Measurements for the Assessment of UK Air Quality Models

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Overview

- Measurement activities supporting model development & validation:
  - EMEP Super-site 'Auchencorth'
  - EMEP European Intensive Campaigns
  - AMPEP round-Britain flights
  - Micrometeorological measurements of urban emission
- Modelling activities:
  - Interpretation of AMPEP flights using NAME
  - Interpretation of AMPEP flights using Models-3
  - UK Modelling using EMEP4UK

EMEP Measurement Programme

- UK is signatory to the UNECE Convention for Long-Range Transboundary Air Pollution (CLRTAP)
- Additional legislative framework to the EC Directives
- Regulation through 8 Protocols
  (e.g. Gothenburg Protocol (e.g. Nitrogen, S, VOC emissions, Heavy Metals, POPs, …)
- CLRTAP body EMEP: “Co-operative programme for monitoring and evaluation of long-range transmission of air pollutants in Europe”
  - Collection of official national emission inventories
  - Central modeling activities (e.g. Meteorological Synthesizing Centres East & West)
  - Collection of national monitoring data (measurement sites, Level-2 and Level-3 super-sites, intensive measurement periods)
  - Focused on rural and remote sites

UK EMEP Super-sites
Auchencorth Moss (& Harwell)

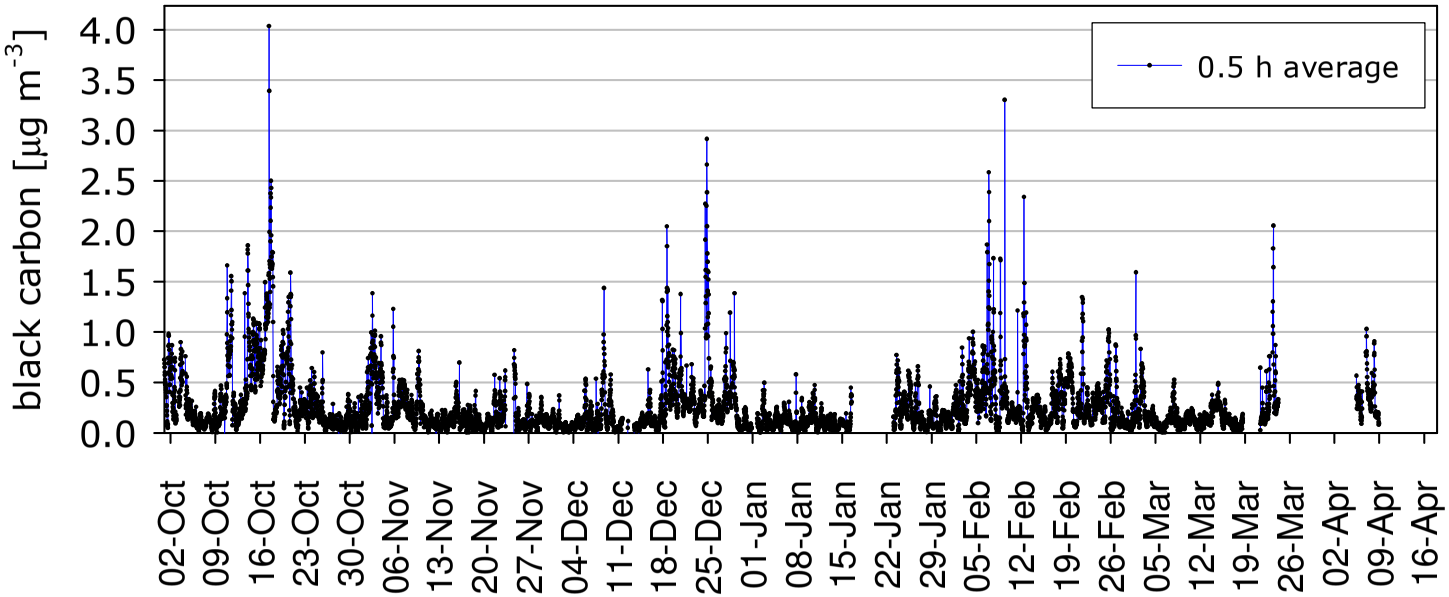
Measurements:

- NO, NO_2, O_3, SO_2 (hourly)
- Hg_0, Hg_p, RGM (hourly)
- Heavy metals (weekly)
- Wet deposition (daily)
- Reactive gases: NH_3, HNO_3, HCl, (HONO) (hourly)
- $PM_{2.5}$/PM_{10} mass (TEOM FDMS & gravimetric)
- Inorganic aerosol: NH_4^+, Na^+, Mg^{2+}, K^+, Ca^{2+}, NO_3^-, SO_4^{2-}, Cl^- in $PM_{2.5}$ and PM_{10} (hourly)
- BC (10 min)
- VOCs (hourly)
- Trace gas fluxes (NO, NO_2, O_3, SO_2, NH_3, CH_4, N_2O, CO_2)

Still coming:

- EC/OC
- Carbonyls
- Crustal (Si, Fe, Al)
- Aerosol size-distribution?

Black Carbon (Aethalometer)



MARGA (**M**onitoring instrument for inorganic **A**e**R**osol composition and acidifying **GA**ses)

Measured compounds:

- **Gas**: NH_3, HCl, HNO_3, HONO, SO_2
- **$PM_{2.5}$**: Cl^-, NO_3^-, SO_4^{2-}, NH_4^+, Na^+, K^+, Mg^{2+}, Ca^{2+}
- **PM_{10}**: Cl^-, NO_3^-, SO_4^{2-}, NH_4^+, Na^+, K^+, Mg^{2+}, Ca^{2+}

Sampling:

- Inlet, Cyclone

Sample collection:

- Wet rotating denuder (gases)
- Steam-Jet-Aerosol-Collector

Analysis:

- Online ion chromatography

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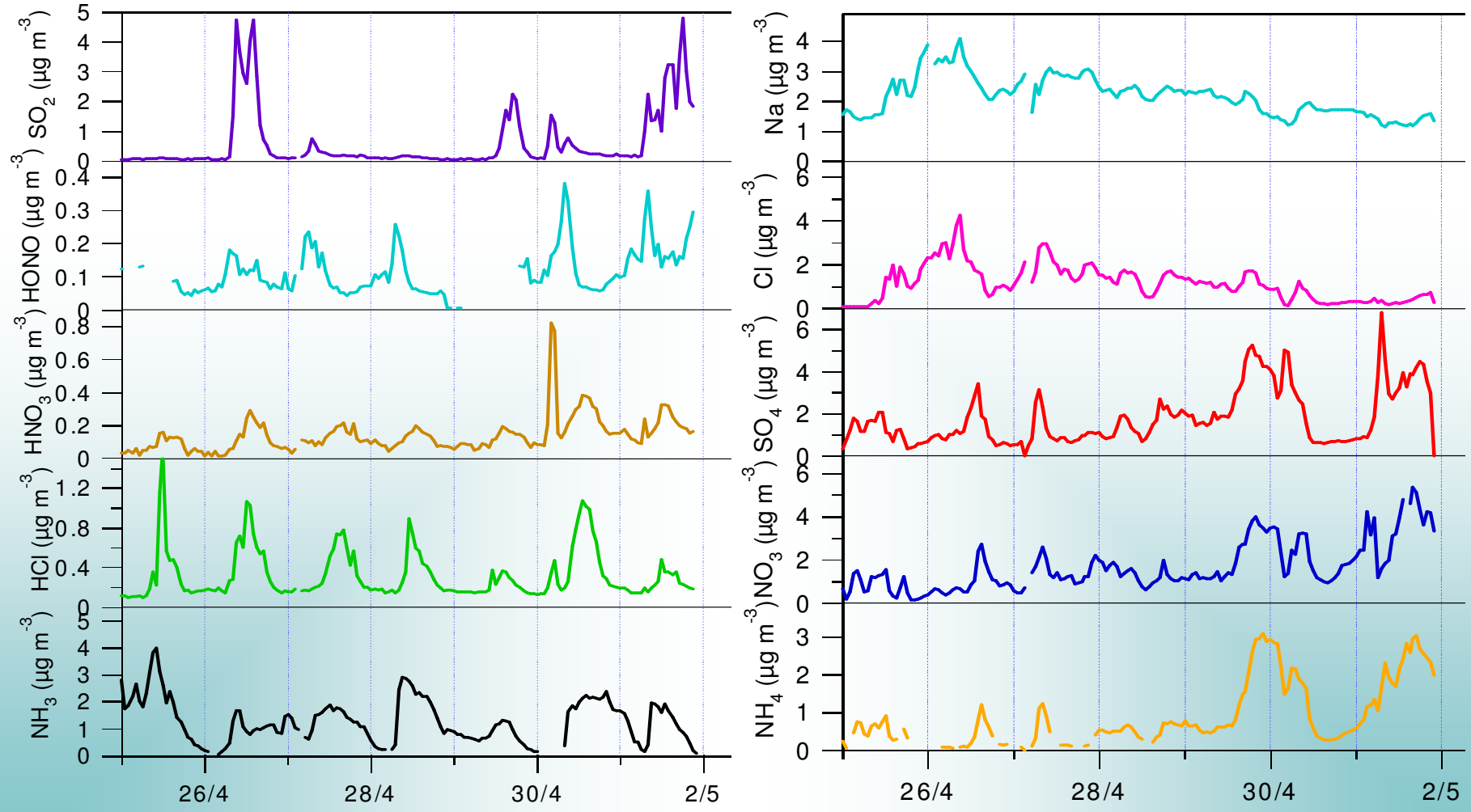
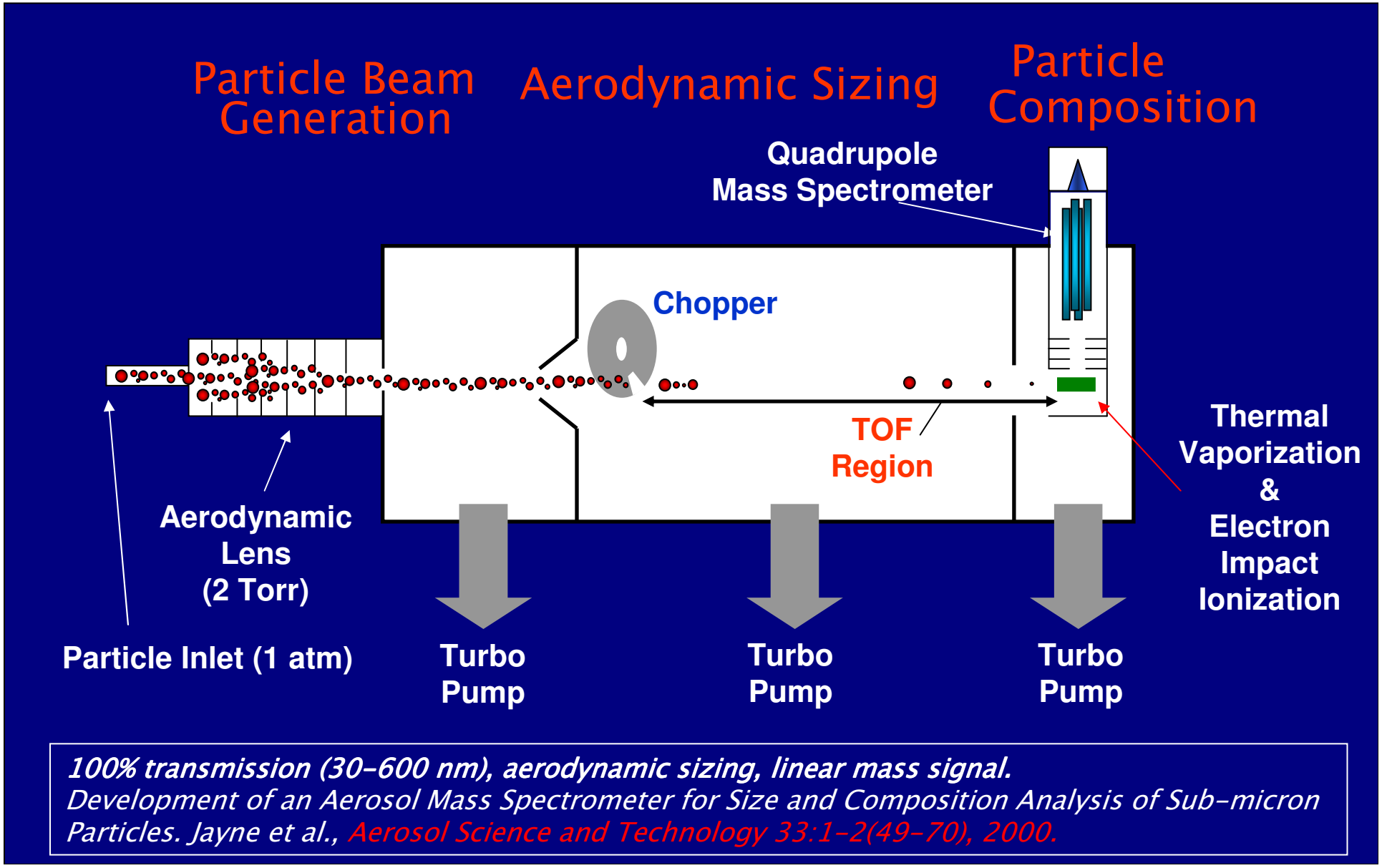
Example Time Series Auchencorth



Aerodyne Aerosol Mass Spectrometer (AMS)



100% transmission (30-600 nm), aerodynamic sizing, linear mass signal.
Development of an Aerosol Mass Spectrometer for Size and Composition Analysis of Sub-micron Particles. Jayne et al., Aerosol Science and Technology 33:1-2(49-70), 2000.

