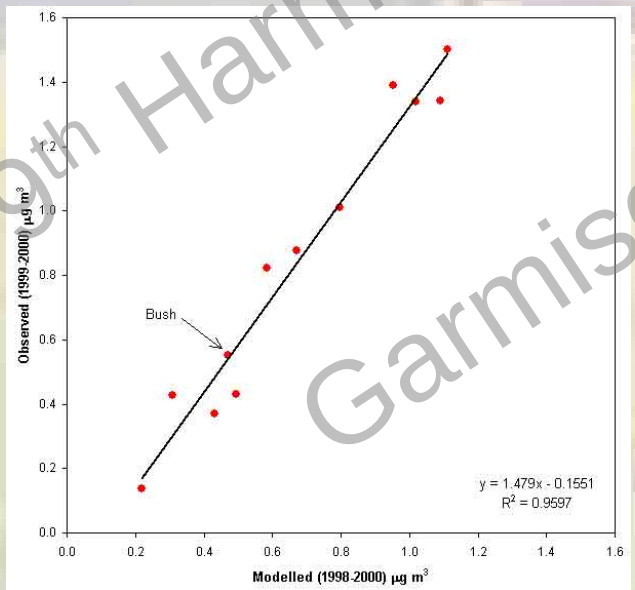
SO_4



NO_3



NH_4



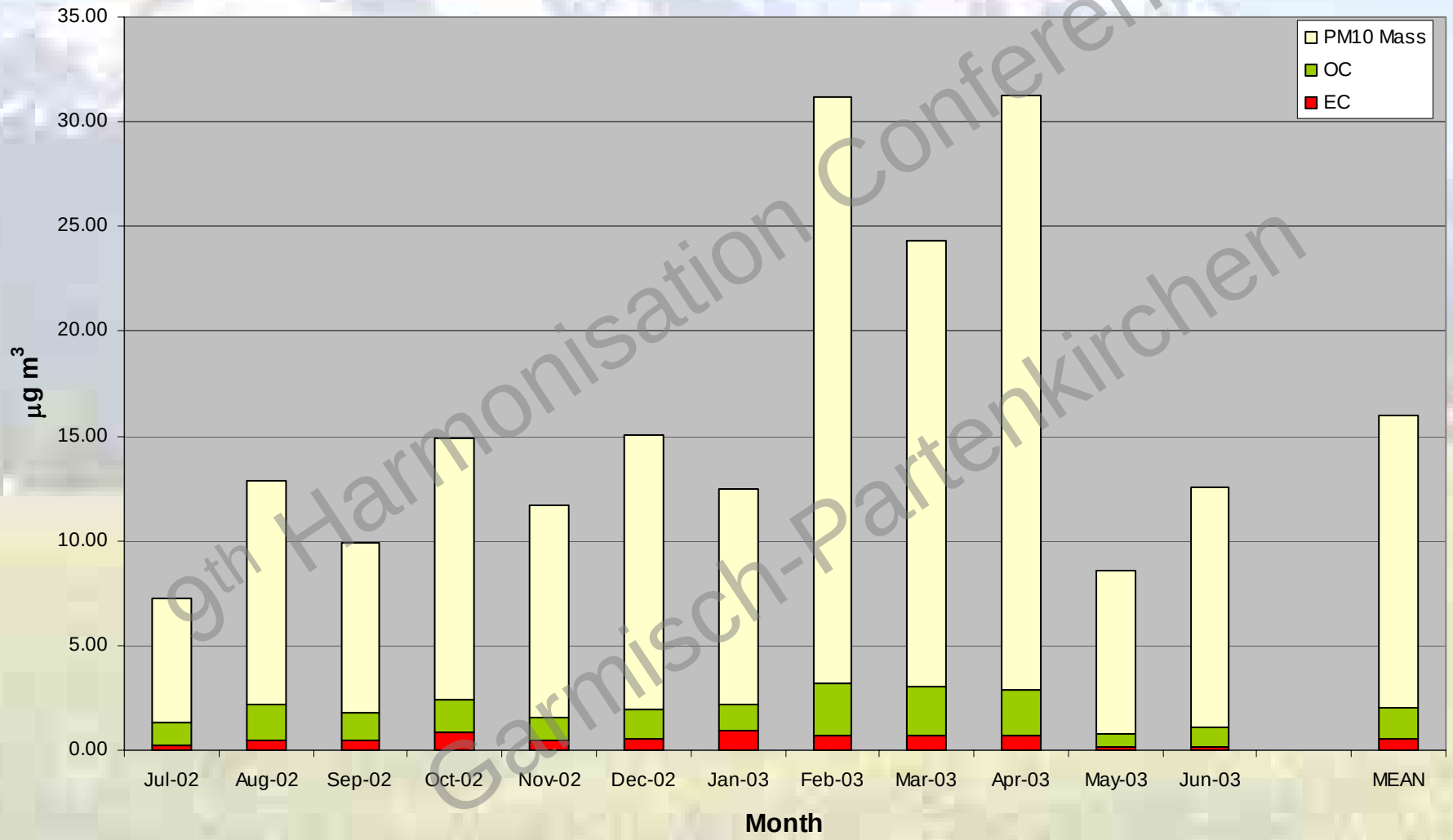
Good level of agreement with observed SO_4, NO_3 and NH_4 over the period 1999-2000.

