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Lessons learnt from the first EMEP intensive measurements: June 2006 and January 2007

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+ many others**

TFMM workshop, JRC, 19-20 November 2009



EMEP Intensive measurement periods 2006-2007

- Focus on:
 - **PM mass closure**
 - **Artefact free measurements of gaseous and particulate nitrogen species**
 - **High temporal resolution data (diurnal variations)**

	mass		daily						Intensive, hourly			
	mass		Inorg		EC/OC		Crust		Inorg		EC/OC	
	June	Jan	June	Jan	June	Jan	June	Jan	June	Jan	June	Jan
AT02	PM10,PM2.5,PM1	PM10,PM2.5,PM1	FP	FP						SO4	PM2.5	
CH02	PM10,PM2.5,PM1	PM10,PM2.5,PM1	FP, PM10, PM1						AMS	AMS	PM2.5	PM2.5
CZ03	PM10, PM2.5	PM10,PM2.5			PM10	PM10						
DK41	PM10,PM2.5, PM1	PM10,PM2.5, PM1							NOx,O3	NOx,O3		
DE02	PM10,PM2.5,PM1	PM10,PM2.5,PM1	FP									
DE03	PM10, PM2.5	PM10, PM2.5	FP									
DE07	PM10,PM2.5	PM10,PM2.5	FP									
DE43	TSP	TSP		Berner							BC	BC
DE44	PM10,PM2.5,PM1 + Berner (5 sizes)		X	X	X	X				AMS		AMS (OM)
ES31	PM10,PM2.5, PM1	PM10,PM2.5, PM1	PM10 & PM2.5		TC	TC	PM10 & PM2.5					
FI17	PM10,PM2.5,PM1	PM10,PM2.5,PM1	X	X								
HU02	PM10				TC							
IE31									AMS		AMS	
IT01	PM10,PM2.5	PM10,PM2.5	X	X	X	X	X	X				
IT04	PM10,PM2.5	PM10,PM2.5	PM2.5	PM2.5	PM2.5	PM2.5			X	X		
NL11	PM10, PM2.5	PM10,PM2.5							X	X		
NO01	PM10,PM2.5,PM1	PM10,PM2.5,PM1	X	X	X	X						
SE12	PM2.5,PM1				X	X						
GB33									AMS		AMS (OM)	
GB36	PM10,PM2.5								X			
GB40									X	X		