EMEP Intensive Measurement Period Jun 06 & Jan 07: Gas / aerosol partitioning of inorganic aerosol

(contributions: ECN, NL; Univ. Kuopio, FI; PSI, CH)

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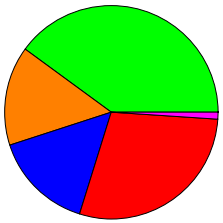
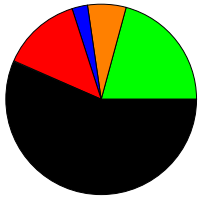
Auchencorth, Scotland

Total PM_1 = 8.6 µg m^{-3}



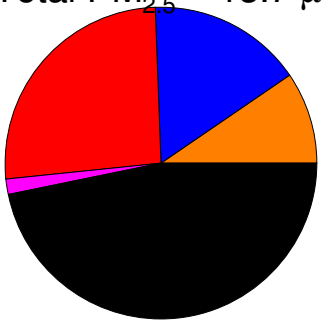
Mace Head, Ireland

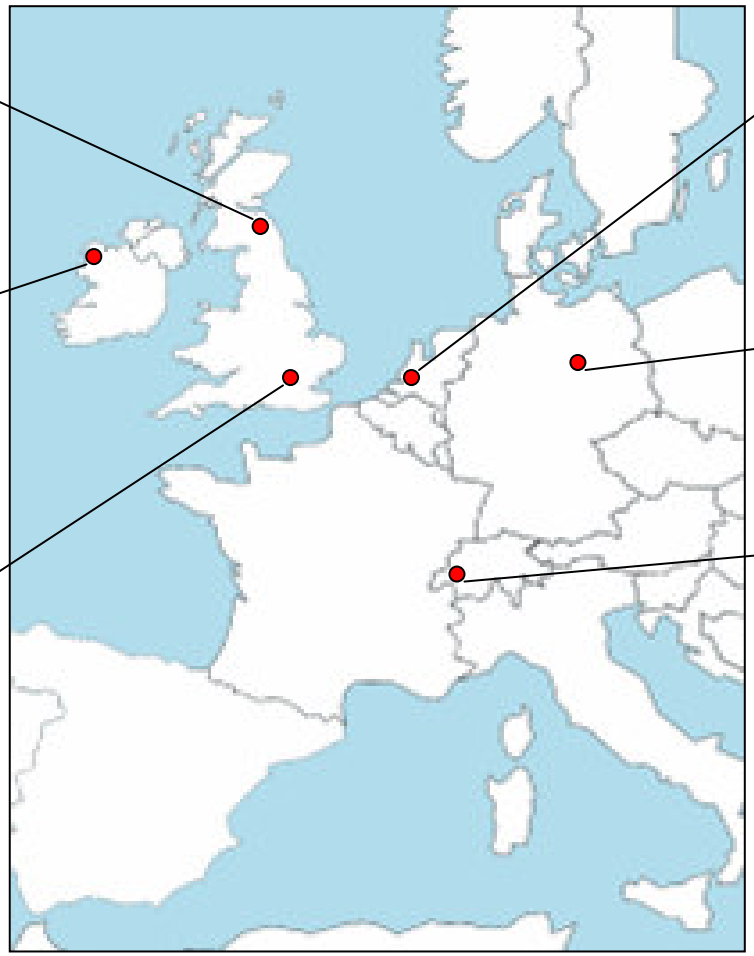
Total PM_1 = 6.0 µg m^{-3}



Harwell, England

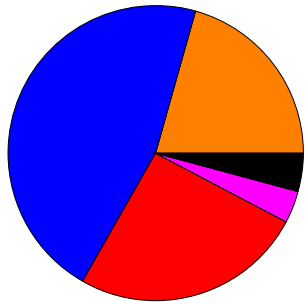
Total $PM_{2.5}$ = 13.7 µg m^{-3}





Cabauw, The Netherlands

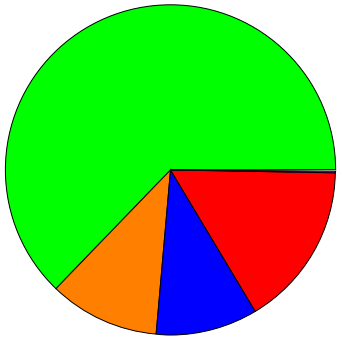
Total $PM_{2.5}$ = > 12.4 µg m^{-3}



Melpitz, Germany

Payerne, Switzerland

Total PM_1 = 19.4 µg m^{-3}



Organics
Ammonium
Nitrate
Sulphate
Chloride
Not resolved

Total Measured Concentrations June 2006

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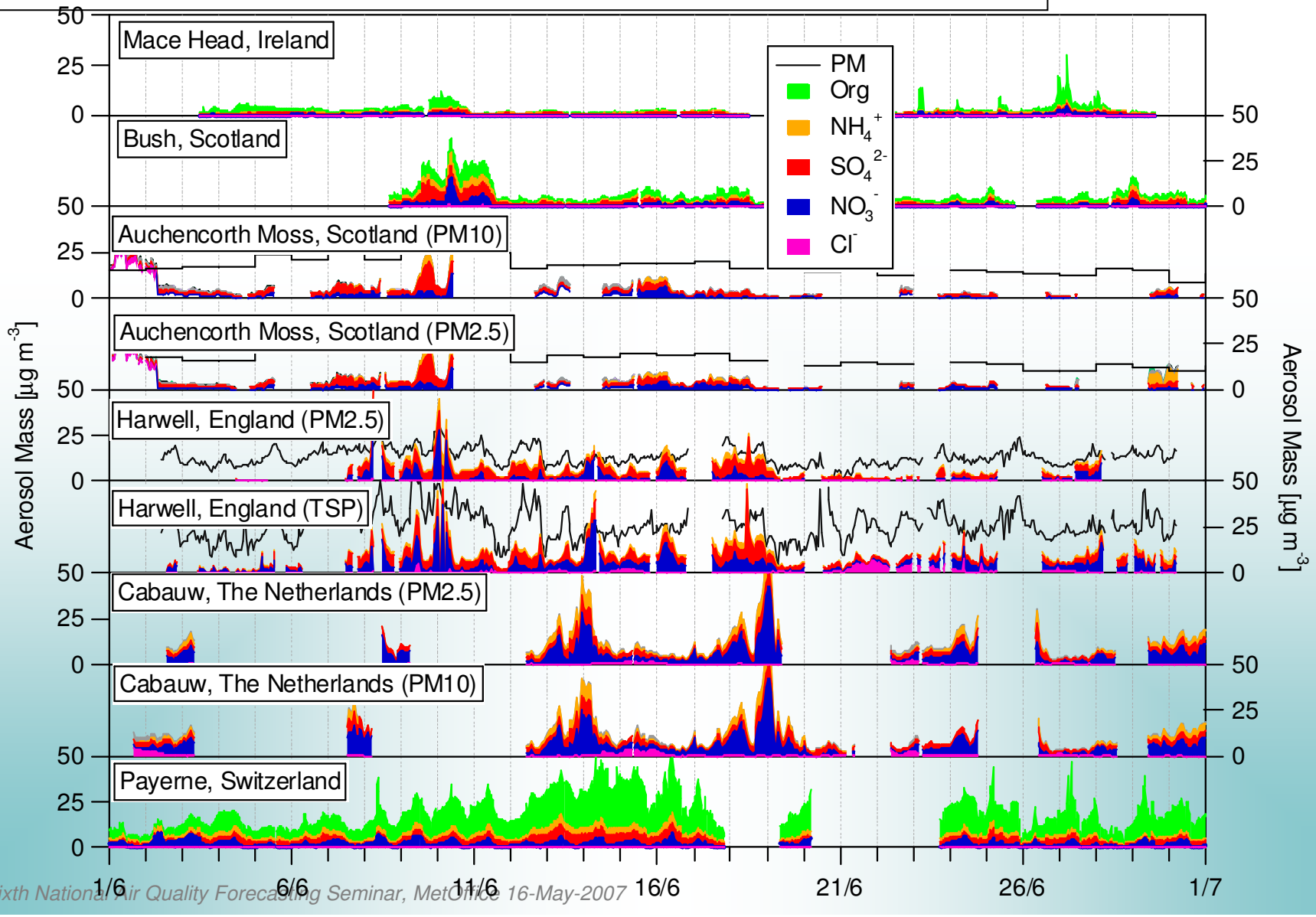
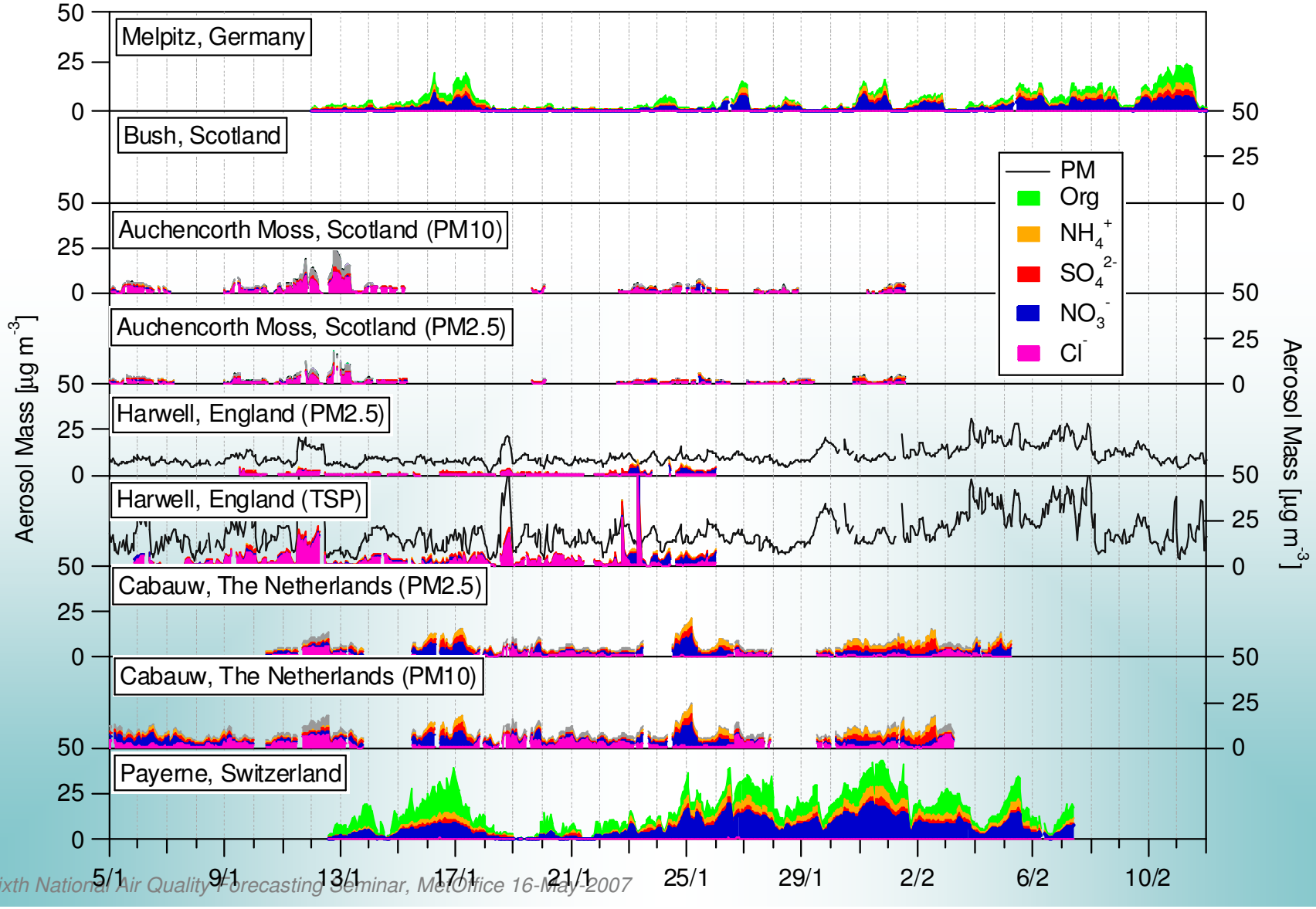
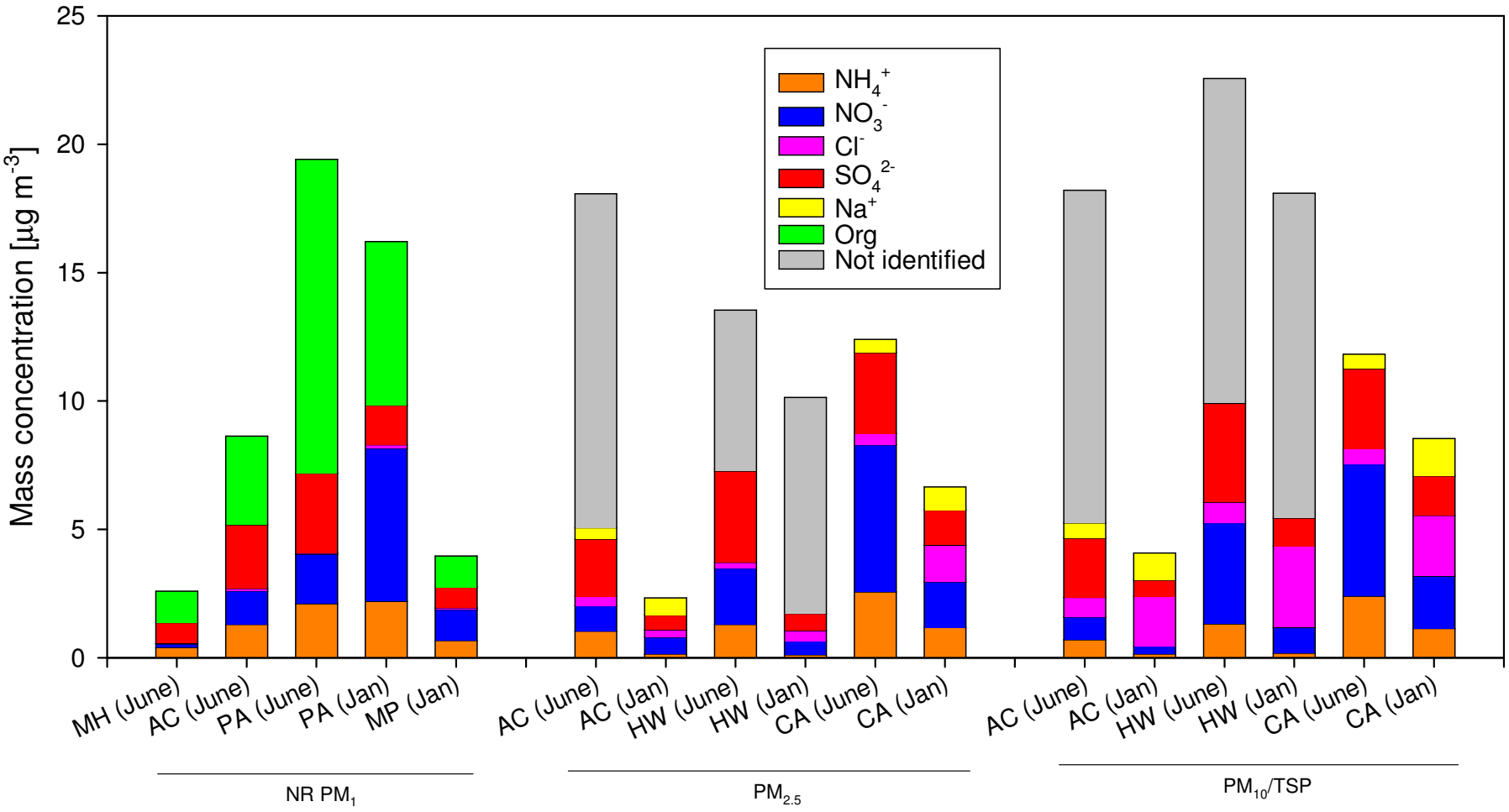