SO_4	$R^2 = 0.62$
NO_3	$R^2 = 0.90$
NH_4	$R^2 = 0.96$

Deriving Estimates of SOA from EC/OC Measurements

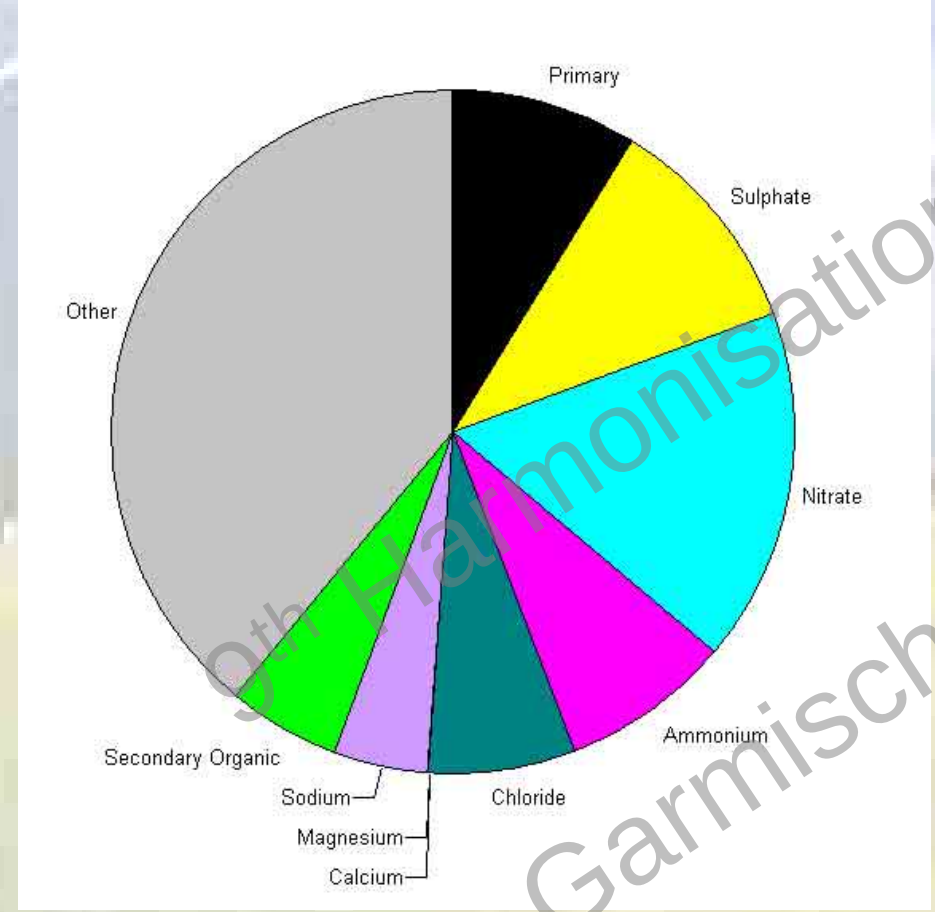
- Elemental Carbon and Organic Carbon measurements taken weekly at Bush Estate as part of wider EMEP Campaign
- OC is a mixture of primary and secondary organic matter
- The typical OC/EC ratio in primary emissions is close to 1:1 and is assumed to be represented by winter measurements
- Subtracting the winter OC/EC ratio from the summer OC/EC ratio reflects the secondary component of OC
- This approach has been applied to data from the Bush Estate to derive an annual mean estimate of SOA for comparison with ELMO

