School of Earth and Environment
INSTITUTE FOR CLIMATE & ATMOSPHERIC SCIENCE

UNIVERSITY OF LEEDS

Nucleation and global aerosol: *Connecting AEROCOM and EUCAARI*

Ken Carslaw

Co-workers: Dominick Spracklen, Joonas Merikanto, Graham Mann, Carly Reddington

Thanks to Ari Asmi and EUCAARI data providers

http://researchpages.net/glomap

GLOMAP
Global Model of Aerosol Processes

EUCAARI questions concerning nucleation

UNIVERSITY OF LEEDS

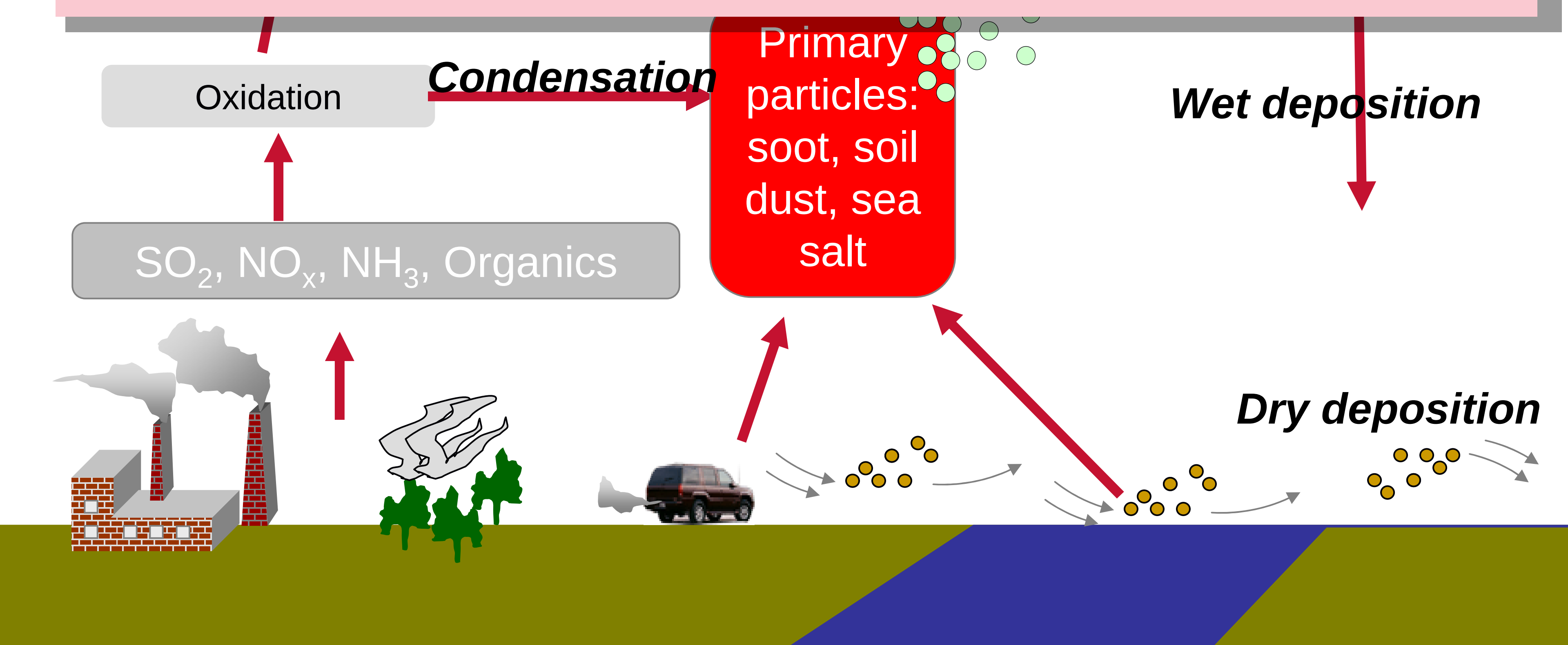
1. What are the relative contributions of primary and secondary aerosol to CCN?
2. How sensitive are different models to BL nucleation, and specifically the mechanism?
3. Can the intensive EUCAARI/EUSAR observations constrain the contribution of nucleation within the uncertainty of the mechanism and rate?

4. Similar set of questions for FT nucleation

Why is nucleation important?

UNIVERSITY OF LEEDS

1. Nucleation is a substantial source of CCN
2. <u>Changes</u> in CCN determine the aerosol indirect forcing
3. Nucleation is susceptible to many environmental changes (H_2SO_4, organics, condensation sink, T, RH, ions, etc) so long term aerosol radiative forcing is "interesting"



Our current understanding of nucleation (CN)

UNIVERSITY OF LEEDS

Atmos. Chem. Phys., 6, 5631–5648, 2006
www.atmos-chem-phys.net/6/5631/2006/

Atmospheric Chemistry and Physics

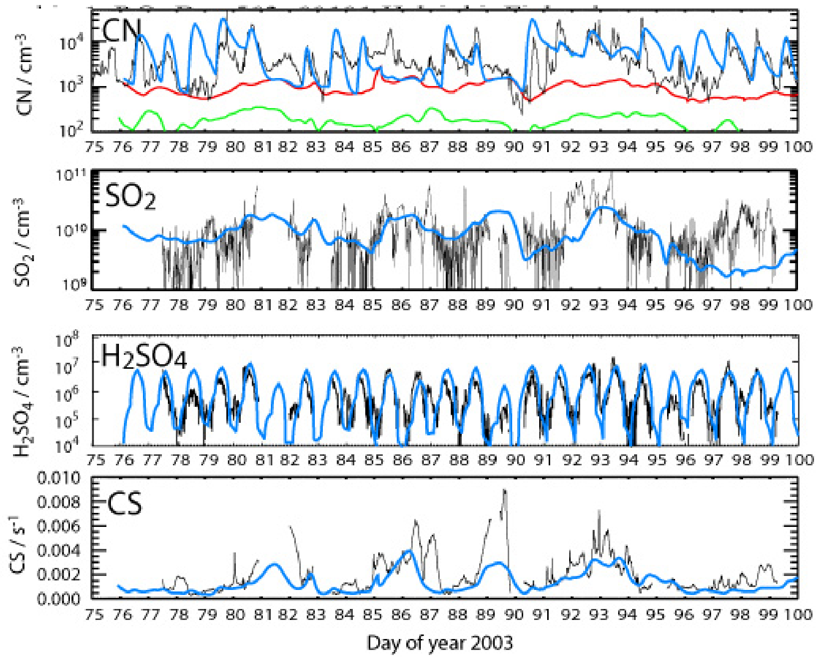
The contribution of boundary layer nucleation events to total particle concentrations on regional and global scales

D. V. Spracklen[1,*], K. S. Carslaw[1], M. Kulmala[2], V.-M. Kerminen[3], G. W. Mann[1], and S.-L. Sihto[2]

[1]School of Earth and Environment, University of Leeds, UK
[2]University of Helsinki, Department of Physical Sciences, P.O. Box 64, 00014 University of Helsinki, Finland
[3]Finnish Meteorological Institute, Climate and Global Change, Erik Palmenin
[*]now at: Atmospheric Chemistry Modelling Group, Harvard University, Caml

Received: 14 June 2006 – Published in Atmos. Chem. Phys. Discuss.: 3 Augu
Revised: 10 November 2006 – Accepted: 1 December 2006 – Published: 18 D

The predicted global distribution of particle formation events broadly agrees with what is expected from available observations. Over relatively clean remote continental locations formation events can sustain mean total particle concentrations up to a factor of 8 greater than those resulting from anthropogenic sources of primary organic and black carbon particles. However, in polluted continental regions anthropogenic primary particles dominate particle number and formation events lead to smaller enhancements of up to a factor of 2.