Total Measured Concentrations Jan 2007

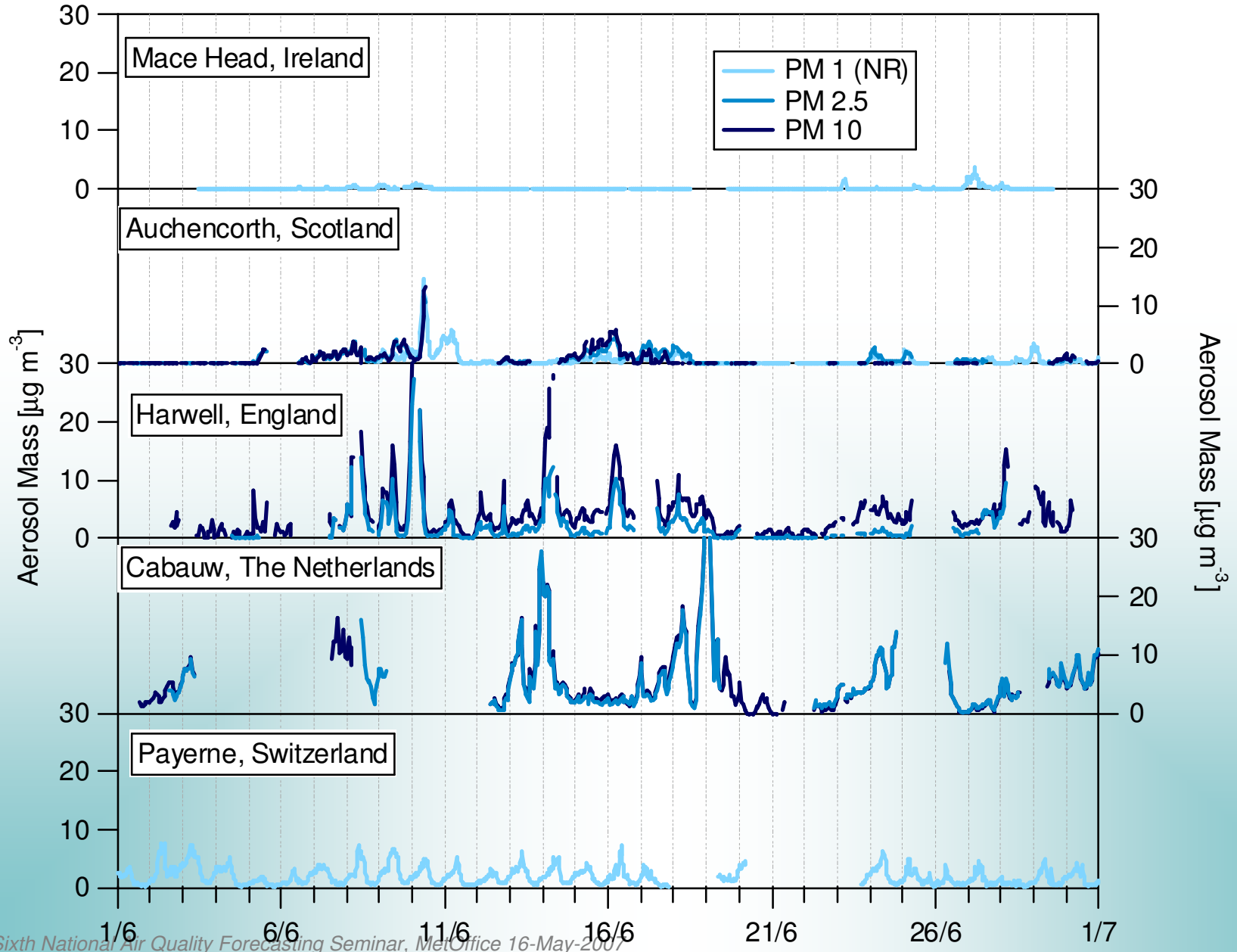
Centre for Ecology & Hydrology



Comparison of the two Intensive Periods



June 2006: Nitrate



Averaged Diurnal Cycles

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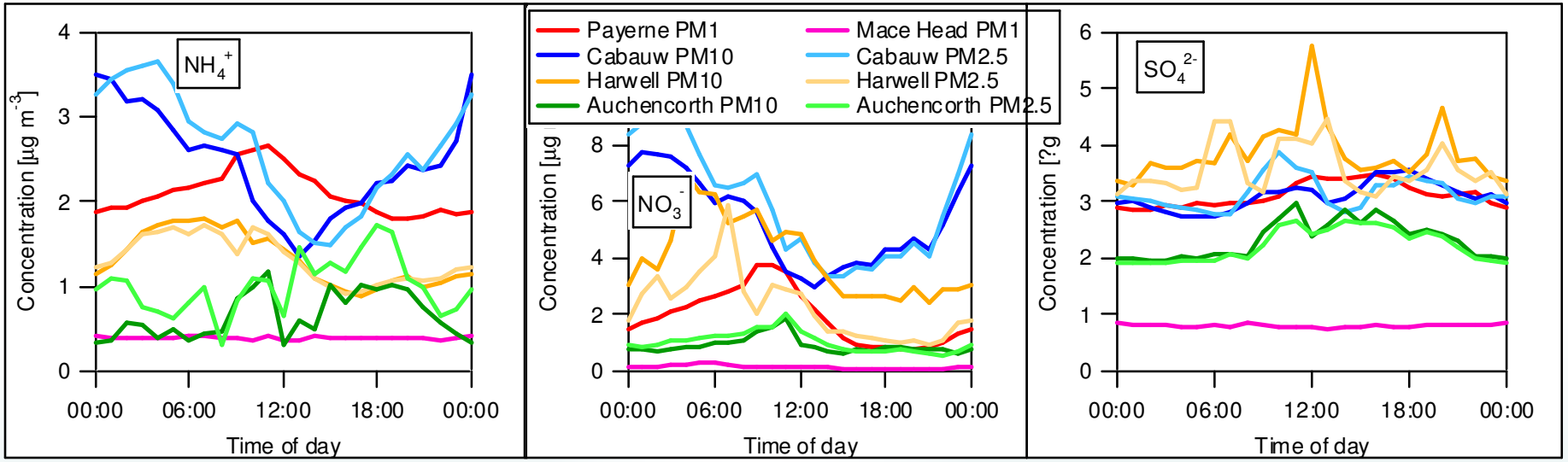
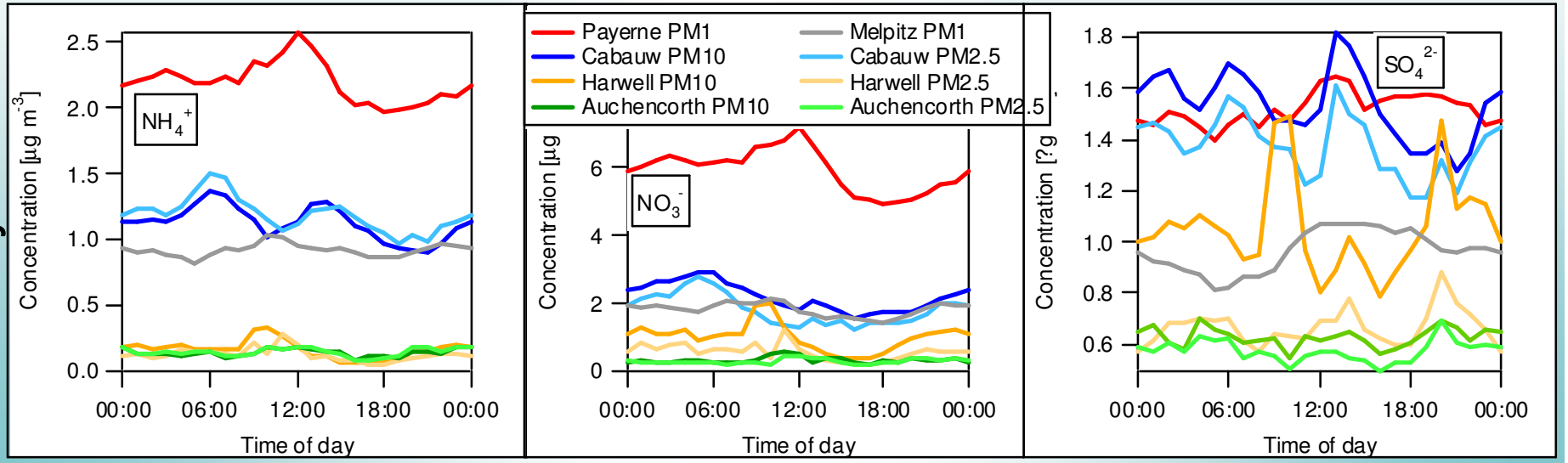
June 2006



January 2007



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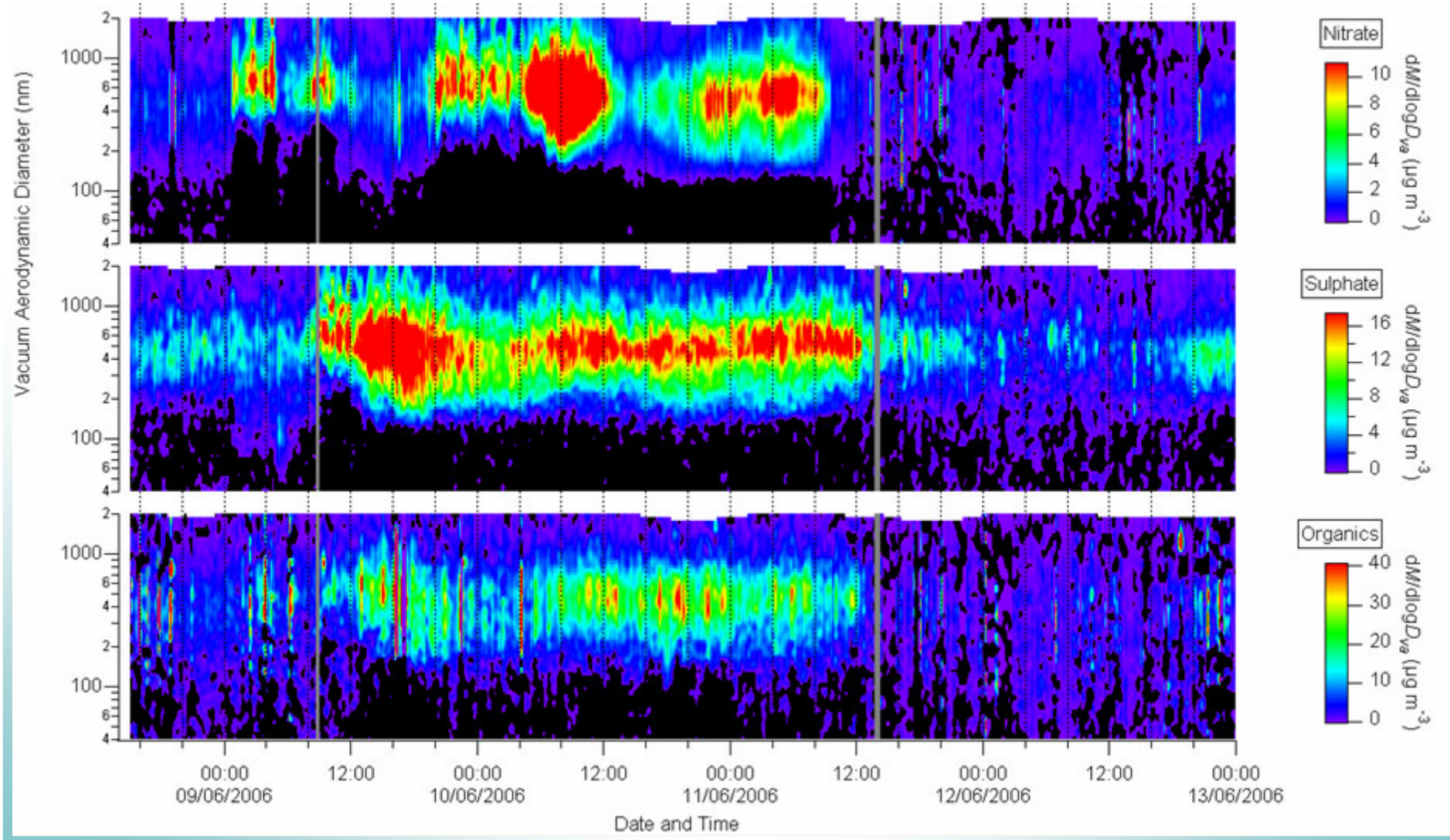
AMS Size Distribution at CEH EMEP (Bush)



Future Plans (EMEP Intensive Campaigns)

- Inter-comparison of European-scale chemical transport models using intensive campaign data (incl. EMEP Eulerian)

- Future intensive campaigns:
  - Preliminary dates: Feb & Sep. 2009
  - Include more continuous measurements (Aerosol Mass Spec., HNO_3, NH_3)
  - Measure vertical aerosol gradients (LIDAR)

NERC Project AMPEP Objectives

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Use of 'round-Britain' aircraft flights to:

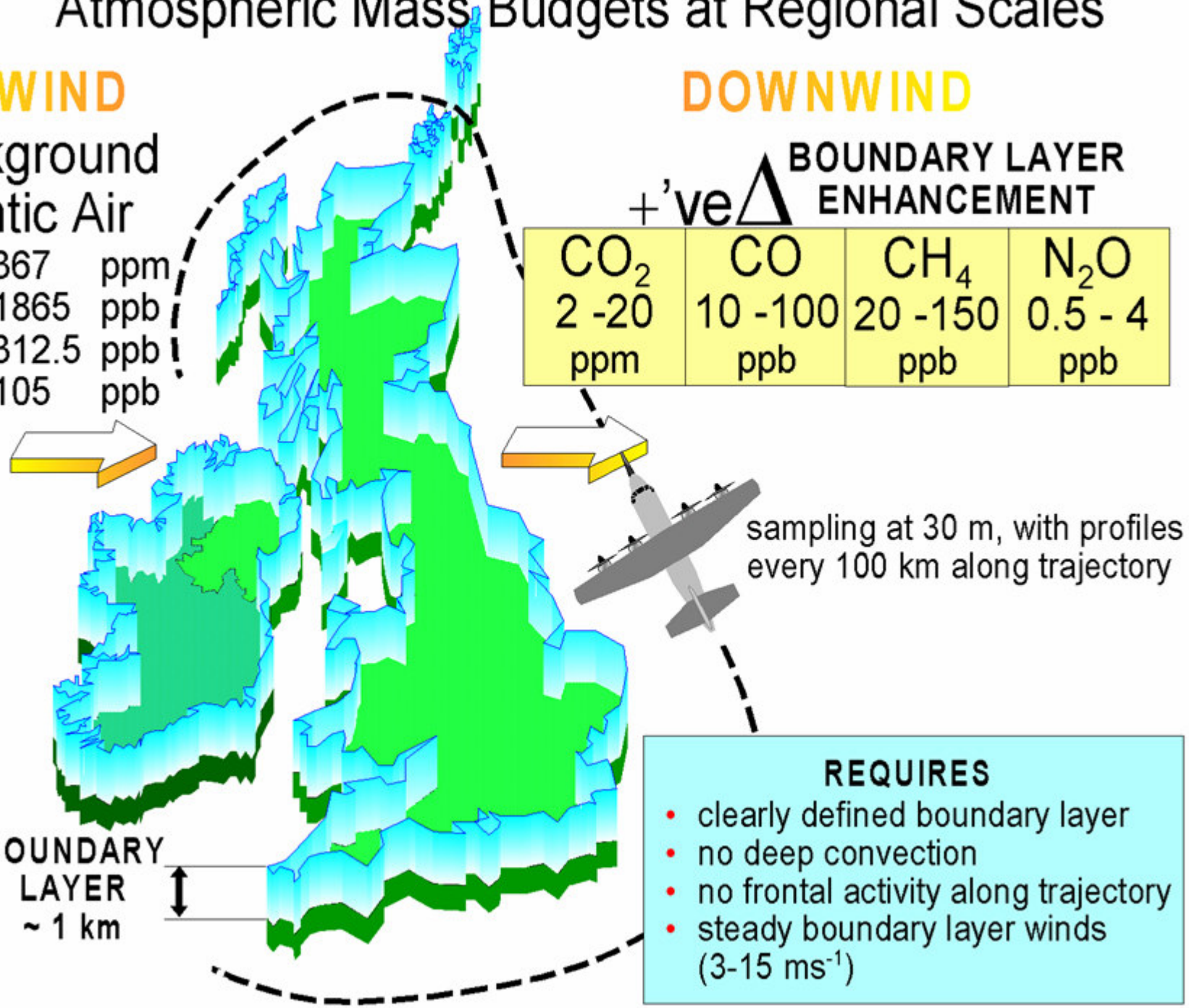
1. Deduce mass balance and the spatially disaggregated emissions of CO, CO_2, N_2O, CH_4 for the UK