EC, OC and PM_{10}, Bush Estate (2002-2003)



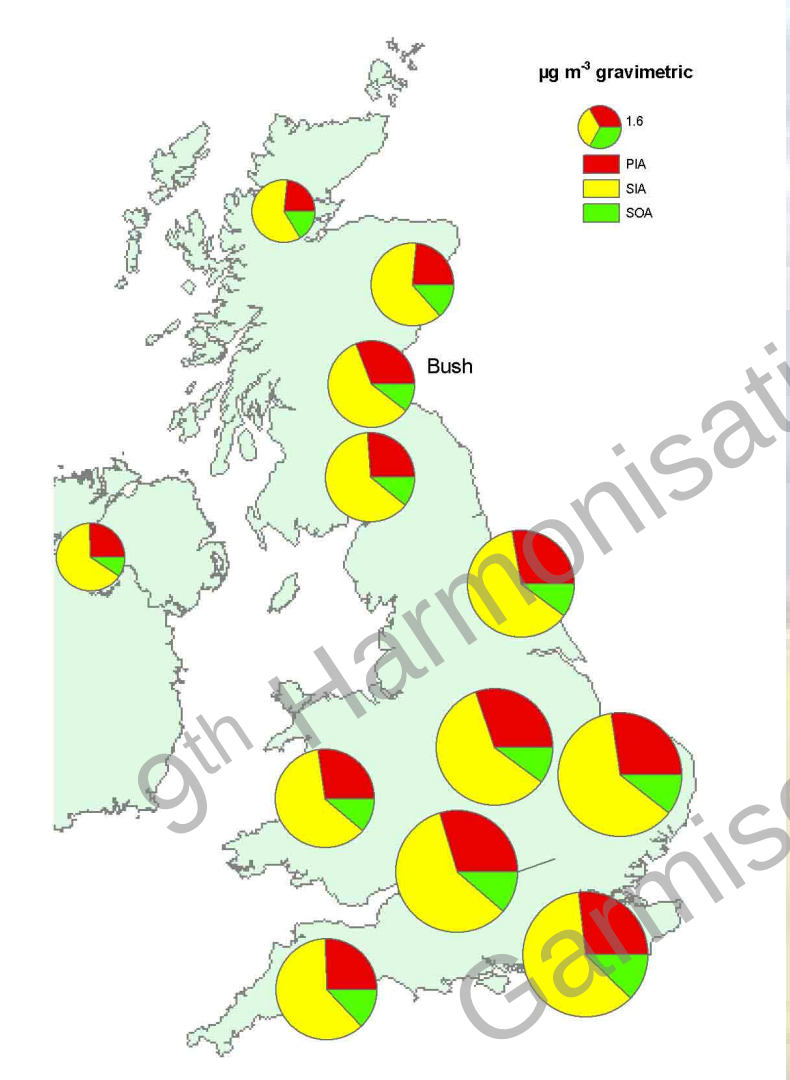
Mean Annual Particle Composition, Bush (2002-2003)

Component	Observed (2002-3)	Modelled (1999)
Primary	1.28 μg m^3	1.73 μg m^3
Sulphate	1.54 μg m^3	1.21 μg m^3
Nitrate	2.45 μg m^3	1.58 μg m^3
Ammonium	1.18 μg m^3	0.47 μg m^3
Chloride	0.98 μg m^3	0.01 μg m^3
Secondary Inorganic	6.15 μg m^3	3.27 μg m^3
Secondary Organic	0.76 μg m^3	0.58 μg m^3
SUM	8.19 μg m^3	5.57 μg m^3
Total PM_{10}	13.9 μg m^3	