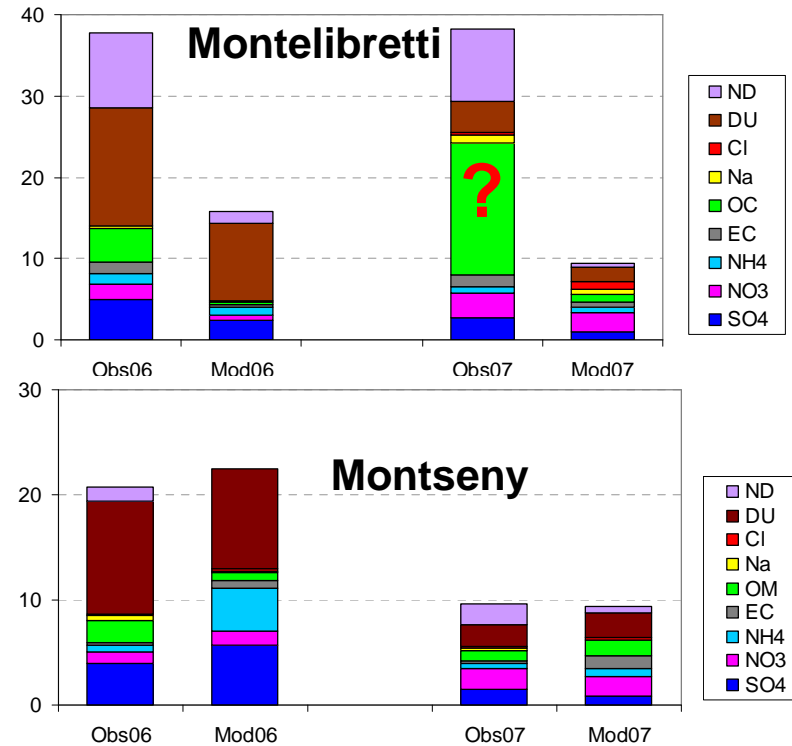
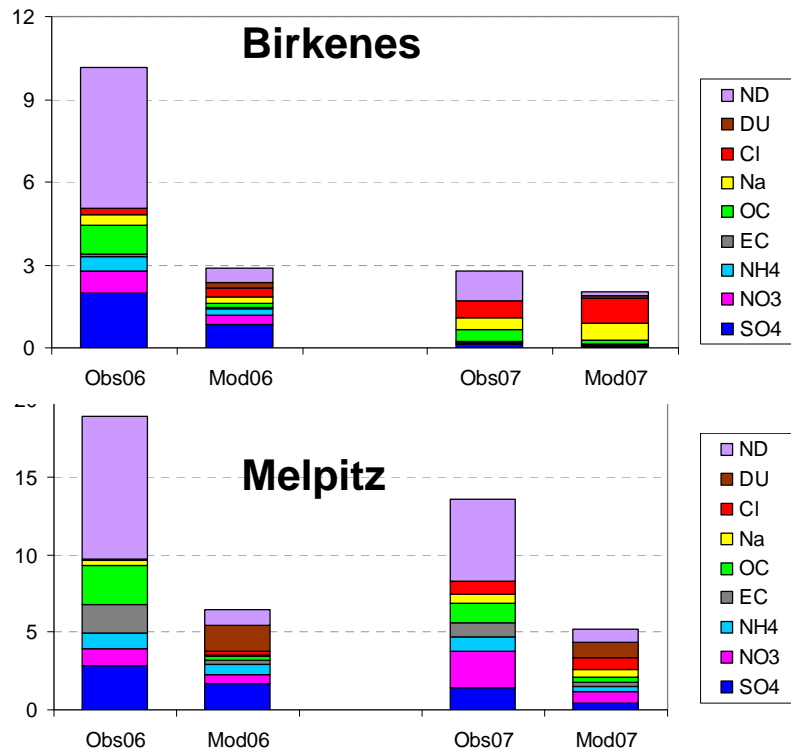


PM mass balance

**Mass closure for PM_{10} and $PM_{2.5}$
at four(5) sites with measurements of all
inorganic and carbonaceous components:**

**Birkenes, Melpitz, Montseny, Montelibretti,
Ispra($PM_{2.5}$)**

PM10 chemical speciation ($\mu\text{g}/\text{m}^3$)