2. Measure gas / aerosol partitioning of pollutants downwind of the UK, deduce chemical processing and compare with models (NO, NO_2, NO_3^-, HNO_3, SO_2, SO_4^{2-}, NH_4^+)

3. Measure the UK atmospheric mass balance UK of a range of metals (Pb, Cd, Zn, Cu, Ni, and Hg)

4. Use chemical transport models (CTMs) to interpret the data.

Atmospheric Mass Budgets at Regional Scales



Time-scales of NH_4NO_3 Formation

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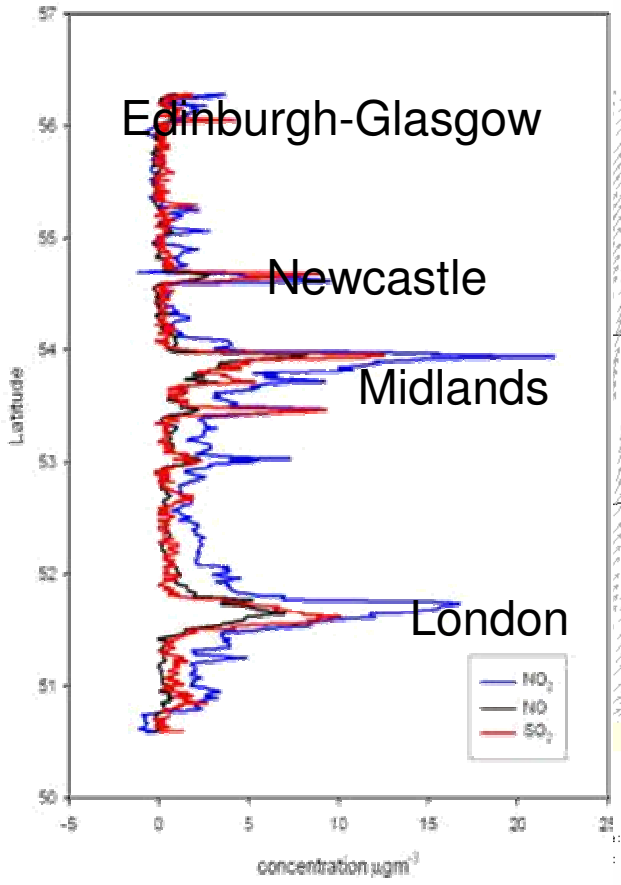
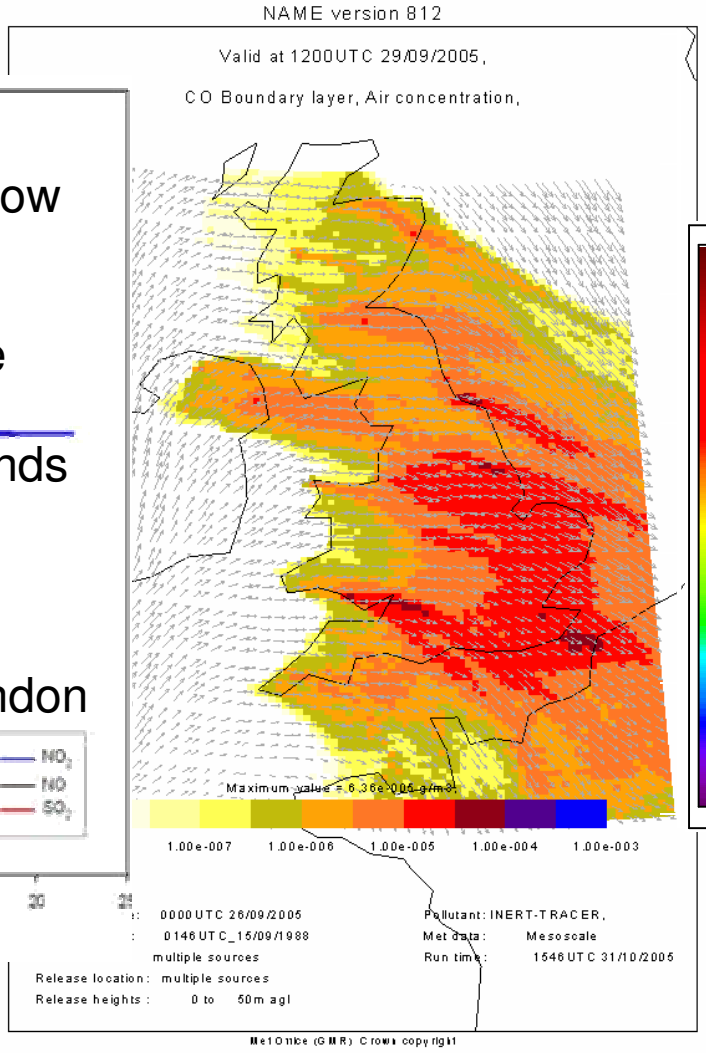
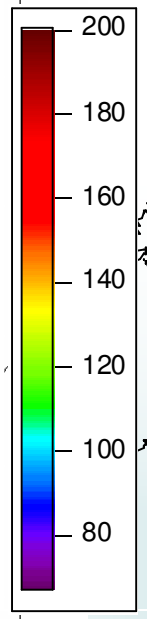
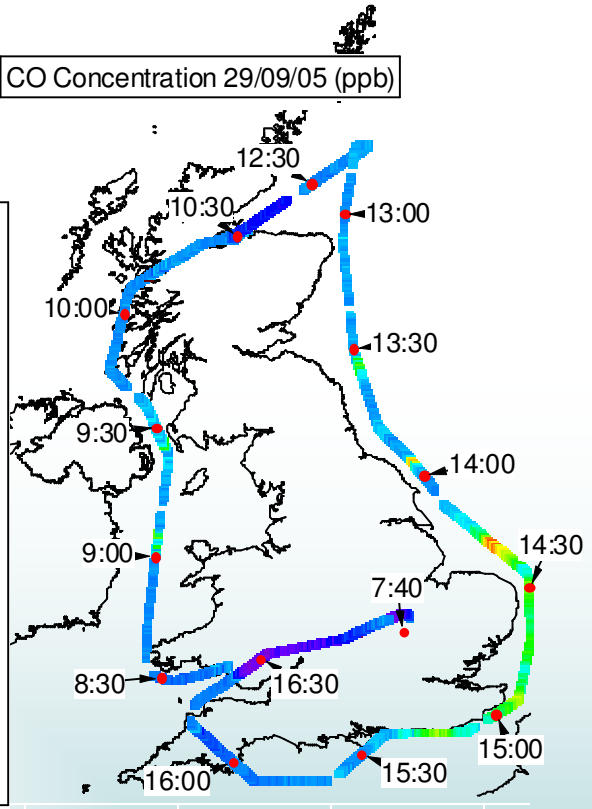


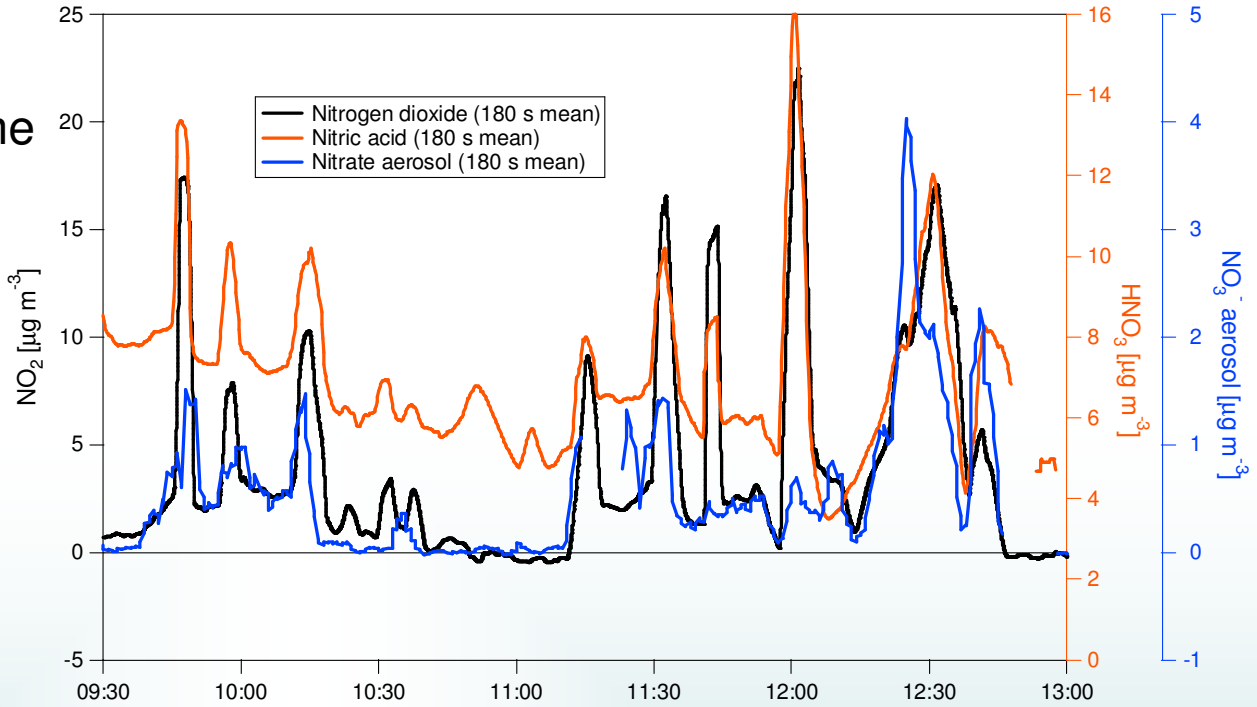






NO_2-NO_3^- oxidation rate (B111-14/07/05)

10:15 – Central Scotland plume
11:35 – Newcastle plume
12:00 – Midlands plume
(power stations)
12:30 – London plume



Average wind speed = 4.48m/s
Average wind direction = 208deg

	10:15	**11:35**	**12:00**	**12:30**
Distance aircraft from source (km)	90	50	70	100
Air parcel travel time (hours)	5.6	3.1	4.3	6.2
% N oxidised to HNO_3 (%)	22.8	20.3	32.5	35.5
% N oxidised to NO_3^- (%)	9.4	7.5	2.1	16.6
Oxidation rate to HNO_3 (%N hr^{-1})	4.1	6.6	7.5	5.7
Oxidation rate to NO_3^- (%N hr^{-1})	1.7	2.4	(0.48)	2.7
Total oxidation rate (%N hr^{-1})	5.8	9.0	8.0	8.4

Nitrogen Oxidation Rates (%N hour^{-1})

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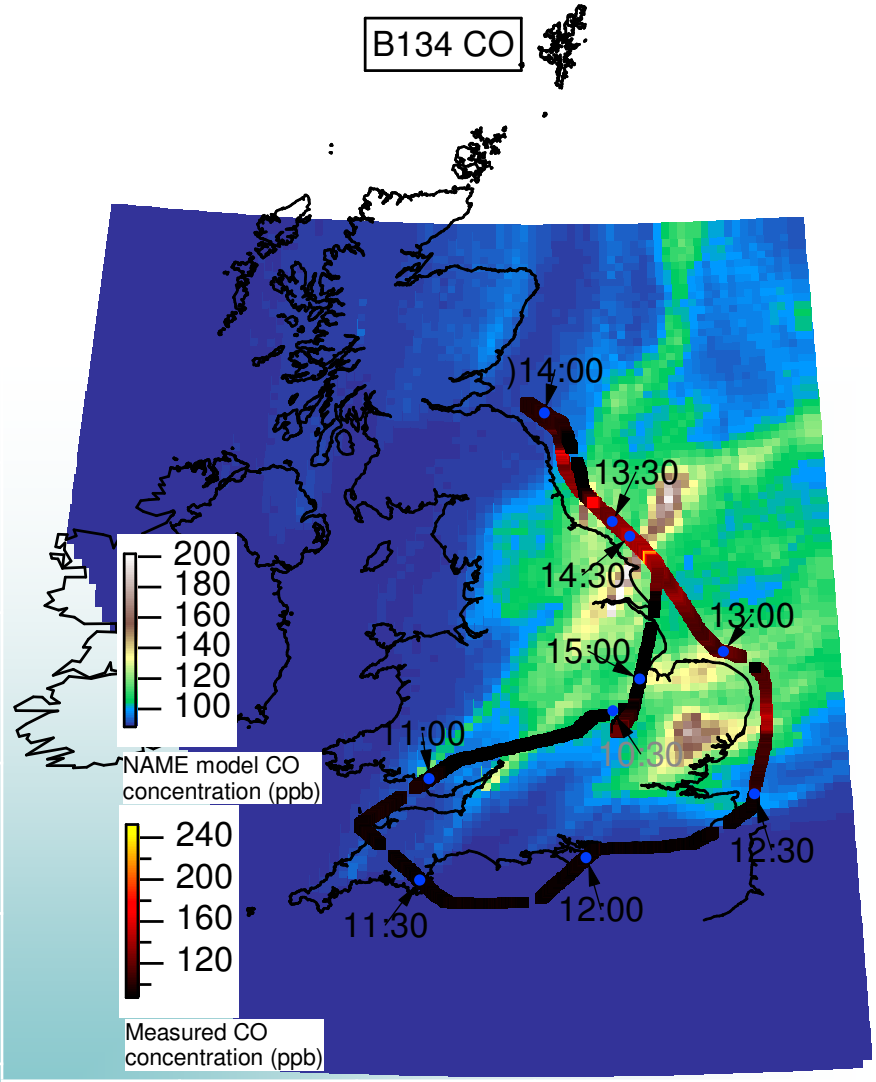
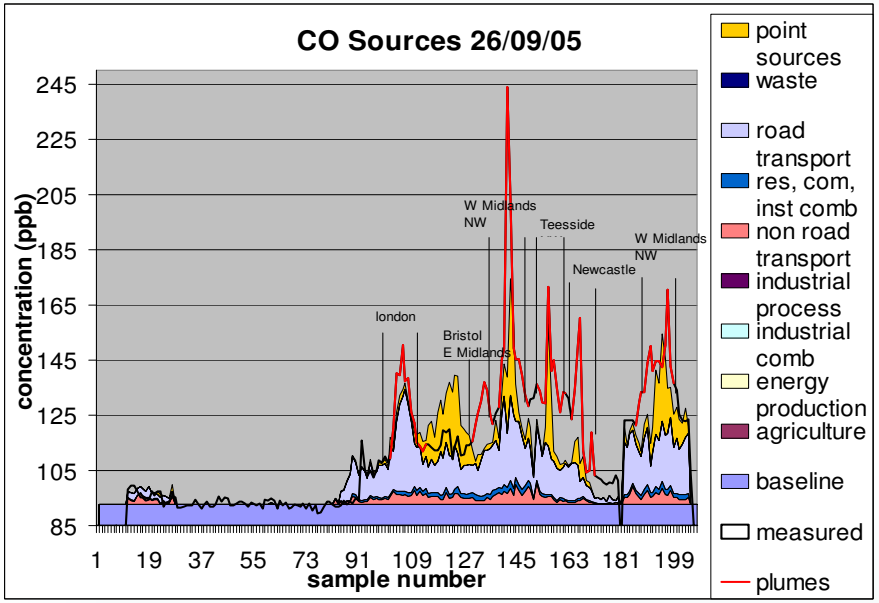
flight	CScotland	Newcastle/ Teeside	Yorkshire/ Midlands	London	Bristol/Channel
B102		1.06	0.53	1.06	
			0.66		
B111	1.68	2.43	(0.48)	2.67	
B112		2.00	4.37	0.94	
			(0.04)		
B118		0.98	0.35	0.68	0.40
			0.52		
			(0.16)		
			1.39		
			1.42		
B119			1.12	0.76	
			1.17		
B136		0.53	0.27	6.79	
			(0.11)		
B102	1.00	0.53	0.66	1.06	
B111	1.68	2.43	(0.48)	2.67	
B112	0.94	4.37	(0.04)	2.00	
B118			0.40	0.68	
	1.39	1.42	0.35	0.52 (0.16)	0.98
B119	1.12	1.17	0.76		
B136	0.53	0.27	(0.11)	6.79	