- Annual mean PM_{10} of 13.79 μg m^3
- Chemical composition known for 65% of mass (including Ca, Mg, Na)
- If we exclude the unknown element then SIA > Primary PM_{10} > SOA
- These ratios are reproduced at other rural aerosol network sites across the UK by the HARM-ELMO model combination

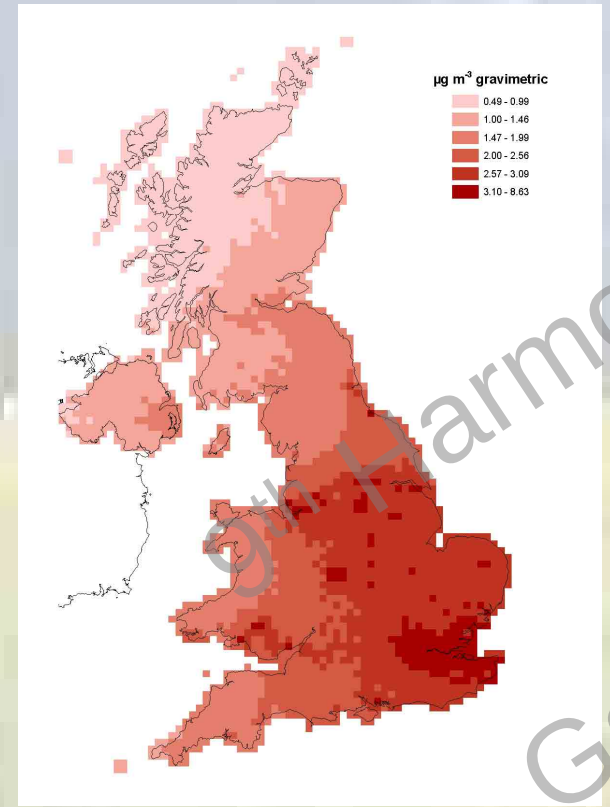
Modelled Source Attribution (1)



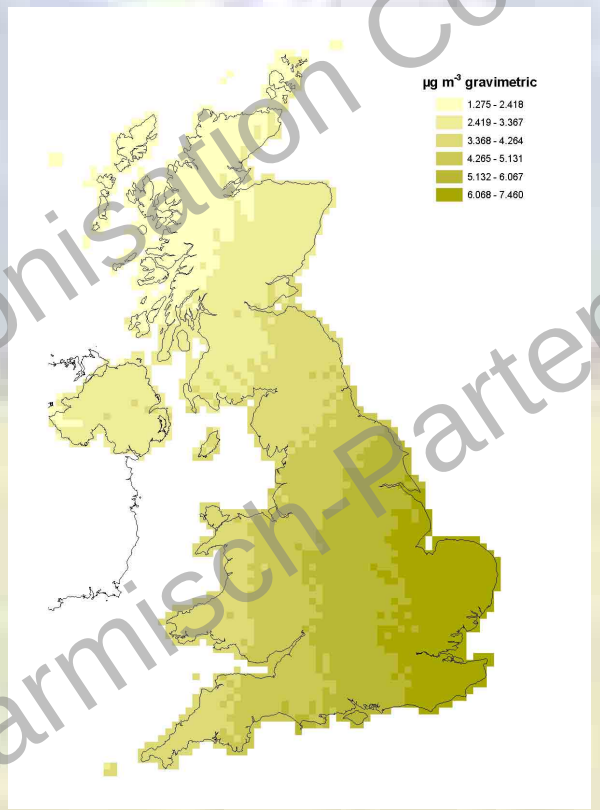
- HARM-ELMO indicates SIA > Primary PM_{10} > SOA across all rural aerosol network sites
- UK sources account for:
  - 40-75% of primary PM_{10}
  - 40-60% of SIA
  - 10-50% of SOA
- SOA accounts for 9-16% of total modelled PM_{10} (excluding coarse component)