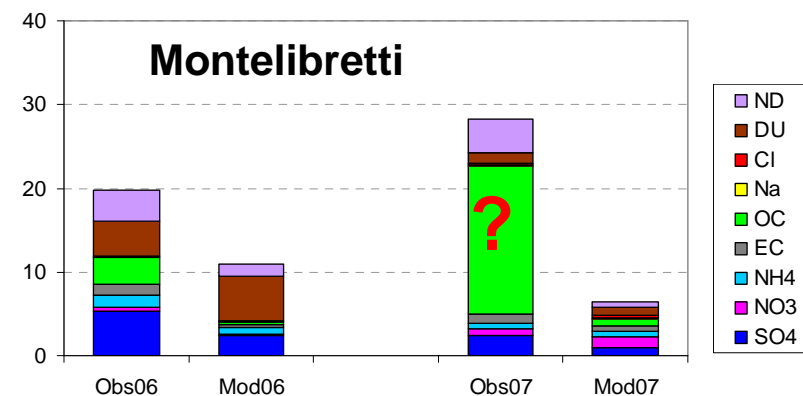
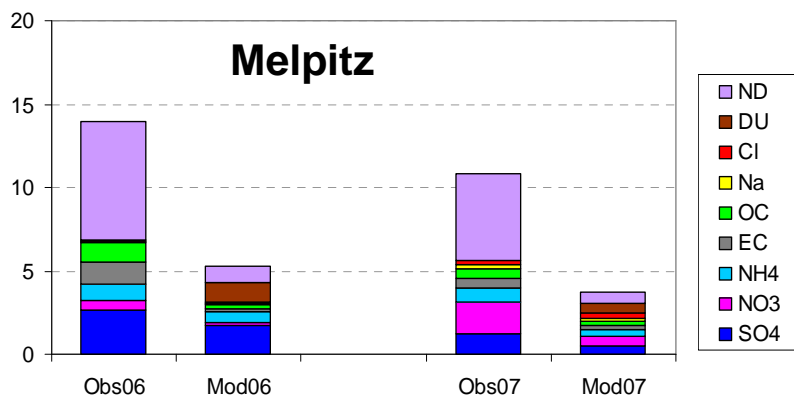
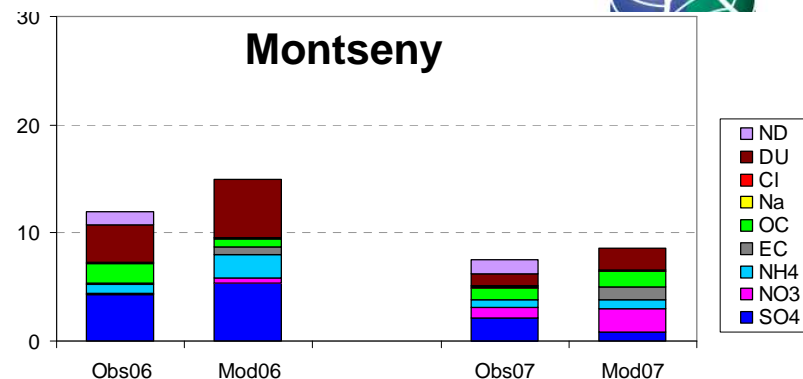
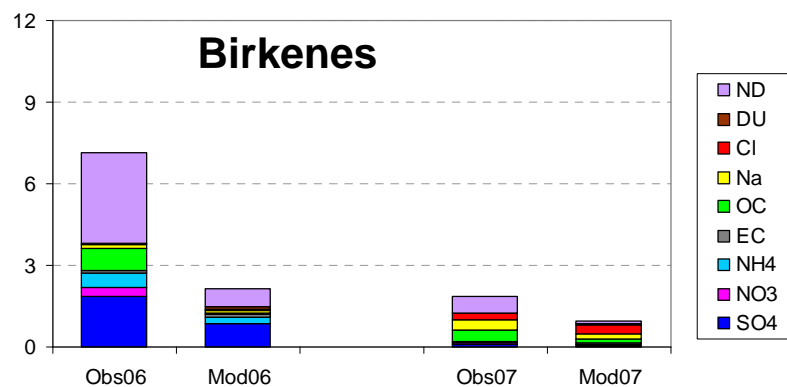
➤ Model underestimates of PM_{10} *)... notice 20-30% of measured PM_{10} is “Not Determined” (50% at NO01 and DE44). Note! modelled OC = anthrop. primary OC

➤ The model tends to underestimate all PM components (exc. at ES17)

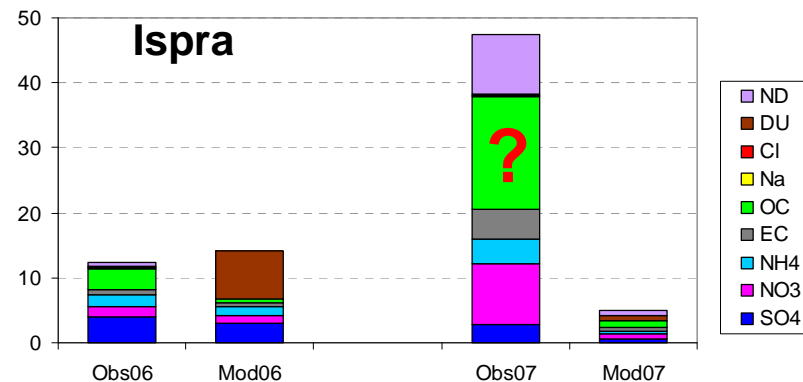
➤ Higher in summer than in winter (IT), larger difference for PM_{10} (but less pronounced in the model)

➤ Carbonaceous matter (EC + OC) main reason for underestimation of PM mass (ND)