Our current understanding of nucleation (CCN)

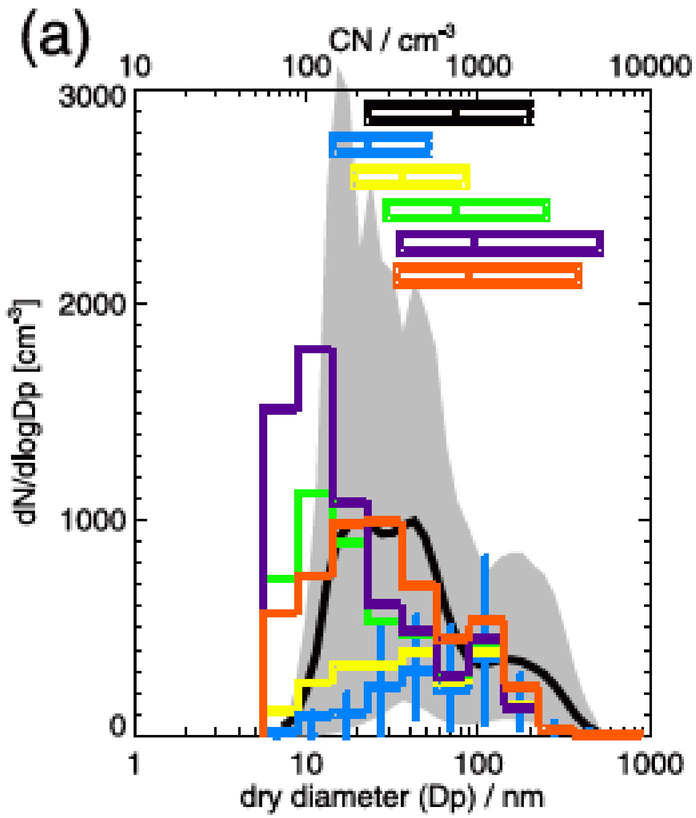
UNIVERSITY OF LEEDS

GEOPHYSICAL RESEARCH LETTERS, VOL. 35, LXXXXX, doi:10.1029/2007GL033038, 2008

Click Here for Full Article

Contribution of particle formation to global cloud condensation nuclei concentrations

Dominick V. Spracklen,[1] Kenneth S. Carslaw,[1] Joonas Merikant
Martyn P. Chipperfield,[1] Markku Kulmala,[2] Sanna-Liisa Sihto,[2]
Veli-Matti Kerminen,[3] Heikki Lihavainen,[3] Alfred Wiedensohler,



in Europe. Particle formation increases springtime BL global mean CCN (0.2% supersaturation) concentrations by 3–20% and CCN (1%) by 5–50%. Uncertainties in particle formation and growth rates must be reduced before the accuracy of these predictions can be improved. These

Our current understanding of nucleation (CCN)

UNIVERSITY OF LEEDS

Atmos. Chem. Phys., 9, 8601–8616, 2009
www.atmos-chem-phys.net/9/8601/2009/

Atmospheric Chemistry and Physics

Impact of nucleation on global CCN

J. Merikanto, D. V. Spracklen, G. W. Mann, S. J. Pickering, and K. S. Carslaw

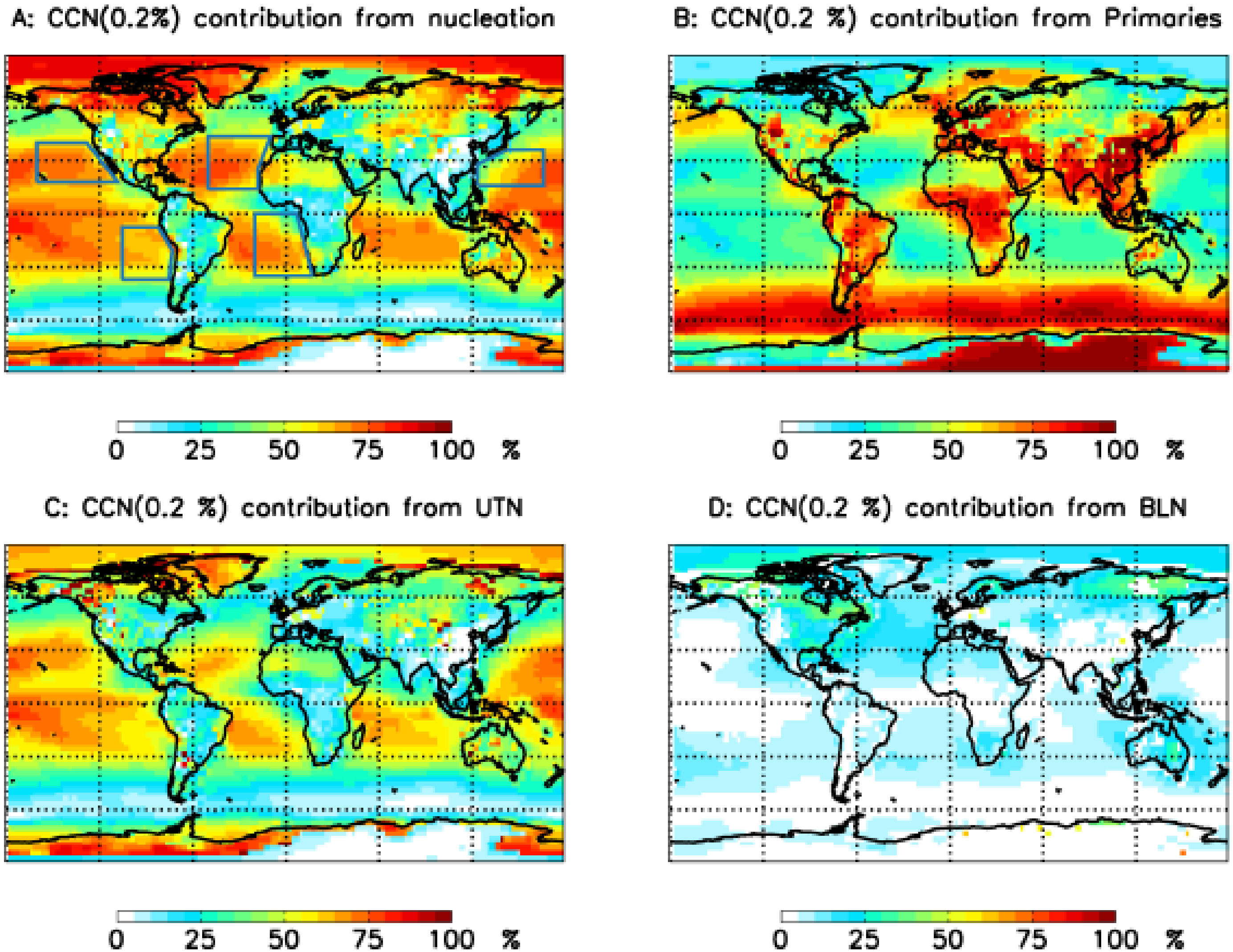
School of Earth and Environment, University of Leeds, Leeds, UK

Received: 27 April 2009 – Published in Atmos. Chem. Phys. Discuss.: 9 June 2009
Revised: 31 October 2009 – Accepted: 3 November 2009 – Published: 12 November 2009

boundary layer. We estimate that 45% of global low-level cloud CCN at 0.2% supersaturation are secondary aerosol derived from nucleation (ranging between 31–49% taking into account uncertainties in primary emissions and nucleation rates), with the remainder from primary emissions. The