Modelled Source Attribution (2)

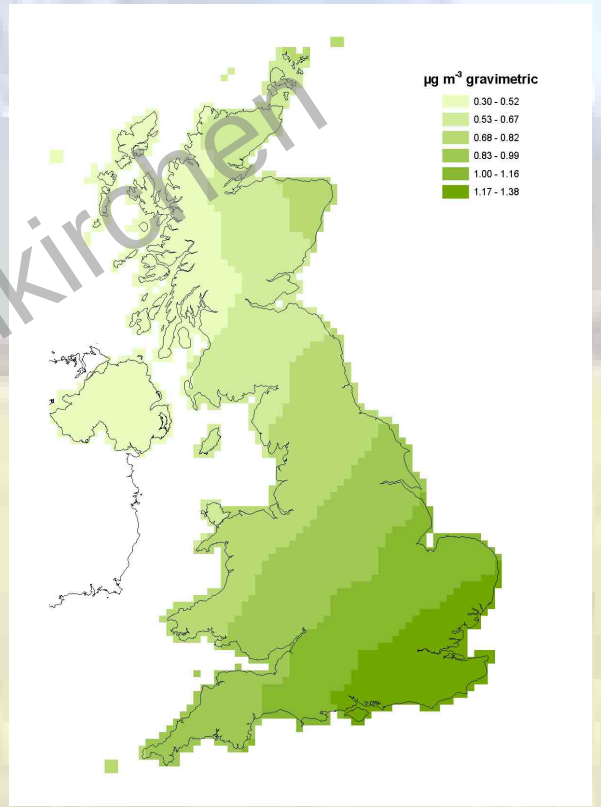
Primary



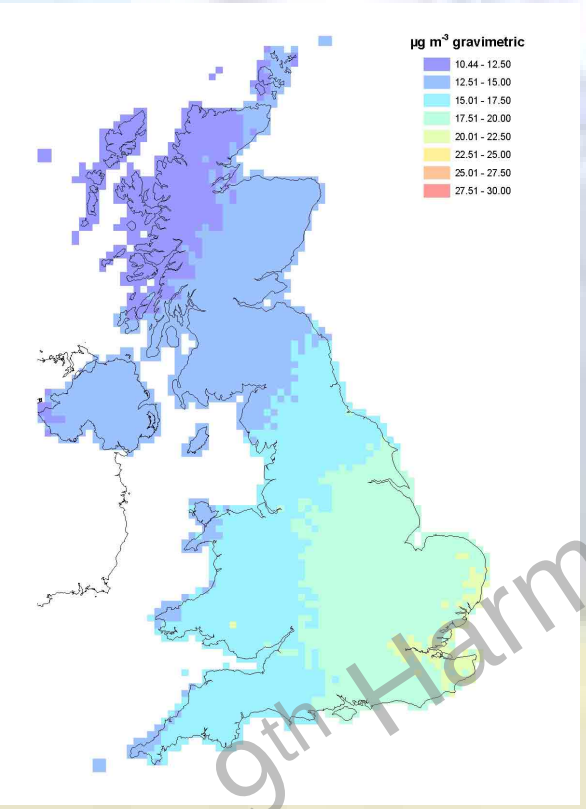
Secondary Inorganic

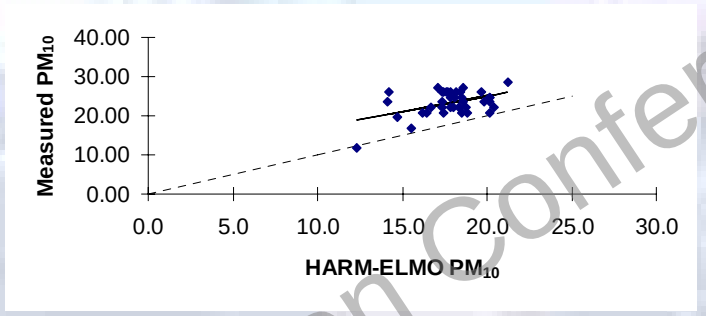


Secondary Organic

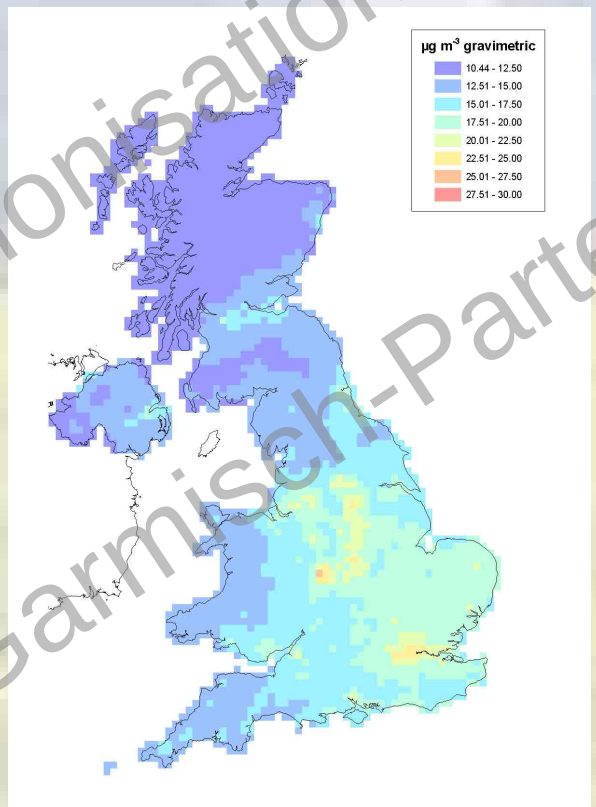


HARM-ELMO output at UK scale

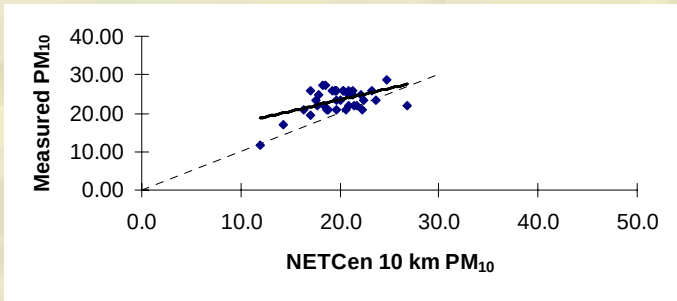




HARM-ELMO
R^2= 0.23
Slope = 0.80

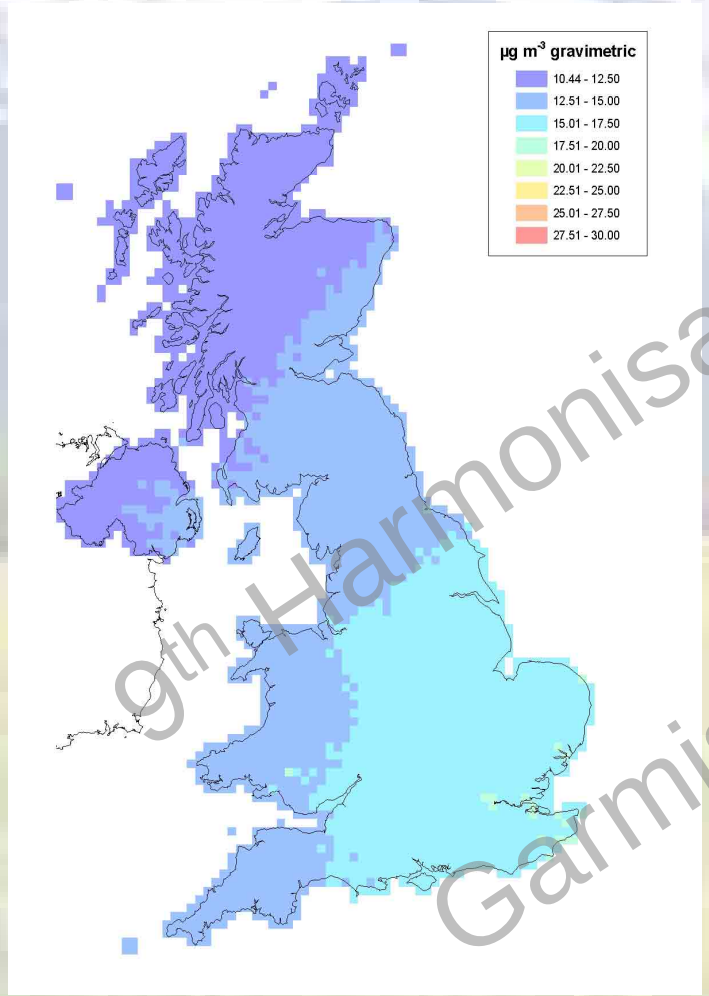


netcen (10km)
R^2 = 0.26
Slope = 0.59