Average **Urban Plumes ~1.7%N h^{-1}**
Industrial/Power Stations ~0.7%N h^{-1}

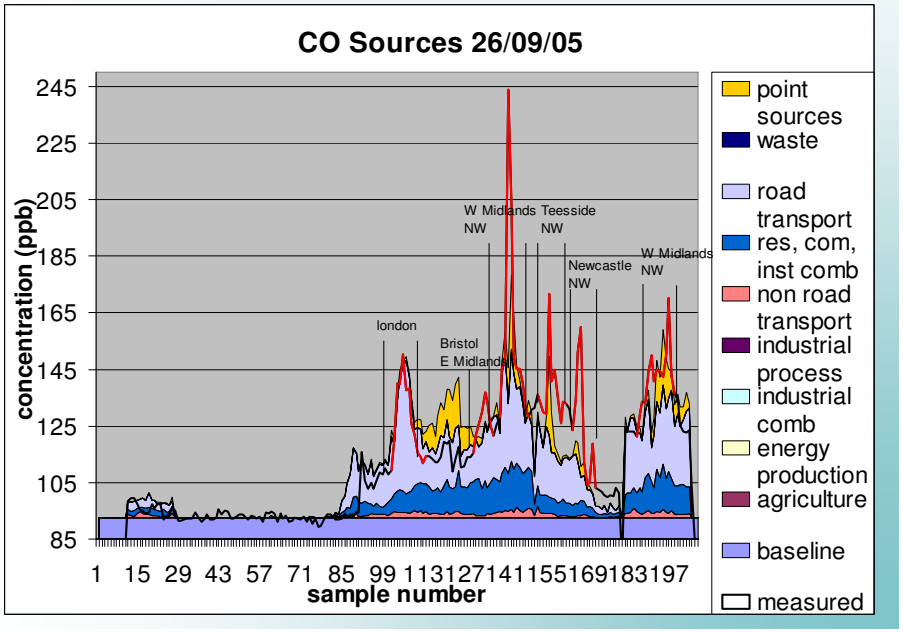
Estimating National CO Emissions using NAME



Original emissions



Adjusted emissions



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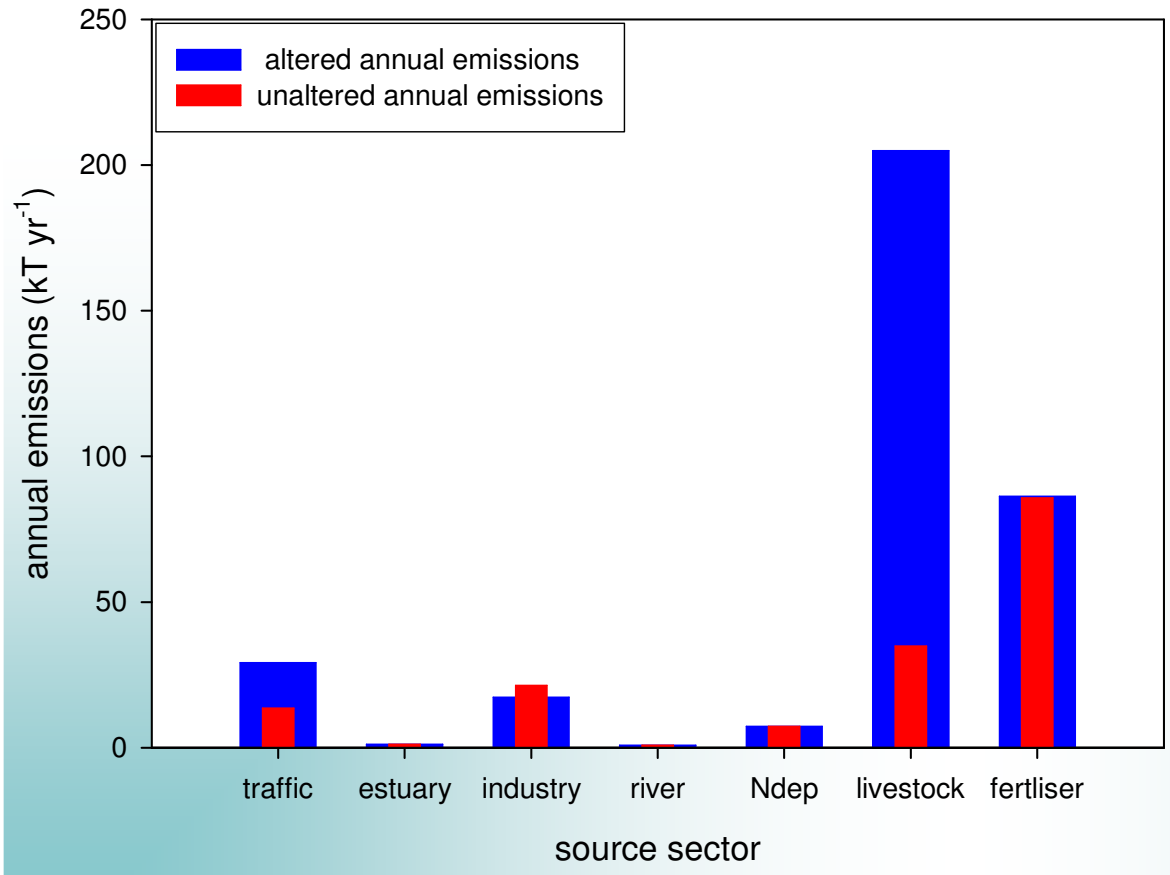
Summary of Annual Emissions Estimates (kt yr^{-1})

	CO	CO_2	N_2O	CH_4
UK bottom-up	2,757	572,196	130	1,933
Ireland bottom-up	239	43,469	31	607
NAME	2,400 ± 226	560,000 $^{139,000}_{133,000}$	350 ± 208	4,000 $^{1400}_{1290}$
Box Approach	2,700 ± 898	670,000 ± 94,000	310 ± 217	4,200 ± 2,130

Average Adjustment of N_2O Emissions Sectors



Source Sector	Relative Adjustment in Annual Emission
Traffic	x 2.2
Estuaries	----
Industry	x 0.8
River	----
N dep	----
Agriculture	x 5.9
Fertiliser	x 1.0

Inverse Modelling Using NAME

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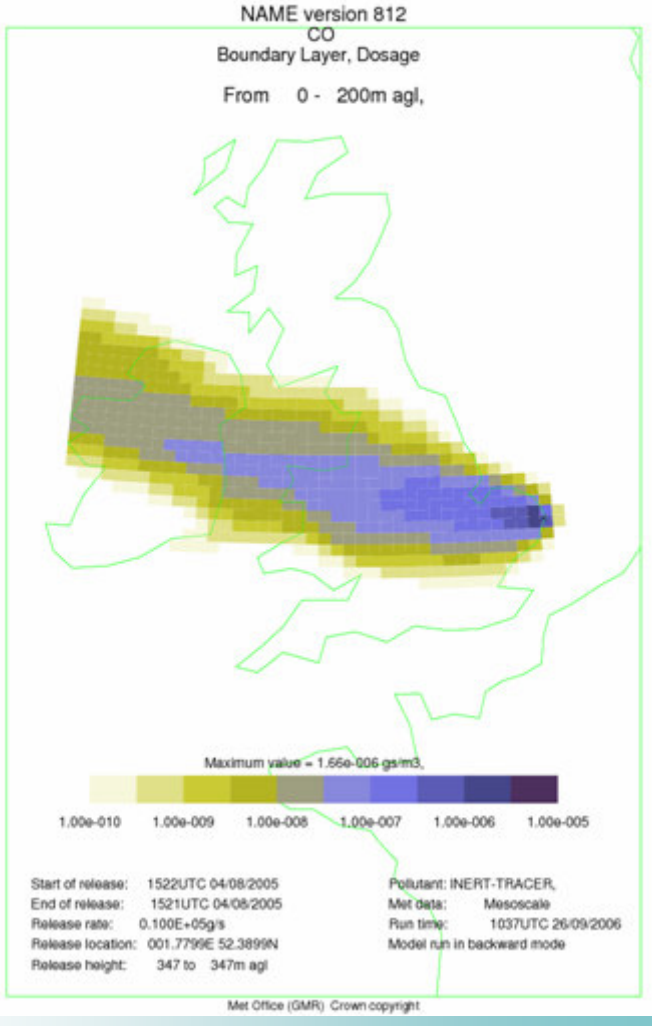
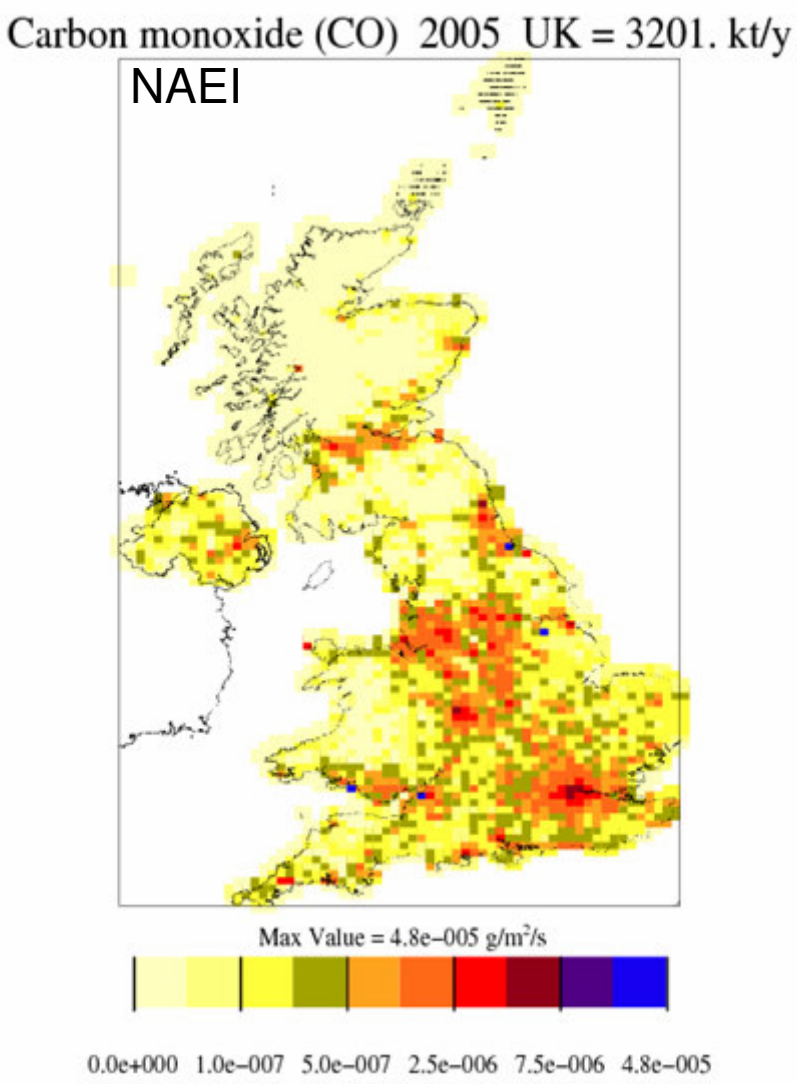
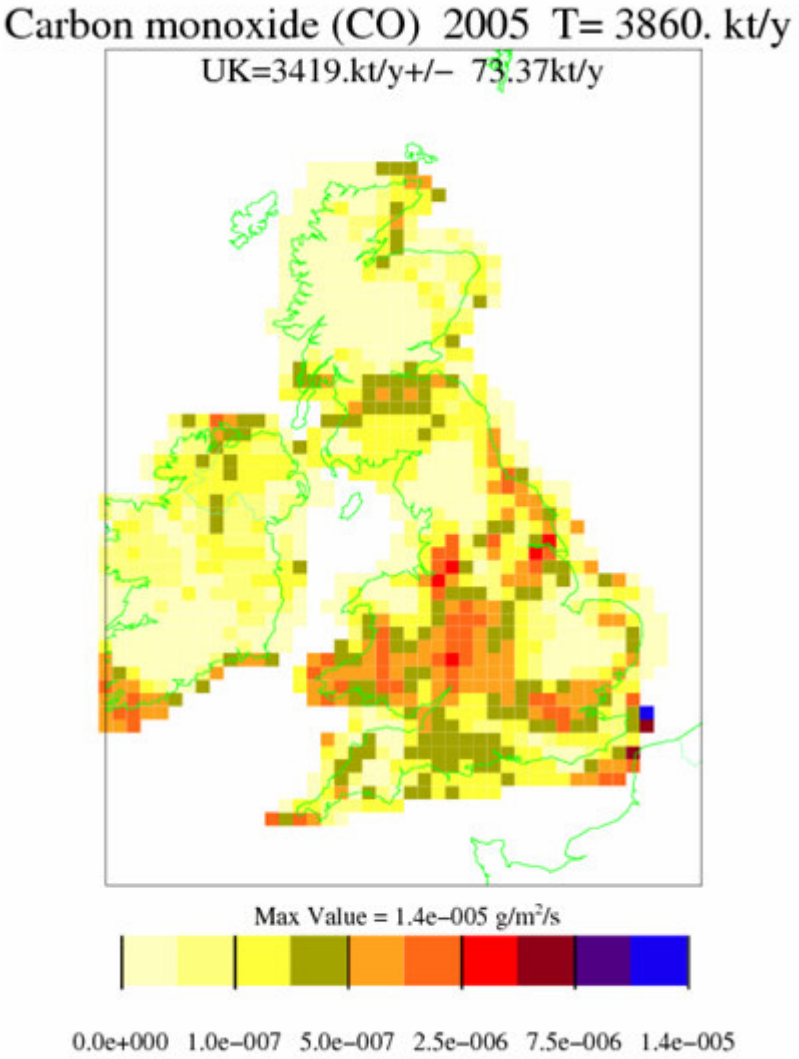
- Generate dosage maps of the origin of the air samples measured on the flights.
- Combine observed concentration enhancement with these dosage maps used to derive UK emissions.
- UK emissions derived using an iterative best-fit technique (simulated annealing), correlation coefficient as statistical score.
- Optional constraint: all emissions have to come from UK and Ireland.