CCN from different sources

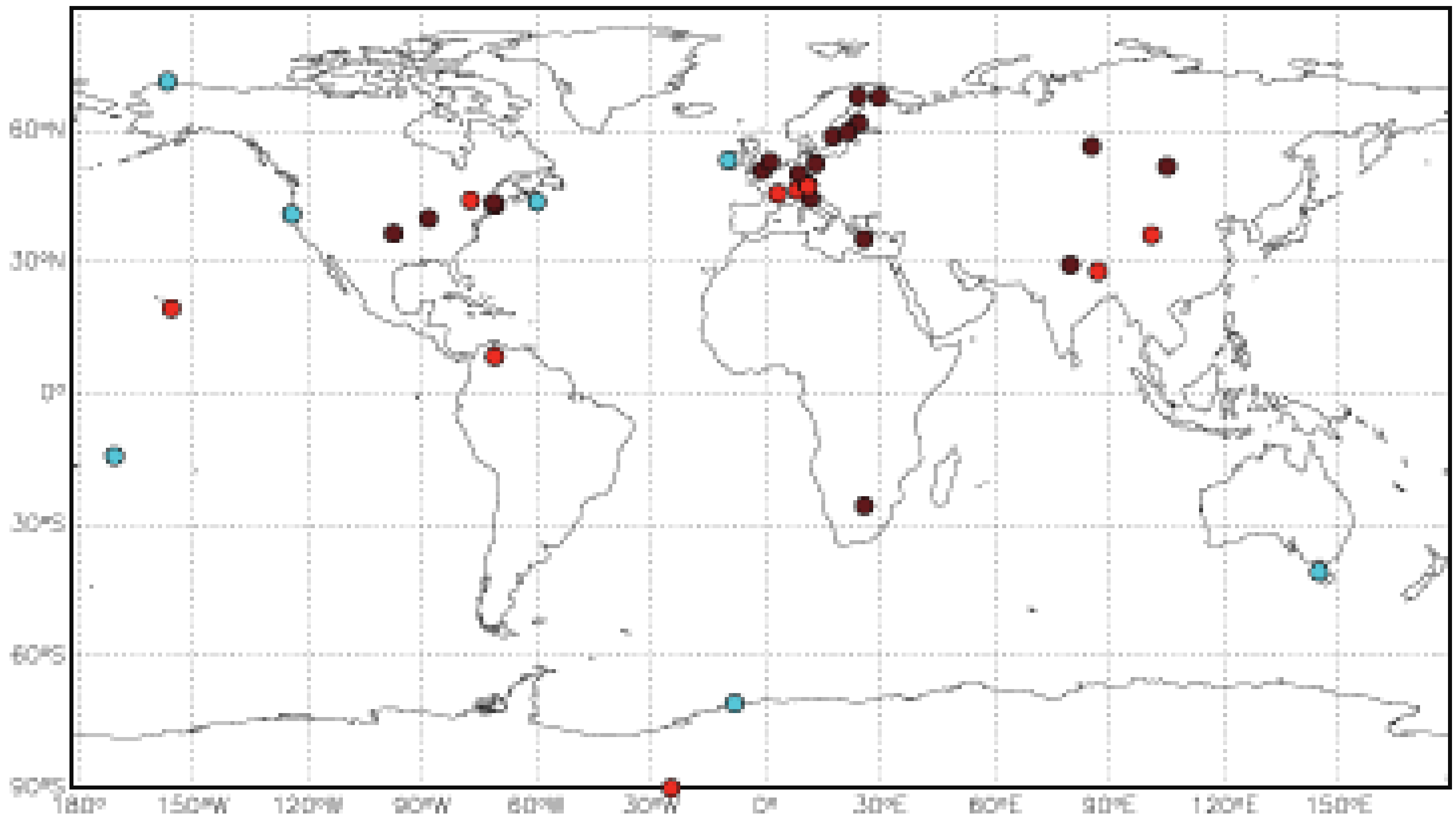
UNIVERSITY OF LEEDS



Global CN observations

UNIVERSITY OF LEEDS

36 surface stations with at least 1 year of CN data



Free trop

Marine boundary layer

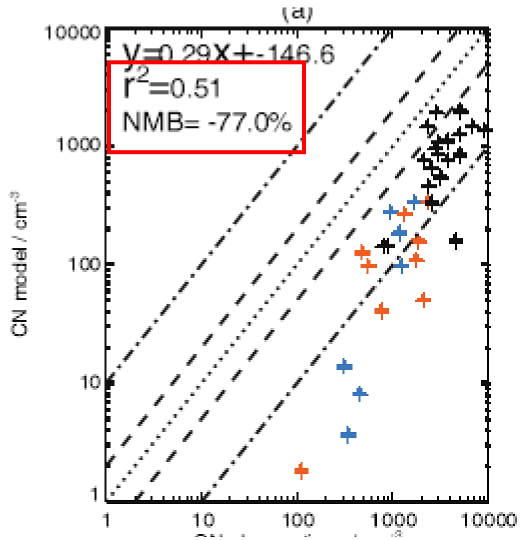
Continental boundary layer

Spracklen et al., ACP 2010

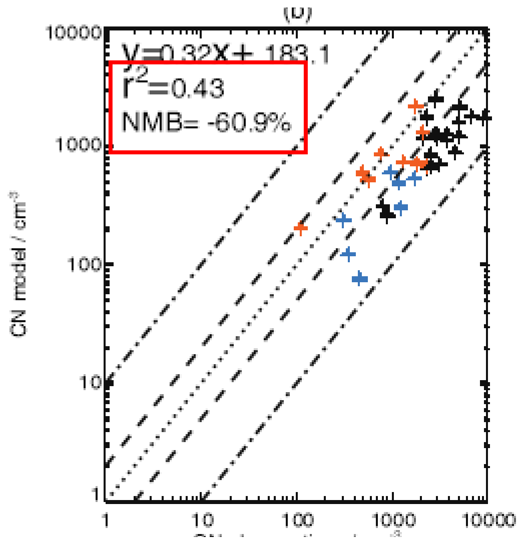
Predicted CN with different emissions and nucleation mechanisms