PM2.5 chemical speciation



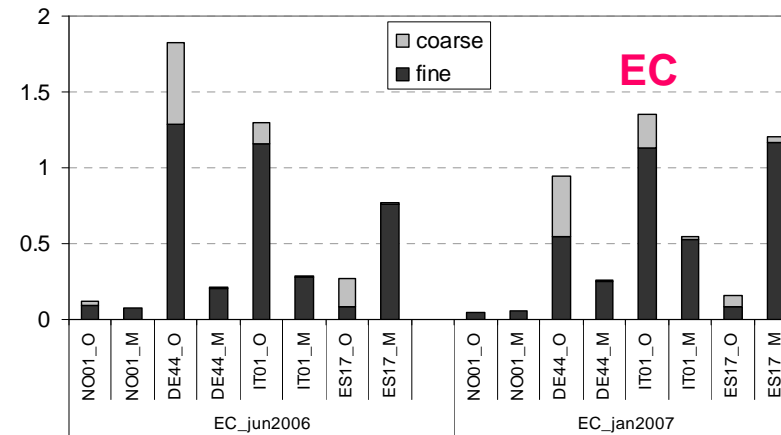
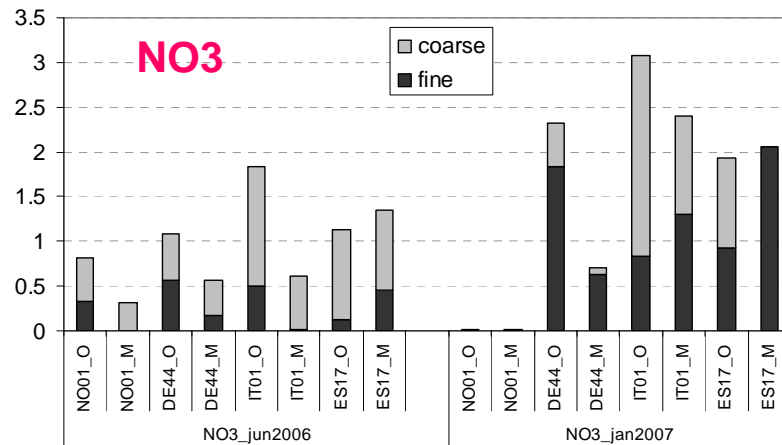
- Smaller contribution from sea salt and mineral dust than in PM₁₀
- EC results consistent with EMEP EC/OC Campaign 2002
- Na⁺ underestimated, mostly in summer
- Mineral dust, mixed results, mostly within +/- 35%



PM size distribution:



Distribution of NO₃⁻, EC, between fine and coarse aerosols from model and measurements



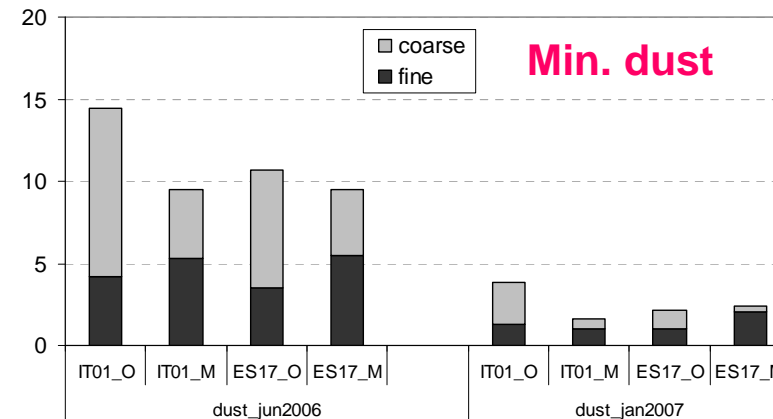
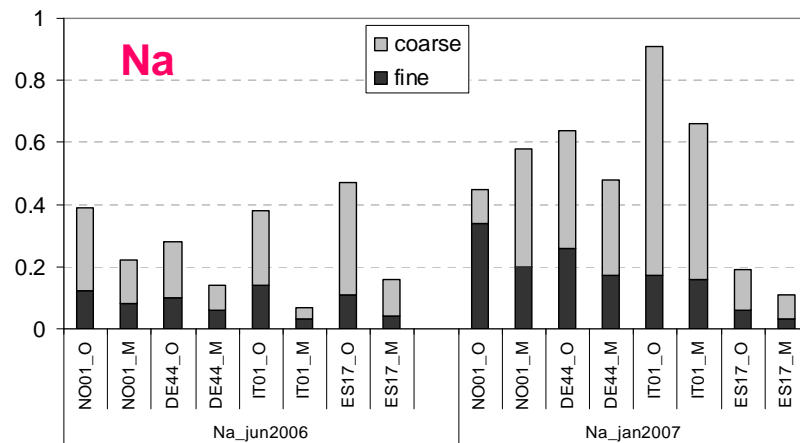
- In observations, a smaller NO₃⁻ mass occurs on fine aerosol in summer (between 10 and 50%) than in winter (between 30 and 80%) - reflected in the model results, but discrepancies can be significant for the individual sites.
- Too little coarse EC in the model results compared to observation, which is due to large uncertainties (underestimation and missing sources) in emission data.