Example 'Dosage Map'
(Single 1-Minute Measurement)

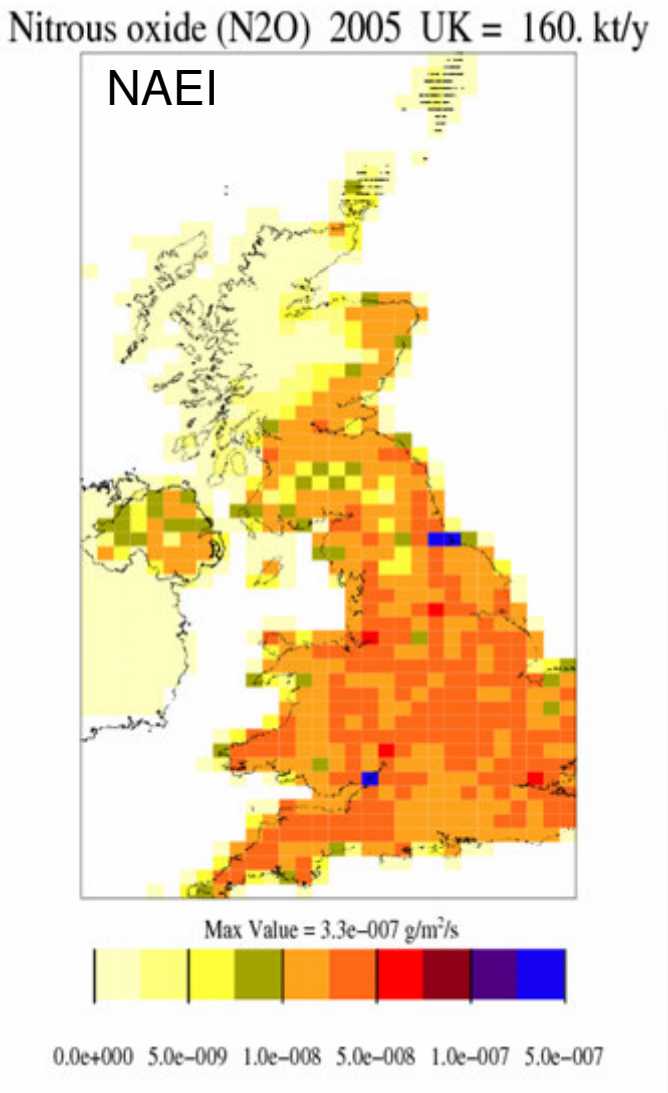


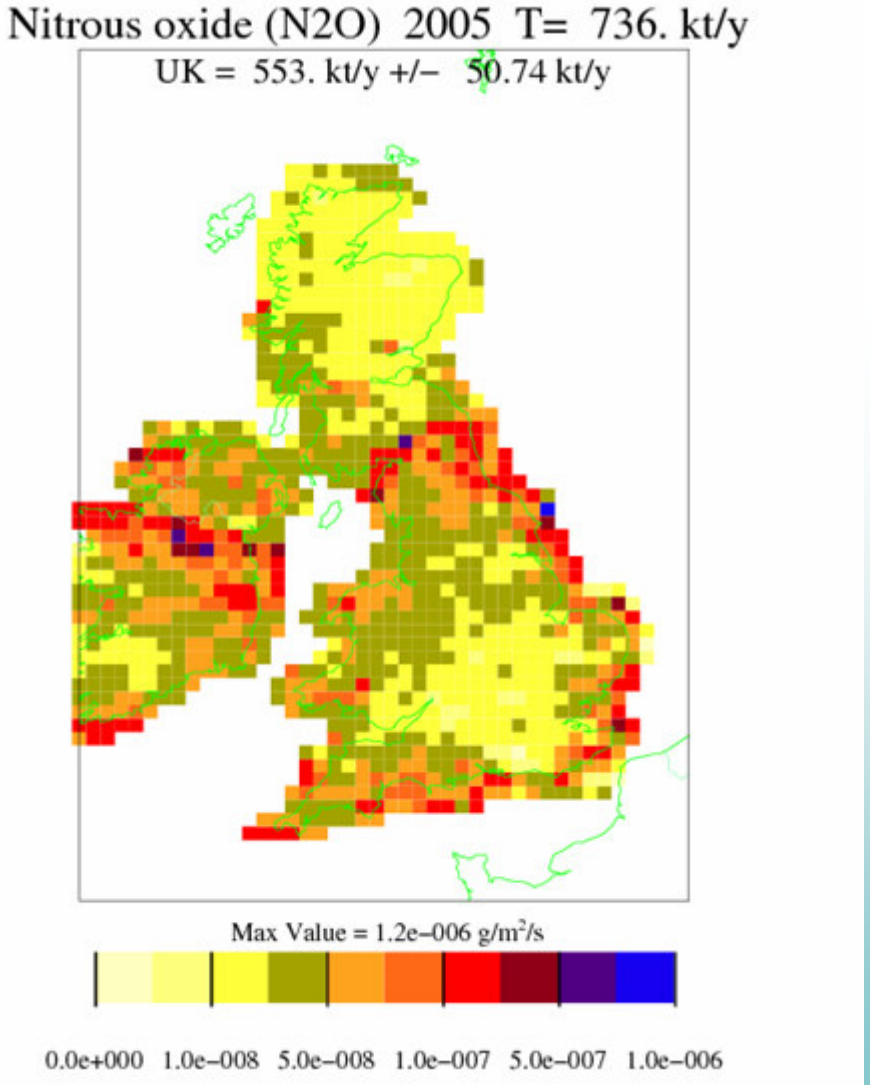
Inverse Calculation of UK CO Emission Field





Inverse Calculation of UK N_2O Emission Field

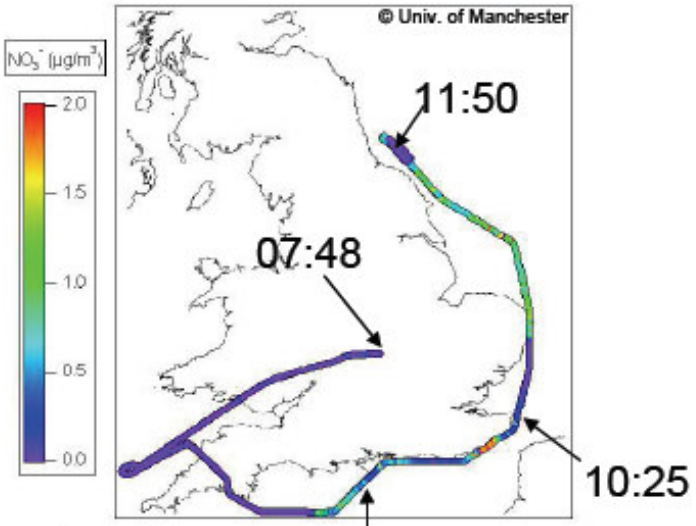




Assessment of Models-3 Using AMPEP Data

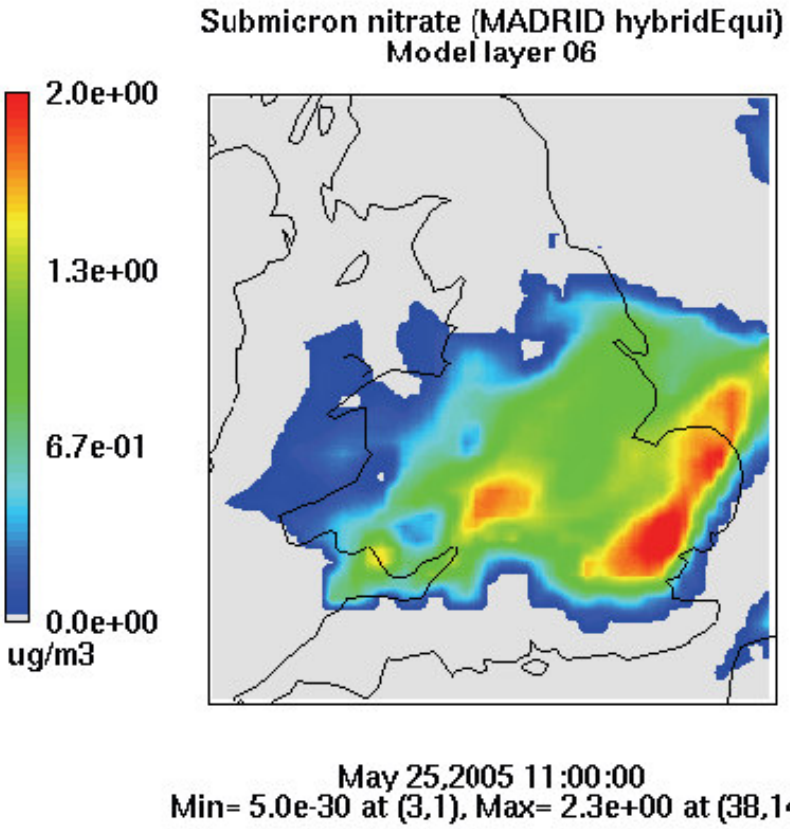
Exploring how Models-3 (standard CMAQ & MADRID) predicts measurements

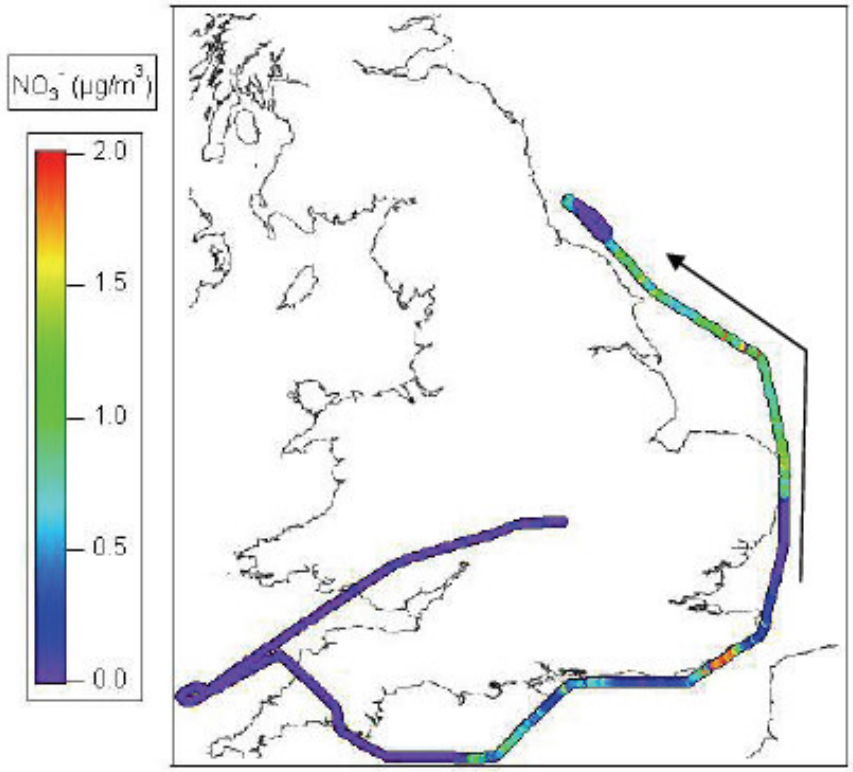
- BAe 146 Flights 2005 / 6,
- focus on B097, AMPEP
  - Anticlockwise, May 2005, (S) Westerlies
  - Aerodyne AMS



NB: altitude of flight varies greatly:

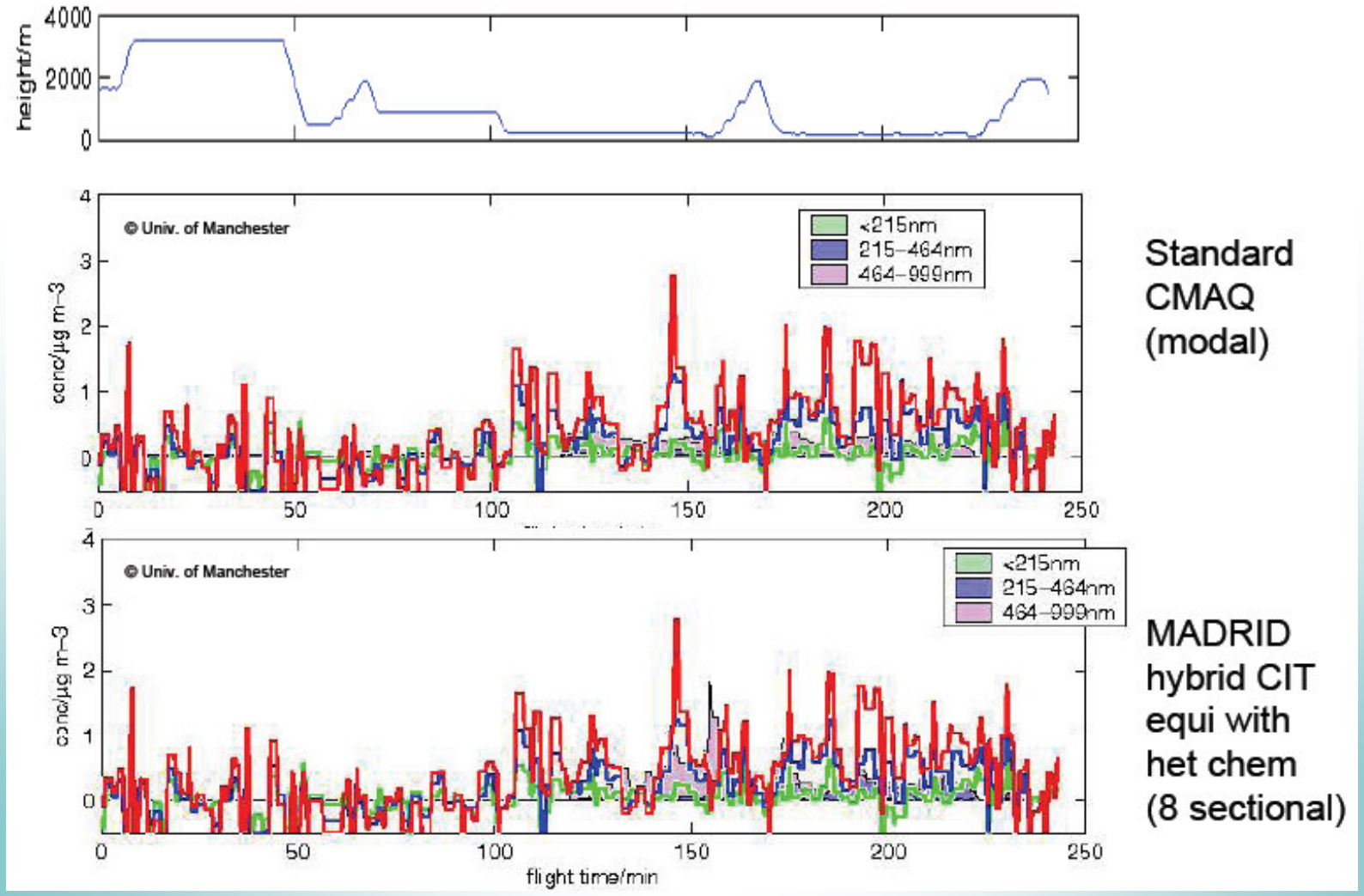
Measured and modelled NO_3^- during flight 25-May-05