UNIVERSITY OF LEEDS

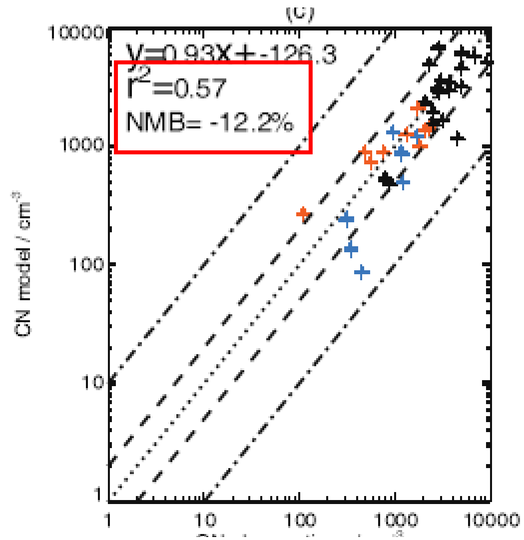
Primary emissions only



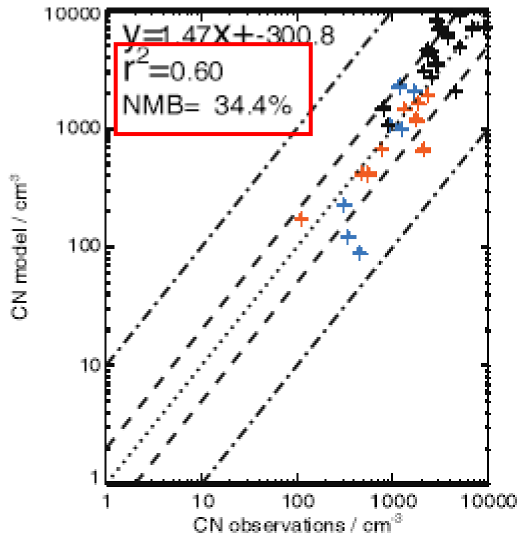
Primary + BHN



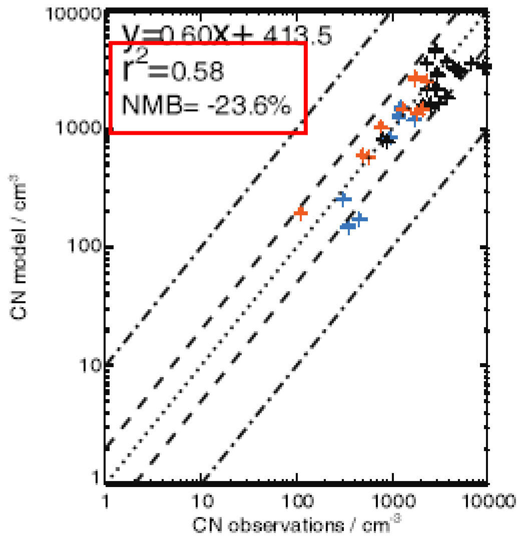
Increased EC/OC primary + BHN



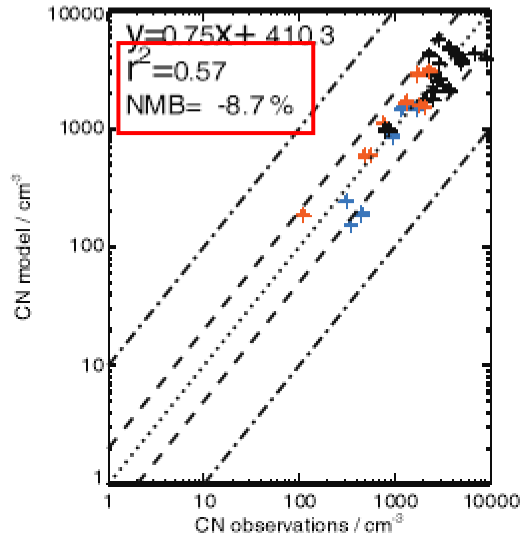
Primary + BHN + increased primary SO4



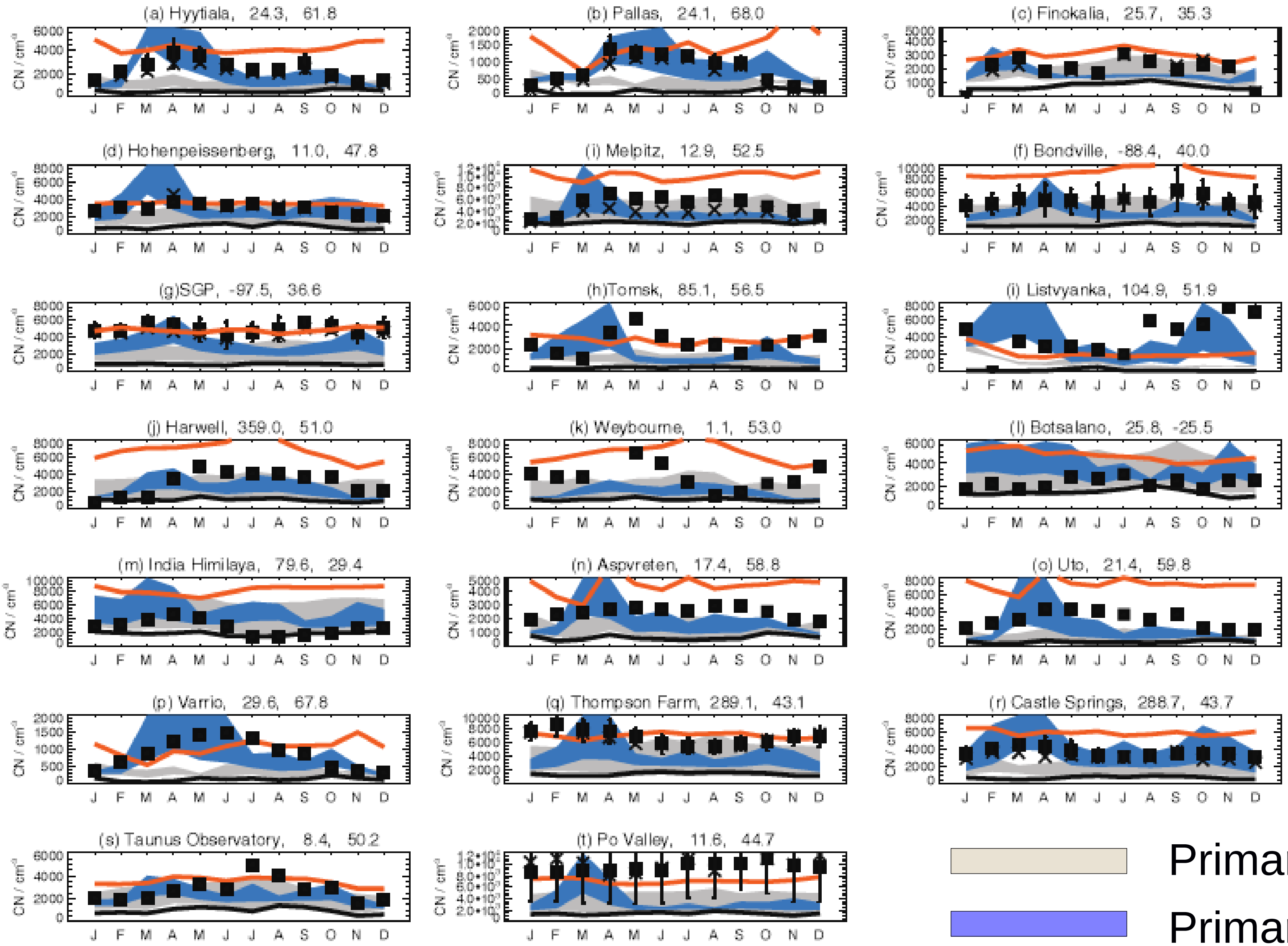
Primary + BHN + BLN-lin



Primary + BHN + BLN-kin



CN seasonal cycle at continental sites

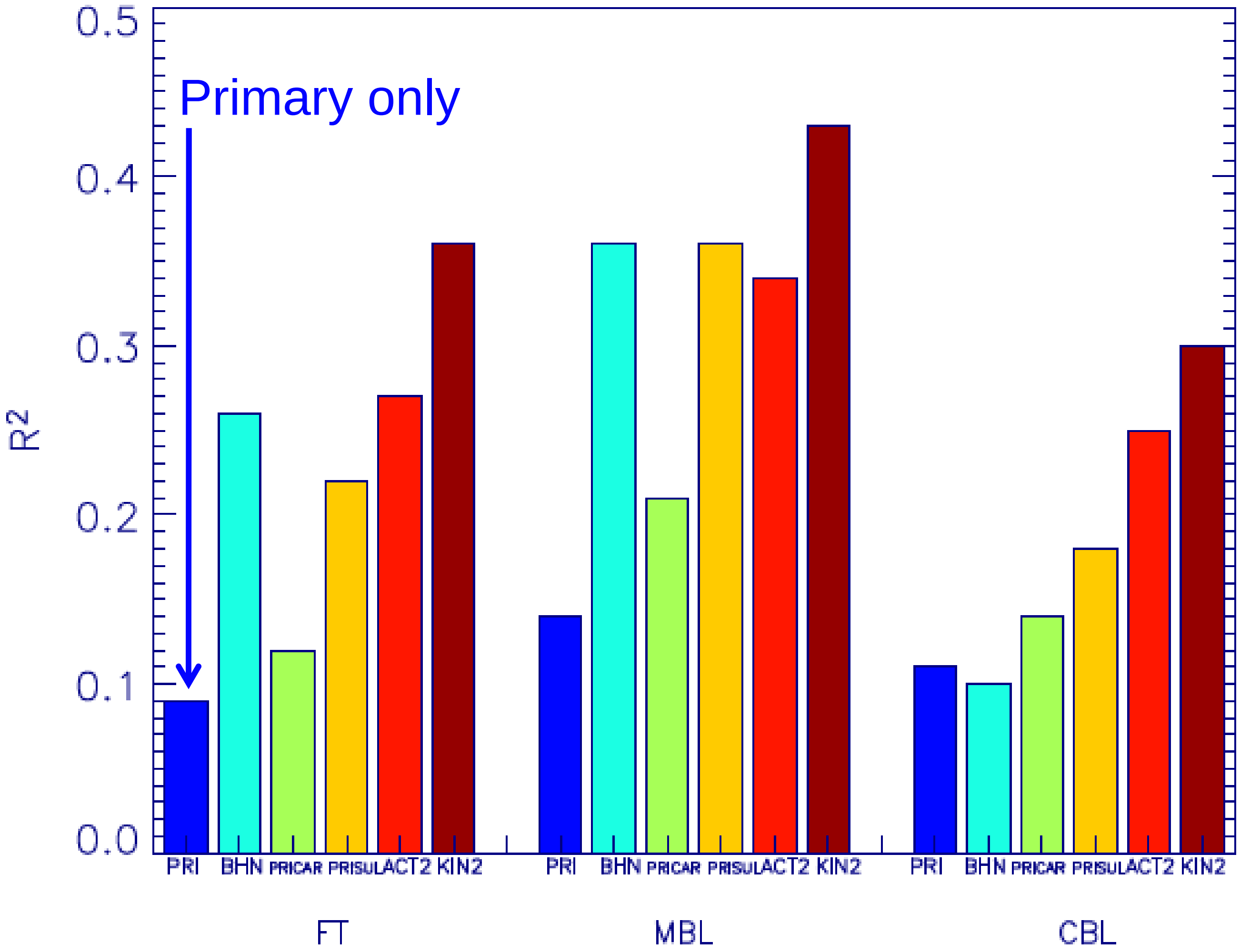


Primary + BHN

Primary + BHN + BLN

Correlation of seasonal CN variation (36 global sites)