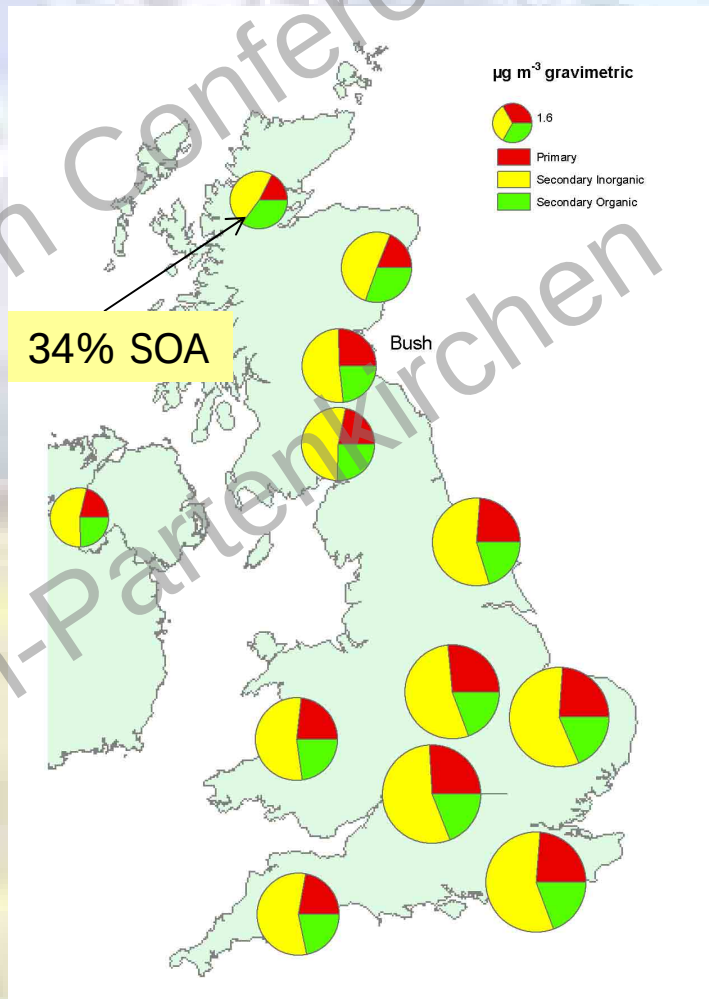


Model Applications: Scenarios and Sensitivity Studies

2010 Scenario (AQS)



2010 Scenario with GB Biogenics x 4



Conclusions

- Using HARM and ELMO we can model many components of PM_{10}
- Lack of validation data for some components
- Near mass closure for site where most data available
- Still under-estimating PM_{10} even with SOA contribution, most noticeably in urban areas
- Missing some known components (Ca, Na, K)
- Primary inventories (EMEP and UK) may not be accurate or complete?
- Model resolution may need to be improved?
- Scope for wide range of source attribution studies and exploration of changes in $PM_{2.5}$:PM_{10} through time

Acknowledgements

- Kjetil Torseth, Norwegian Institute for Air Research, for provision of EC/OC data for Bush Estate, 2002-2003

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