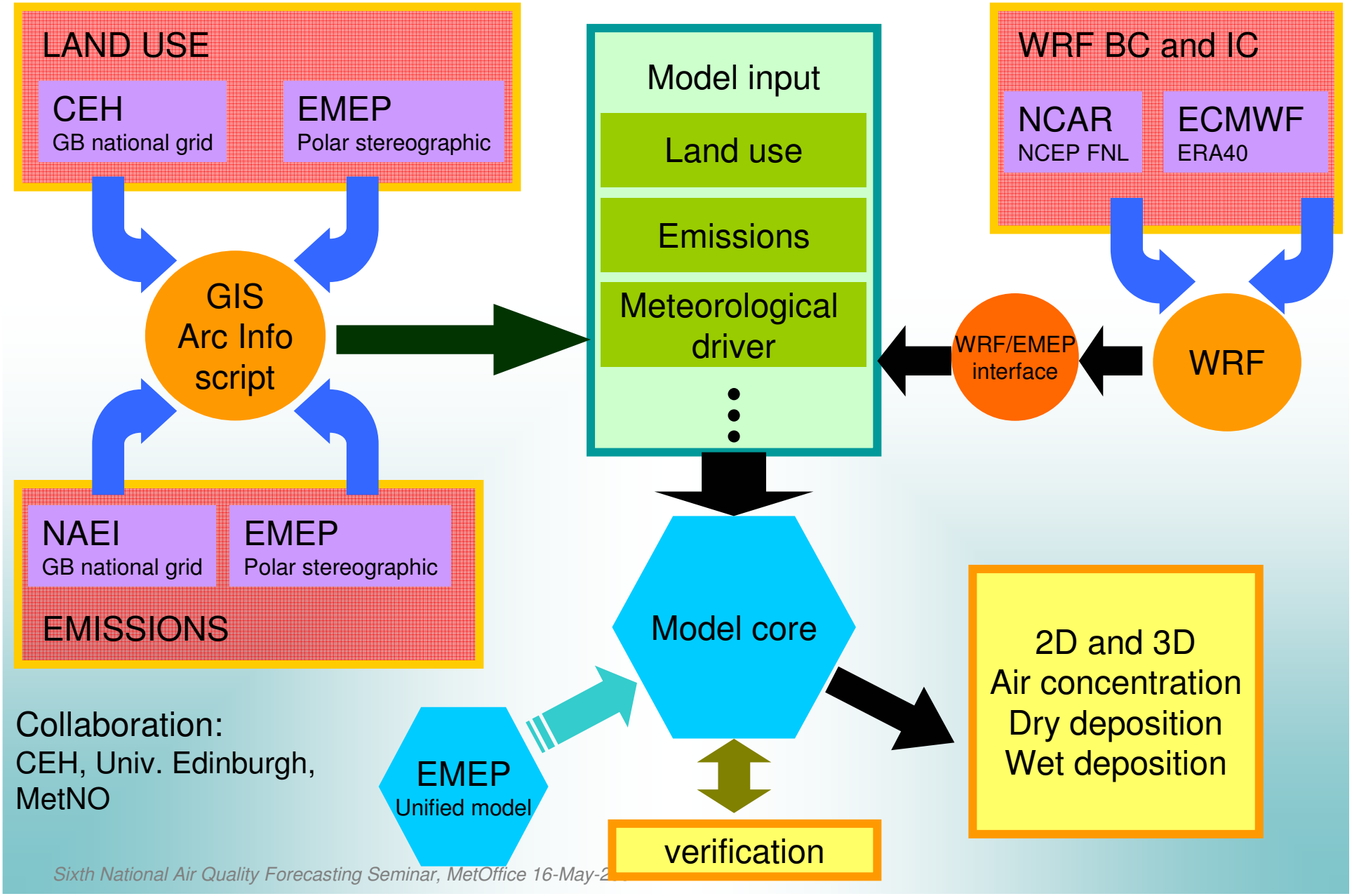


Michael Bane, Gordon McFiggans; SEAES, University of Manchester

Measured & Modelled Time-Series

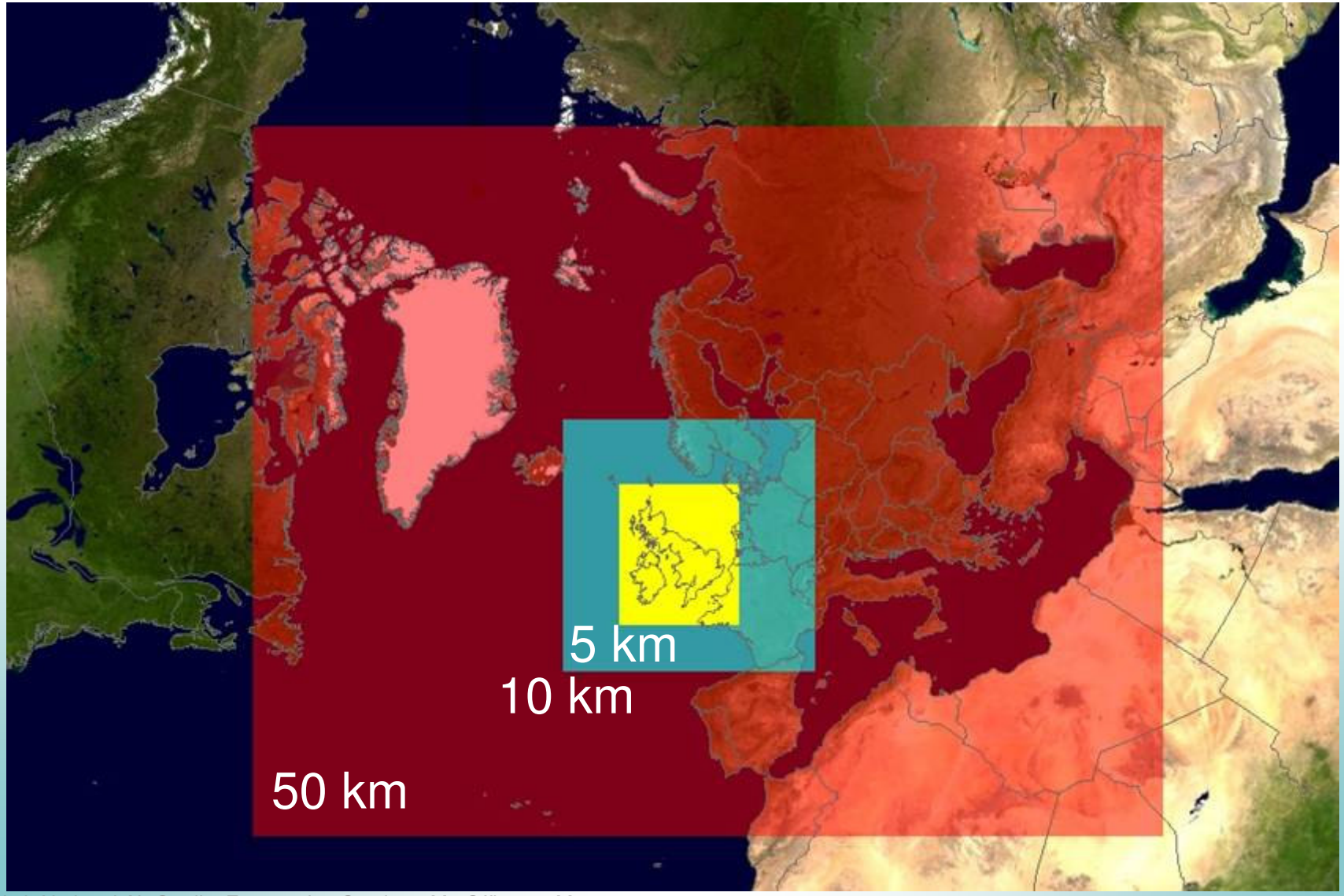


Alternative Eulerian UK modelling framework: EMEP4UK



Collaboration:
CEH, Univ. Edinburgh, MetNO

WRF domain setup



WRF model results surface rainfall

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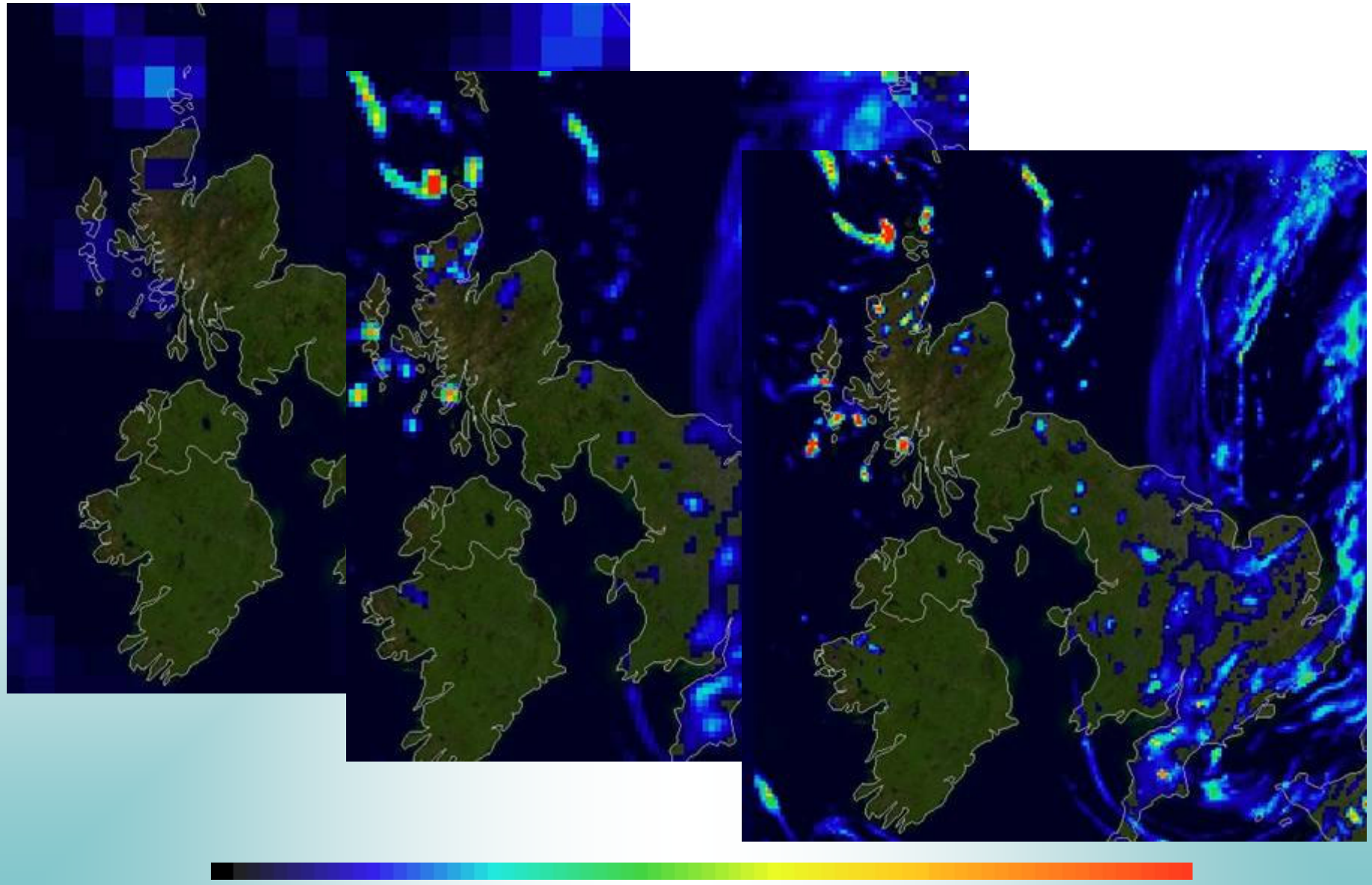
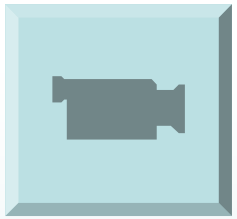


0 5 mm / hr

EMEP4UK Activities

- Operational model focussing on deposition maps
- Currently simulated months 2003
- Next steps:
  - ➢Run for full year (2003)
  - ➢Run for EMEP Intensive Campaigns and AMPEP flights
  - ➢Could easily be setup to run as air quality forecasting model