UNIVERSITY OF LEEDS



Nucleation needed to explain seasonal CCN cycle

More difficult to identify best mechanism

Spracklen et al., ACP 2010

Approach

1. Test several new as well as existing “mechanisms” from EUCAARI in a range of models
2. Compare models against each other and observations, focusing on European BL

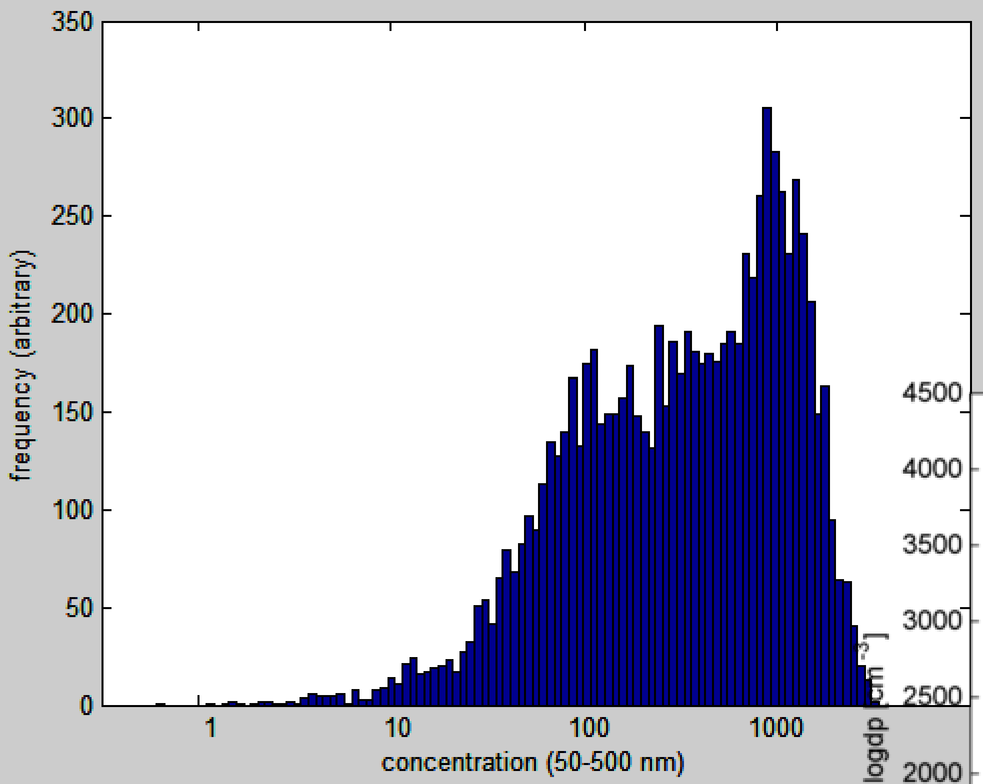
Key quantities and challenges

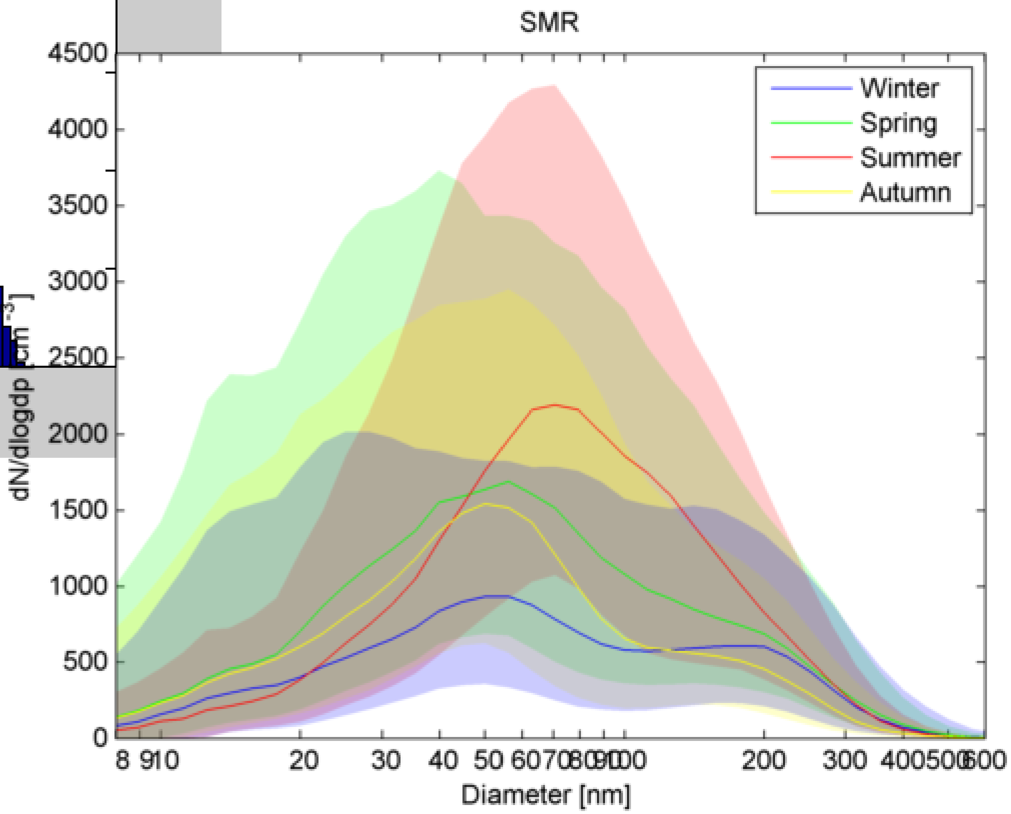
1. Emphasise N50, N100 etc more than N3 (climate-relevant)
2. Nx-50 to test nucleation realism
3. Seasonal / diurnal cycle, variability (e.g., N50 pdf)
4. Needs output at fairly high time resolution (~hours)