PM size distribution:



Distribution of Na⁺ and mineral dust between fine and coarse aerosols from model and measurements

- Na⁺ size distribution is relatively well described by the model, with between 10 and 40% mass in the fine mode (probable measurement problems in January 2007 at NO01).
- For mineral dust, the model tends to underestimate the mass of coarse particles



Artefacts and incompleteness in the measurement data complicates the analysis of results!



Examples:

Rather poor data coverage at ES17 (between 1 and 6 days during each of the campaign periods) and not the same for the different components.

See problem with the SO₄, NO₃ and NH₄ measurements for January 2007 at NO01: PM_{2.5} and PM₁₀ measurements correlate poorly with the regular filter pack measurement going in parallel. The filter pack measurements correlate better with the EMEP model.

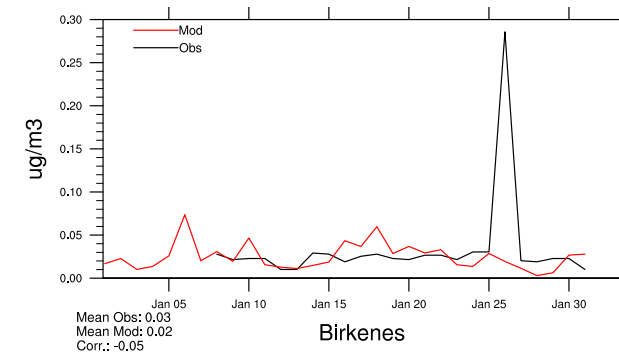
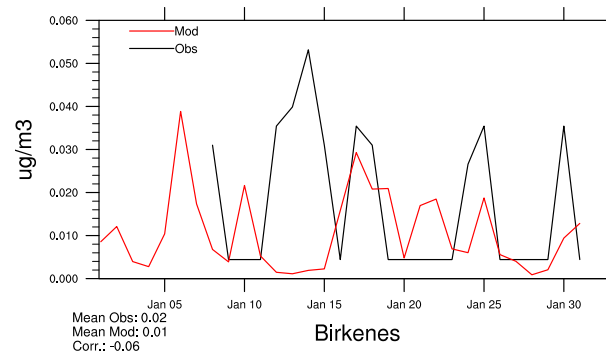
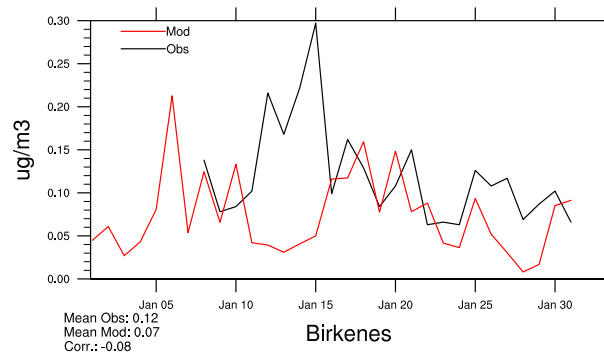
Extremely high the winter levels of OC at the Italian sites which are much higher than any of the other sites and 2-5 times higher than the level in summer, both in PM₁₀ and PM_{2.5}

Probable artefact in the chloride measurements due to evaporation of HCl, as the Na/Cl ratio is often much higher than the sea salt ratio