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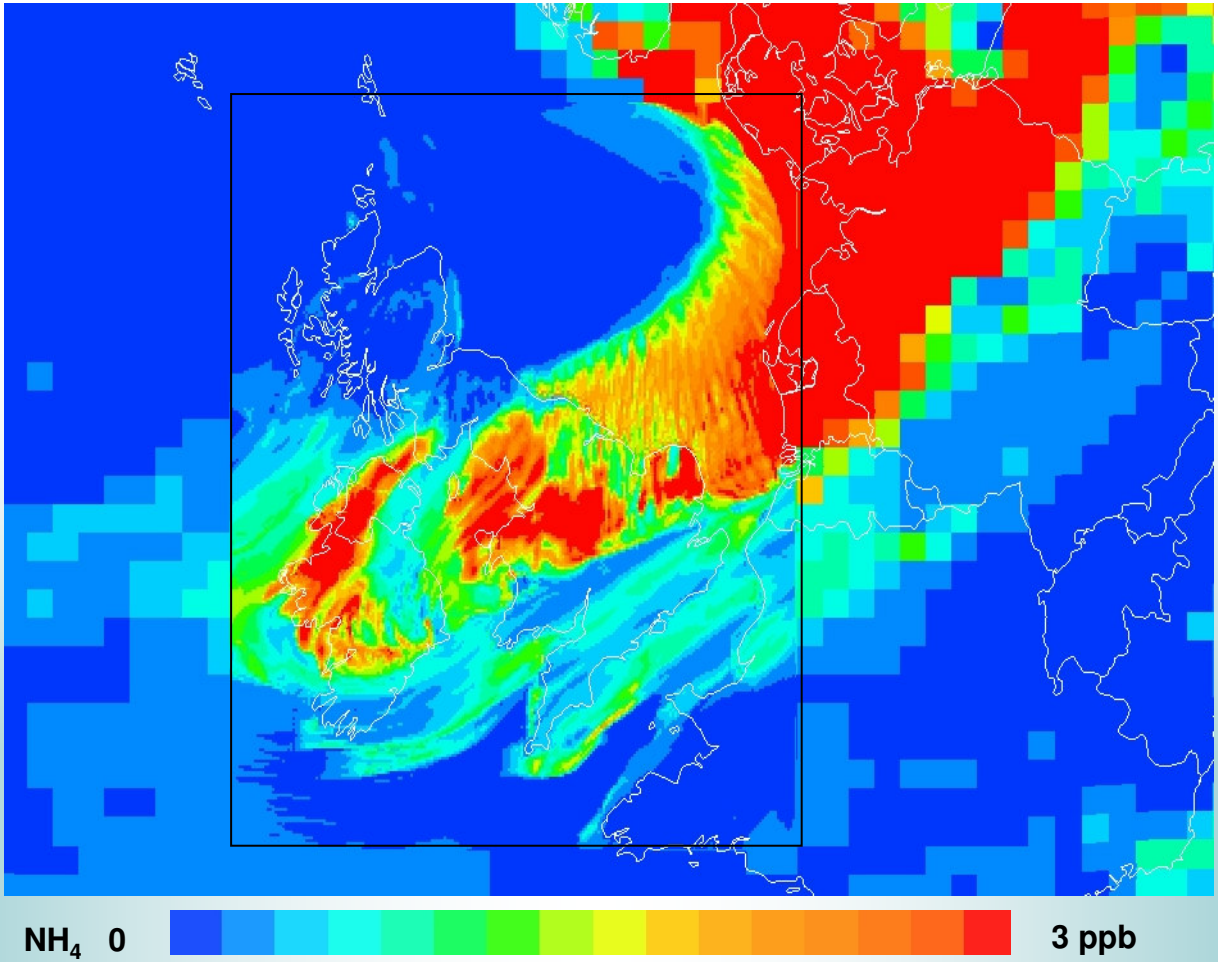
EMEP4UK: NH_4^+ on 02/01/2003 12:00 GMT



Validation of Emission through Urban Flux Measurements BT Tower London (October 2006)

Combined activity of NERC 'CityFlux' & BOC 'REPARTEE'

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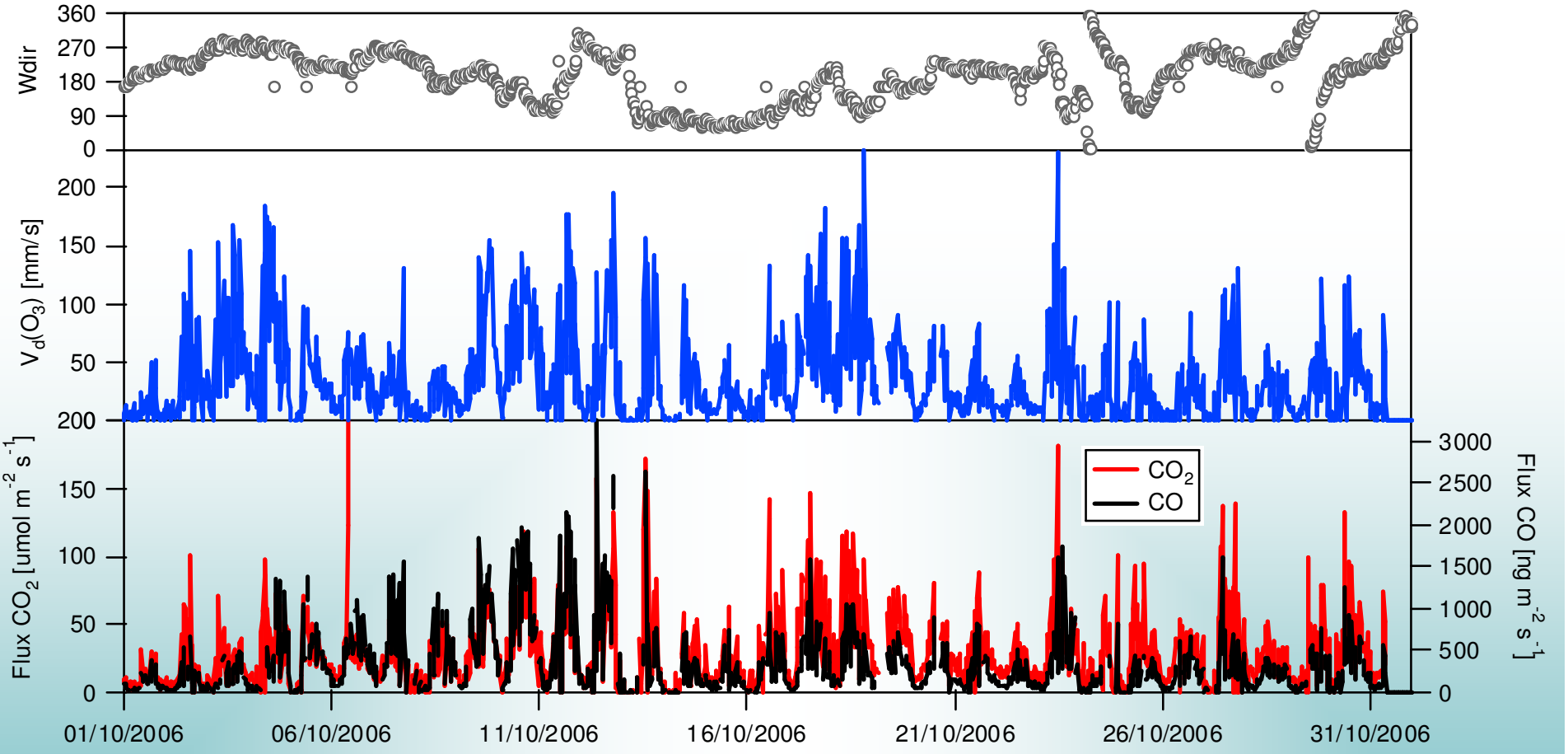
- Measurements supported by:
  - NERC CityFlux (urban fluxes above Gothenburg, Mexico City, Edinburgh, Manchester & London)
  - BOC Foundation 'REPARTEE' (contrasting aerosol at BT Tower and Regents Park)
- Flux measurements of:
  - CO
  - CO_2
  - Aerosols (number, NO_3^-, SO_4^{2-}, organics)
  - VOCs

Time series CO, CO_2, O_3



V_d = - flux/conc

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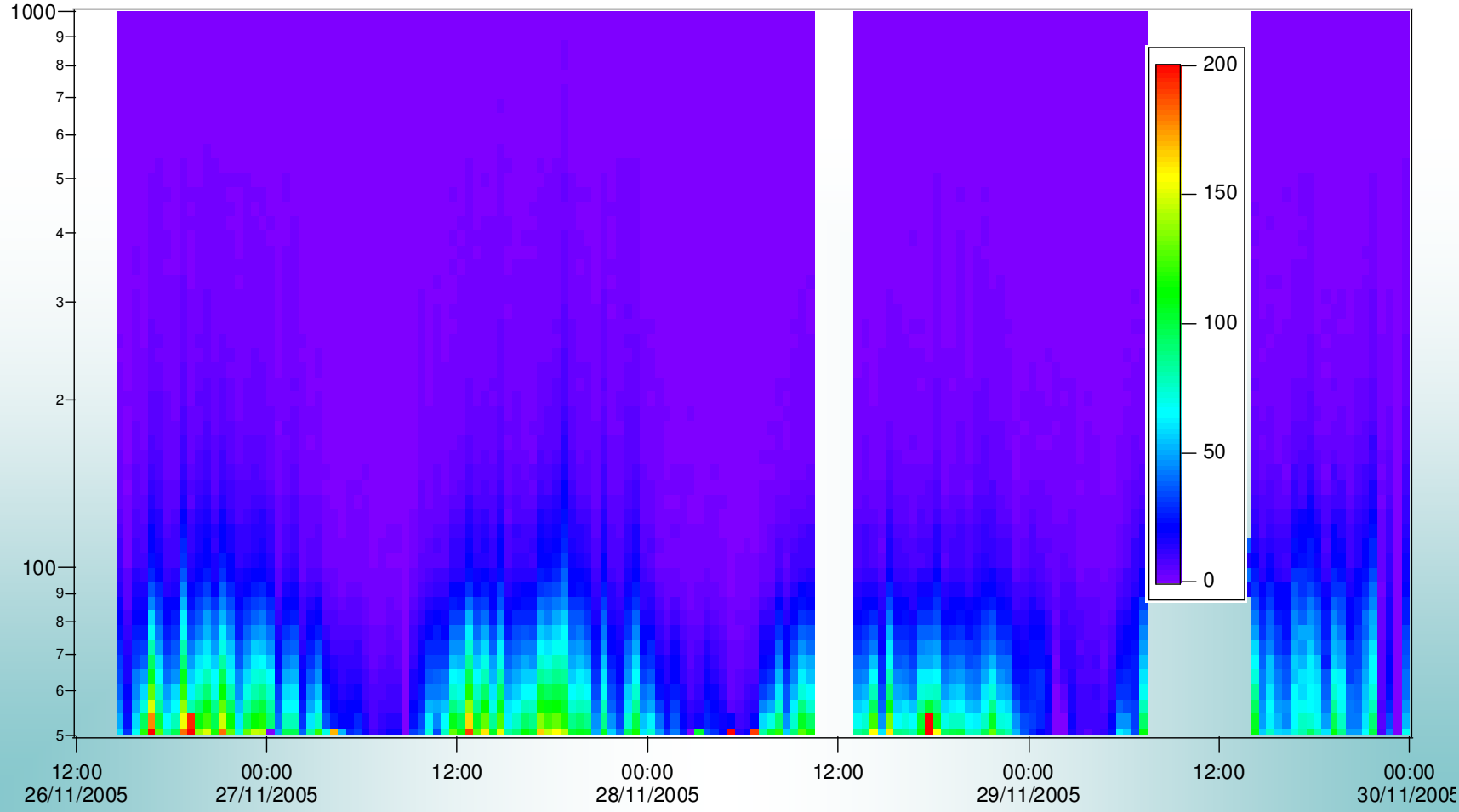
Urban Aerosol Fluxes Measured with an Ultra-High Sensitivity Aerosol Spectrometer (UHSAS) [# cm^{-2} s^{-1}]



VOC Fluxes above London by Proton Transfer Reaction Mass Spectrometer

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Sensible Heat

Methanol (M33)

Mass 41

Mass 42

Acetaldehyde (M45)

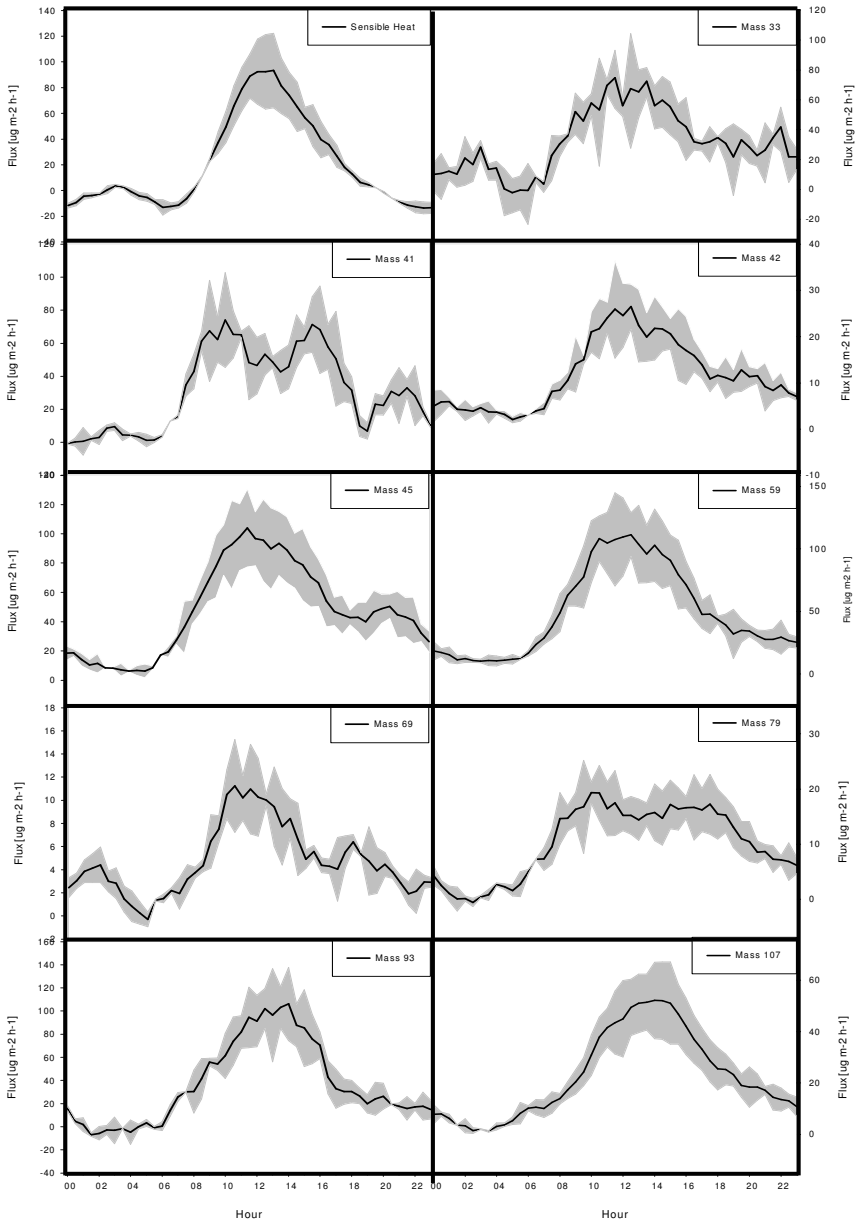
Acetone (M59)

Isoprene (M69)

Benzene (M79)

Toluene (M93)

Ethylbenzene (M107)



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NATURAL ENVIRONMENT RESEARCH COUNCIL

Urban Emissions

- Full analysis of data
- Calculate emission factors relative to CO_2 and CO
- Comparison with emission inventories
- Study chemical transformation (e.g. aerosol NO_3^- & secondary organic aerosol)

Summary

- Test database for model validation, especially inorganic aerosol / gas phase partitioning
  - Case studies & budgets from AMPEP flights
  - Campaign-based at European scale
  - Long-term for Auchencorth
  - Improved oxidation rates from AMPEP flights
- Model development & application
  - Frame inverse modelling
  - Validation of Model-3 / CMAQ
  - Adaptation of European EMEP model to 4 km UK scale

Outlook

EMEP:

- Make Auchencorth data available through CEH website
- Submission to EMEP
- Next intensive campaigns in 2008/09

Round-Britain flights:

- Proposal under review to put NH_3/HNO_3 instrument on FAAM aircraft

Modelling:

- Assess & improve inorganic chemistry modules of Models-3 and EMEP4UK
- Use EMEP4UK to assess the contribution of agricultural NH_3 emissions to UK PM