5. Extension of the AEROCOM microphysics experiments

Example EUSAR data

UNIVERSITY OF LEEDS





Plots from Ari Asmi

First analysis of EUCAARI/EUSAR data

UNIVERSITY OF LEEDS

May 2008 campaign

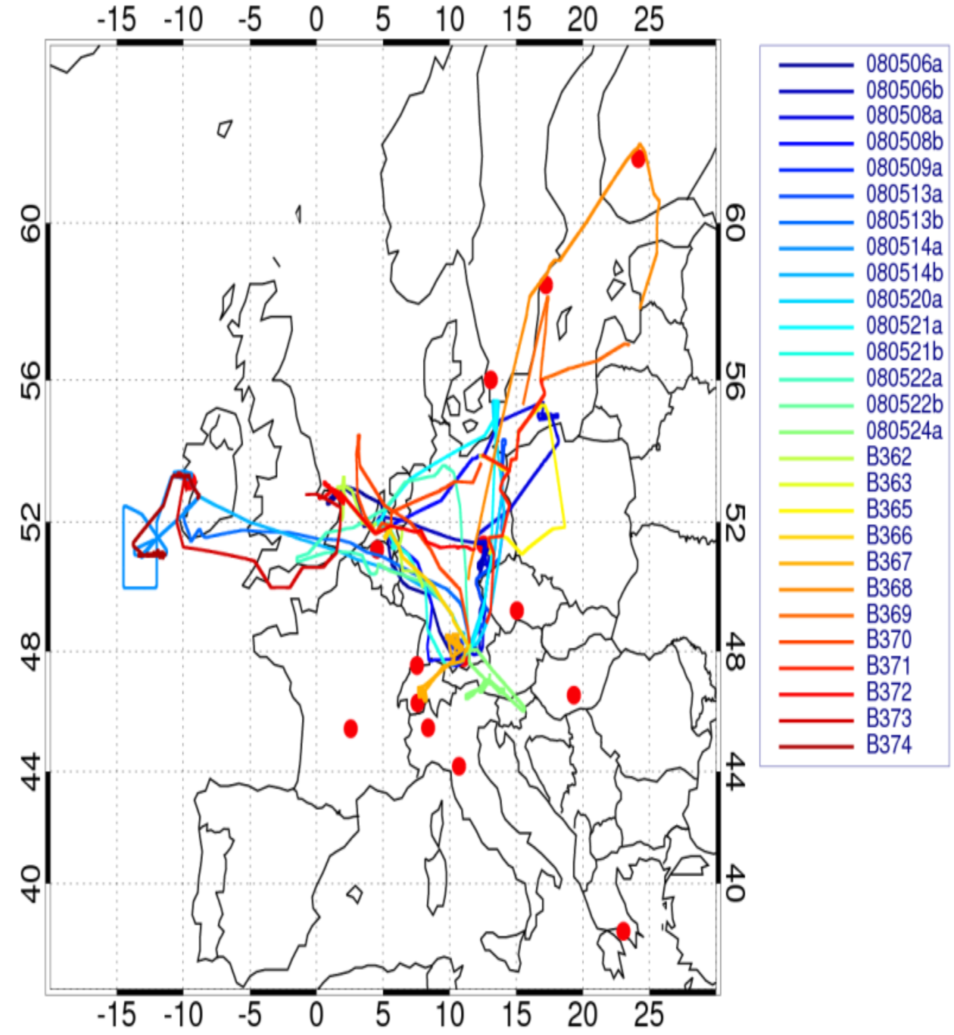
~2 weeks of stable anticyclone, very polluted, followed by weak frontal period

Falcon, ATR and Bae-146 aircraft data

CN, size distributions, BC, non-volatiles, etc

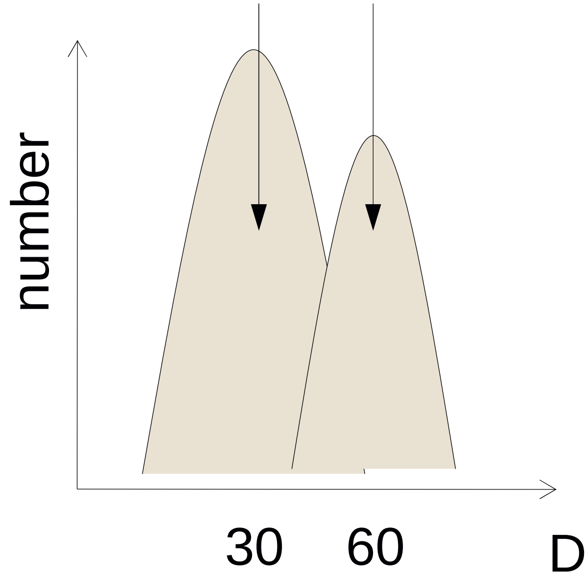
15 EUSAR ground stations

CN, size distributions…



Questions for the EUCAARI analysis

1. What is the contribution of BL nucleation to N3, N50, N100, N160?
2. Is there a statistically significant contribution within the uncertainty of
    1. The size of primary emitted BCOC particles,
    2. The nucleation mechanism?