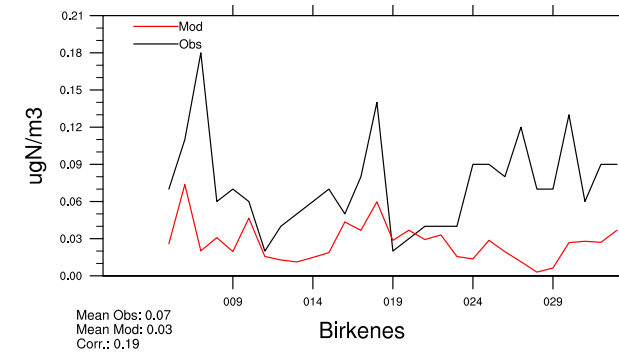
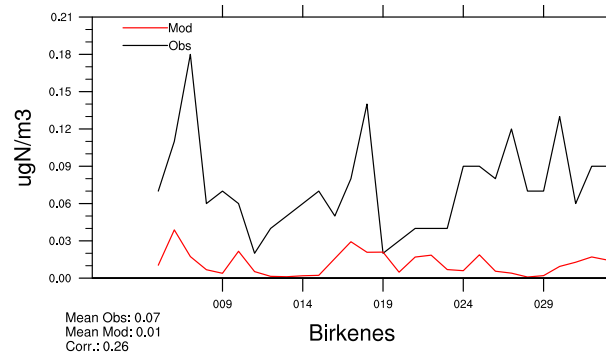
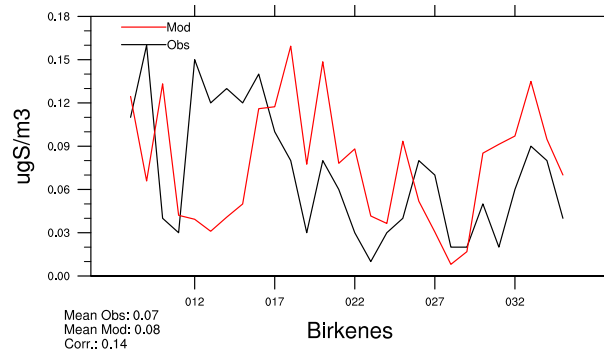


Inconsistent measurement data at Birkeness

Intensive measurements



EMEP network (filter pack)



Artefacts and incompleteness in the measurement data complicates the analysis of results!



Examples:

Rather poor data coverage at ES17 (between 1 and 6 days during each of the campaign periods) and not the same for the different components.

See problem with the SO₄, NO₃ and NH₄ measurements for January 2007 at NO01: PM_{2.5} and PM₁₀ measurements correlate poorly with the regular filter pack measurement going in parallel. The filter pack measurements correlate better with the EMEP model.

Extremely high the winter levels of OC at the Italian sites which are much higher than any of the other sites and 2-5 times higher than the level in summer, both in PM₁₀ and PM_{2.5}

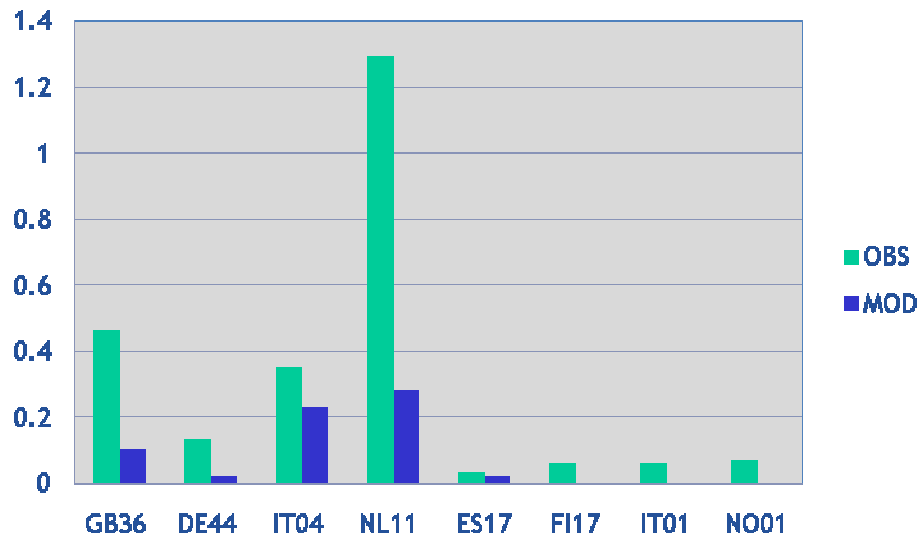
Probable artefact in the chloride measurements due to evaporation of HCl, as the Na/Cl ratio is often much higher than the sea salt ratio