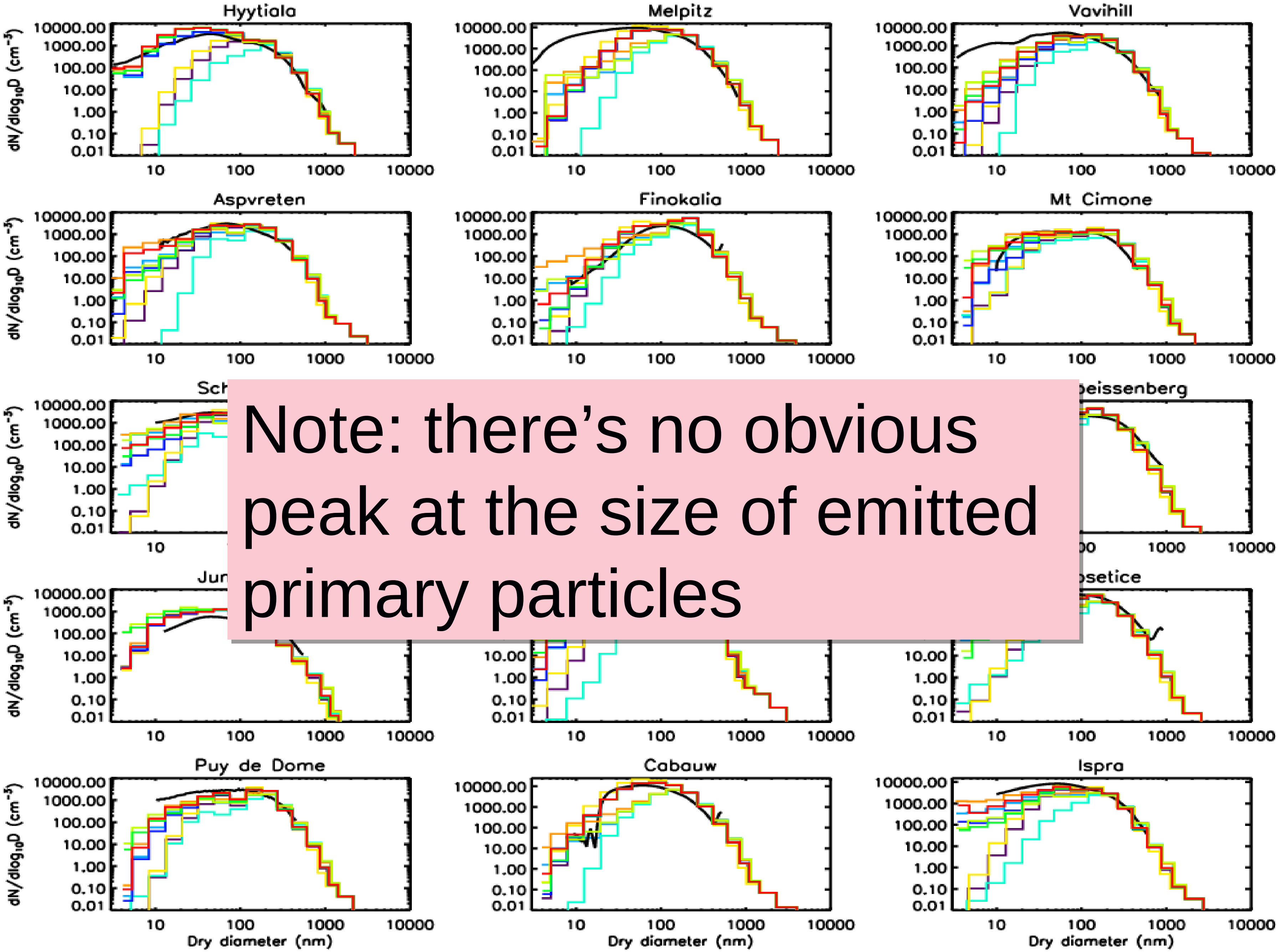


$$J_{nuc} = A[H_2SO_4]$$

$$J_{nuc} = K[H_2SO_4]^2$$

$$J_{nuc} = k[H_2SO_4][\text{organic}]$$

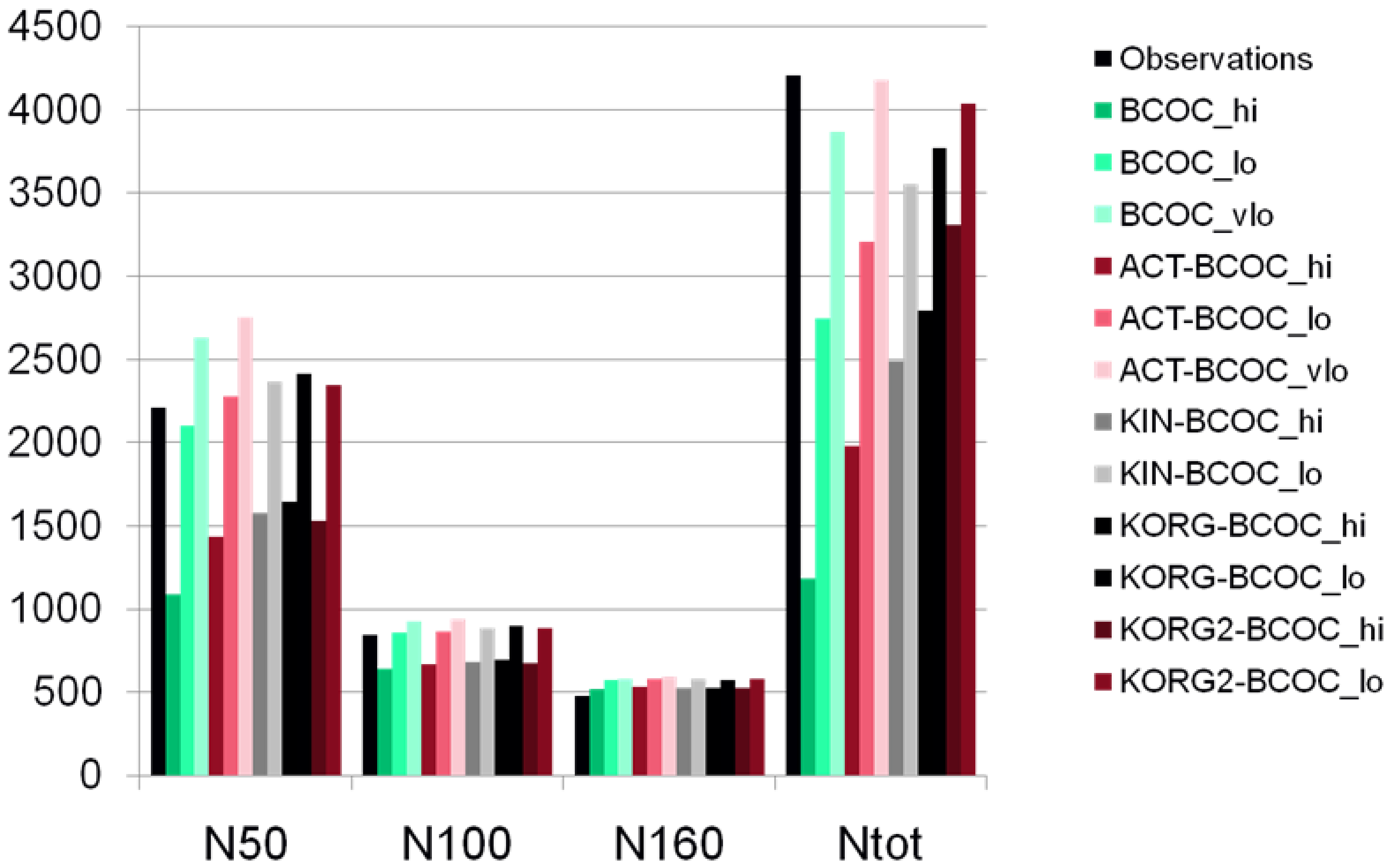
$$J_{nuc} = k_1[H_2SO_4]^2 + k_2[H_2SO_4][\text{organic}]$$



Note: there’s no obvious peak at the size of emitted primary particles

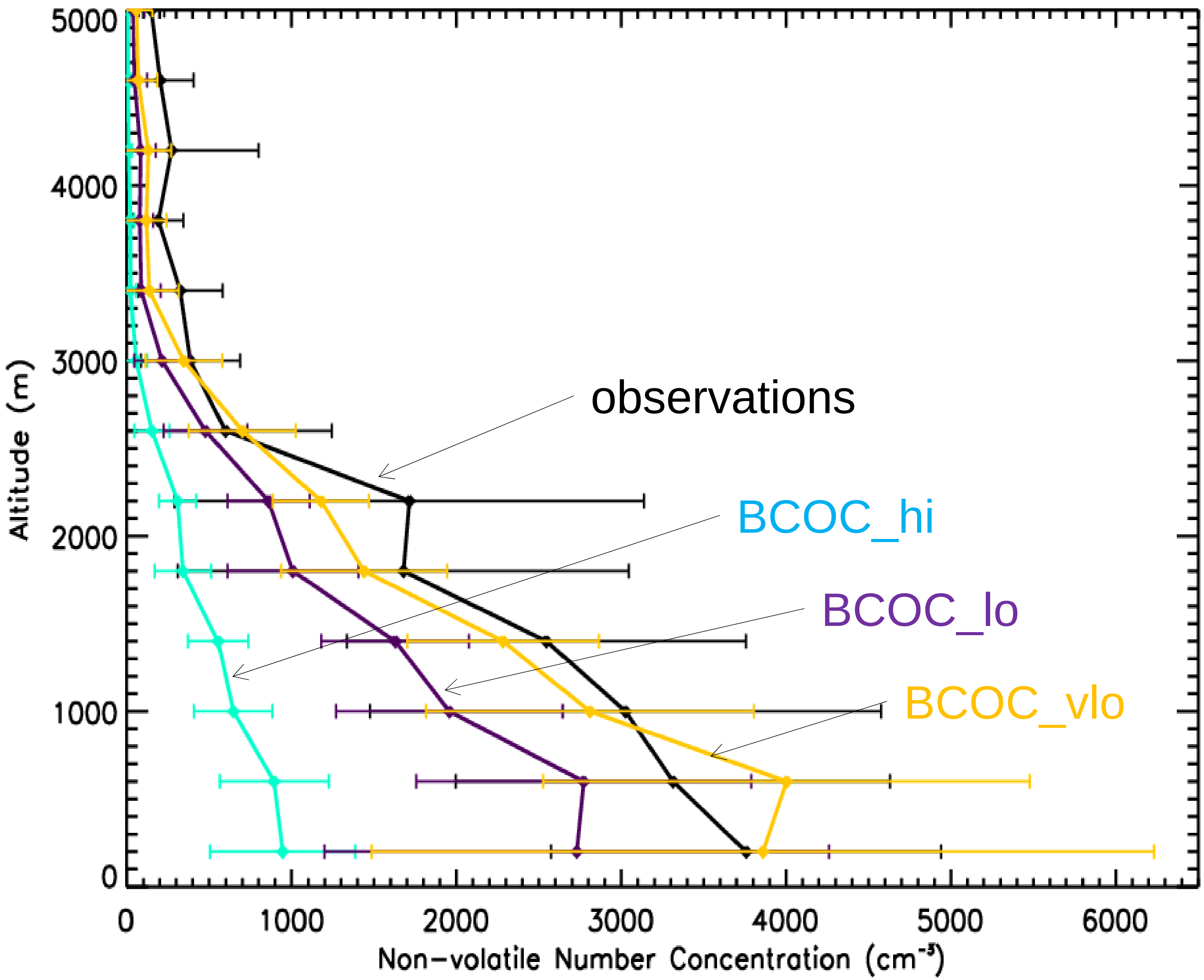
All-site monthly mean

UNIVERSITY OF LEEDS



Non-volatile particles >14 nm
(Falcon all-flight mean vertical profile)

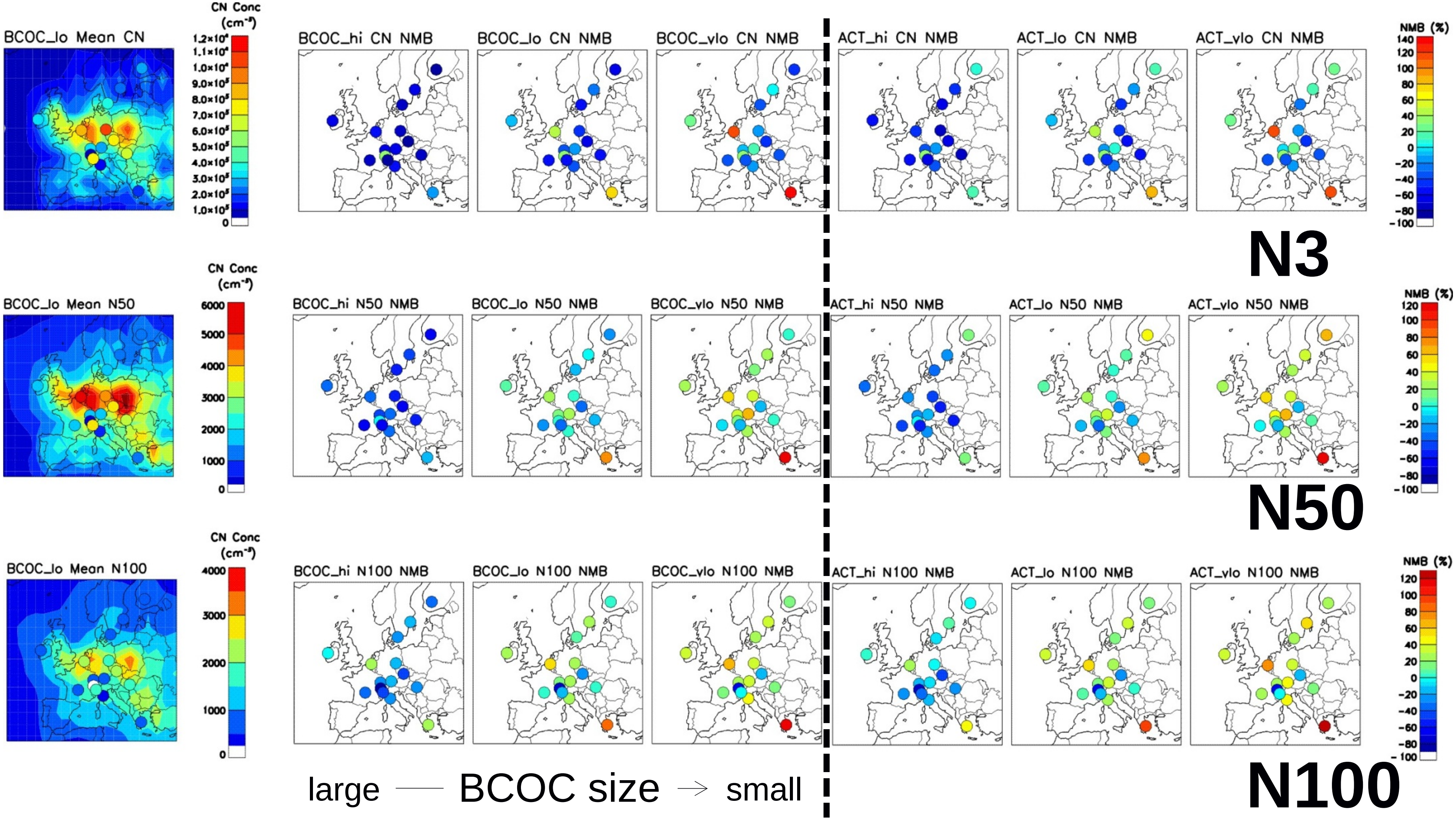
UNIVERSITY OF LEEDS



Sensitivity to BCOC emission sizes

UNIVERSITY OF LEEDS

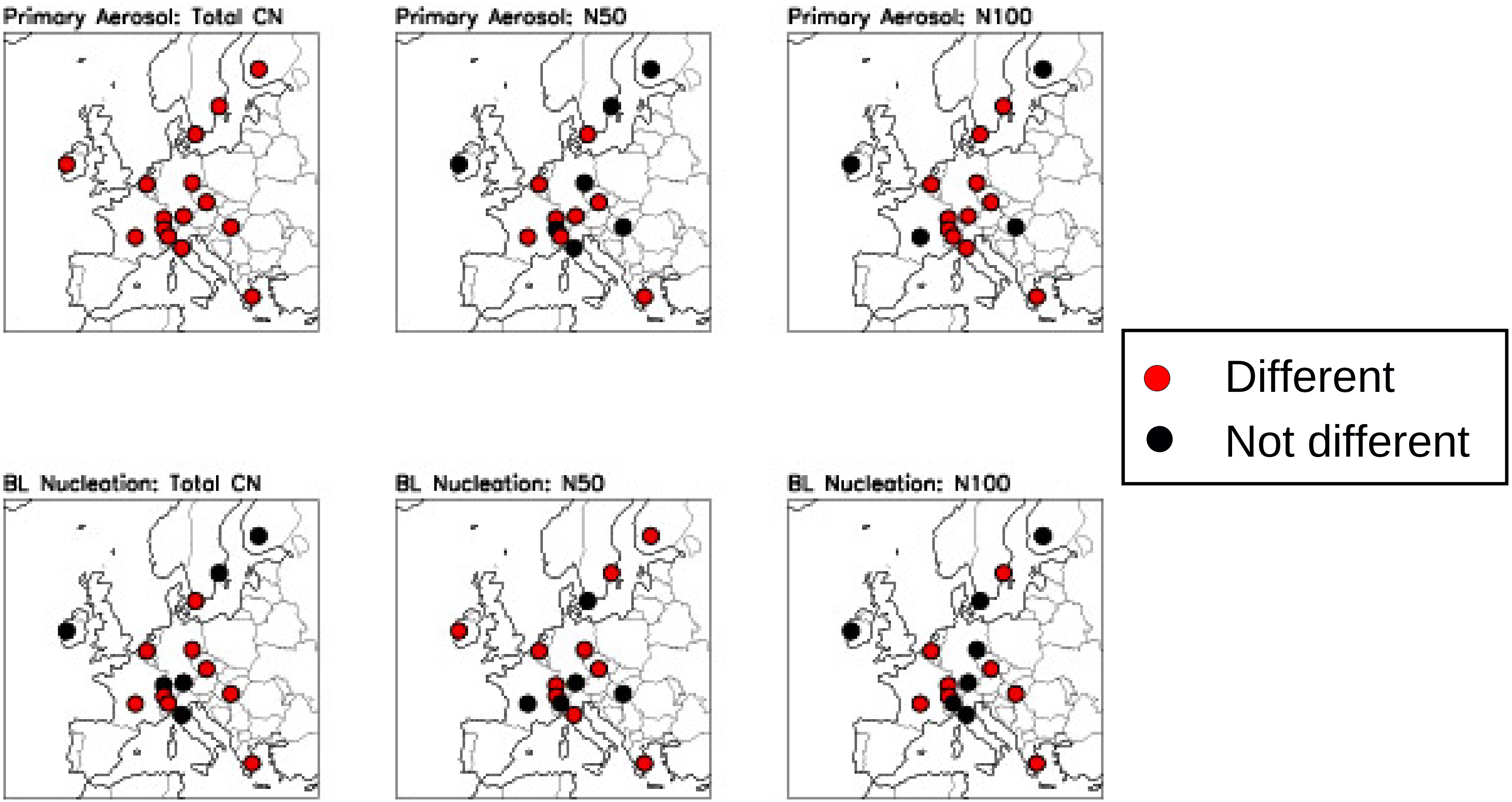
Colour of dot shows model-observation Normalised Mean Bias



Significance of model-observation differences

UNIVERSITY OF LEEDS

Compare model and observation hourly time series. Where are they significantly different at 95% level?



Summary numbers

UNIVERSITY OF LEEDS

Adding BL nucleation increases N50 by 5-50%, but this is statistically significant against the data at only 3 out of the 15 sites

N100 increases by 3-17% and is significant at 5 sites

The EUCAARI / AEROCOM BL nucleation intercomparison

UNIVERSITY OF LEEDS

1. What are the relative contributions of primary and secondary aerosol to CCN?
2. How sensitive are different models (CCN, AOD) to BL nucleation, and specifically the mechanism?

Model experiments

1. 2008 nudged with 2000 emissions
2. 4 simple BL nucleation expressions, or none

Evaluation Data

1. 2008 EUSAR data, global 36 station CN
2. Focus on N3, N50, N100

Timeline

EUCAARI report needed by ~February 2011

Protocol to be sent out very soon