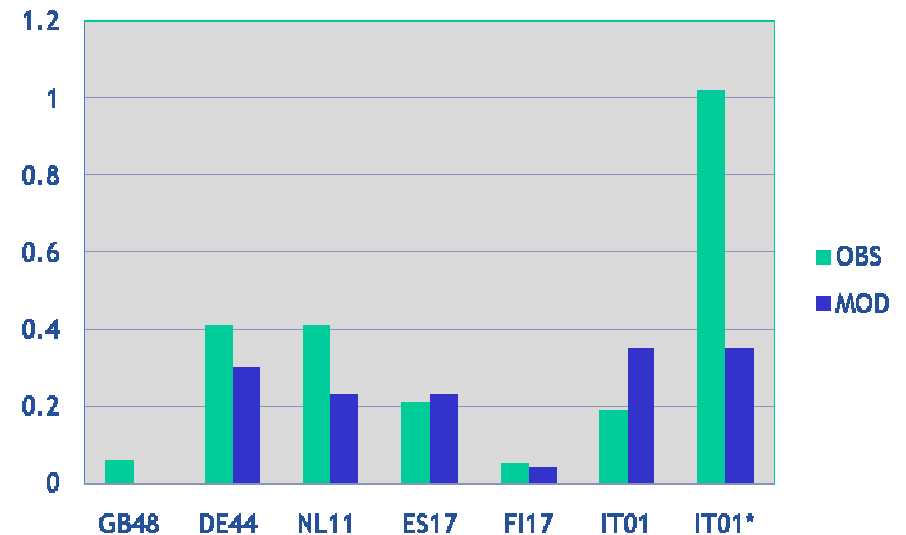
Aerosol/gas partitioning:

- NH_3 - NH_4^+ split modelled well both in summer and winter
- More problems with nitrate

Fine nitrate (PM_{2.5})



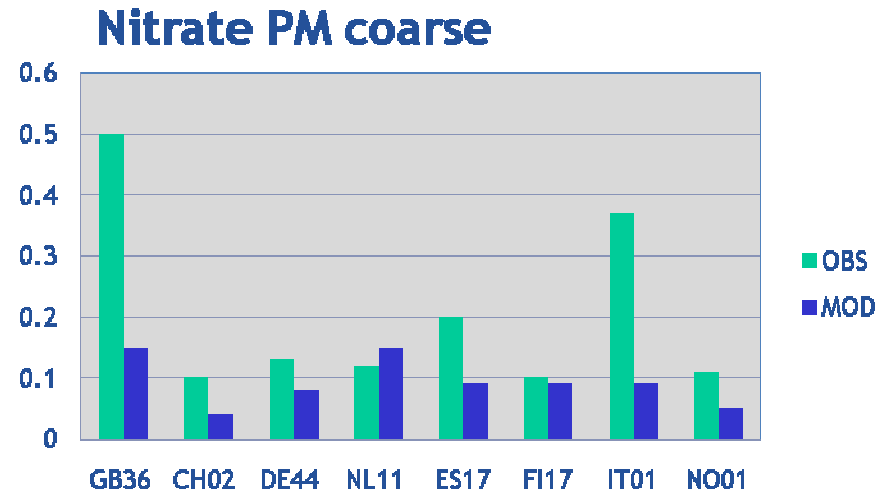
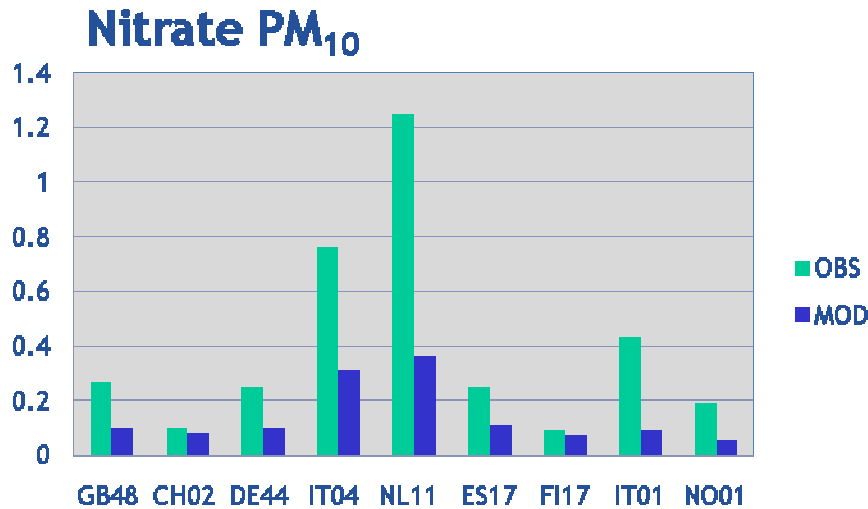
June 2006



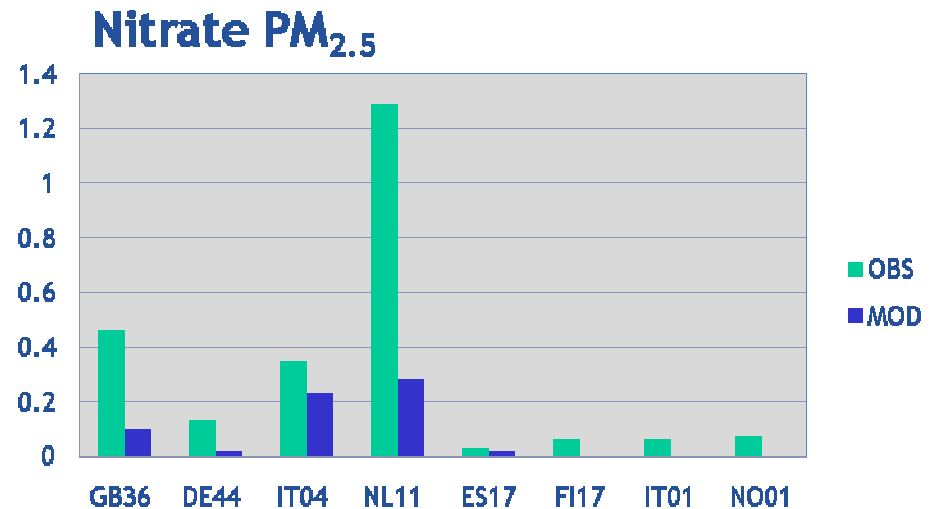
January 2007

- Underestimation in summer and mixed in winter
- HNO₃ mixed in summer, low in winter

Nitrate in different size fractions

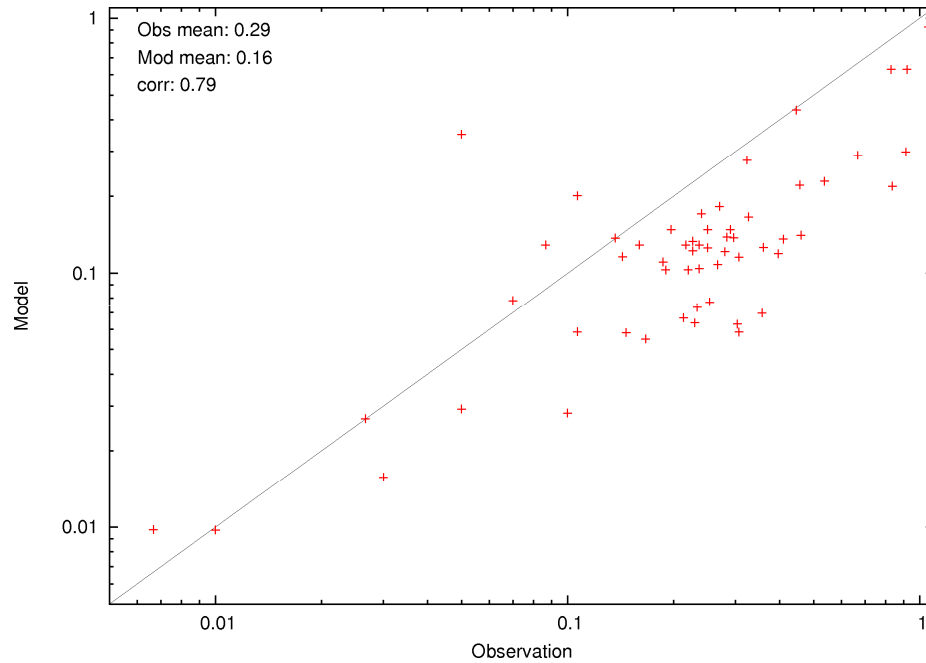


PM₁₀ nitrate underestimated, mostly because of underestimation of fine nitrate

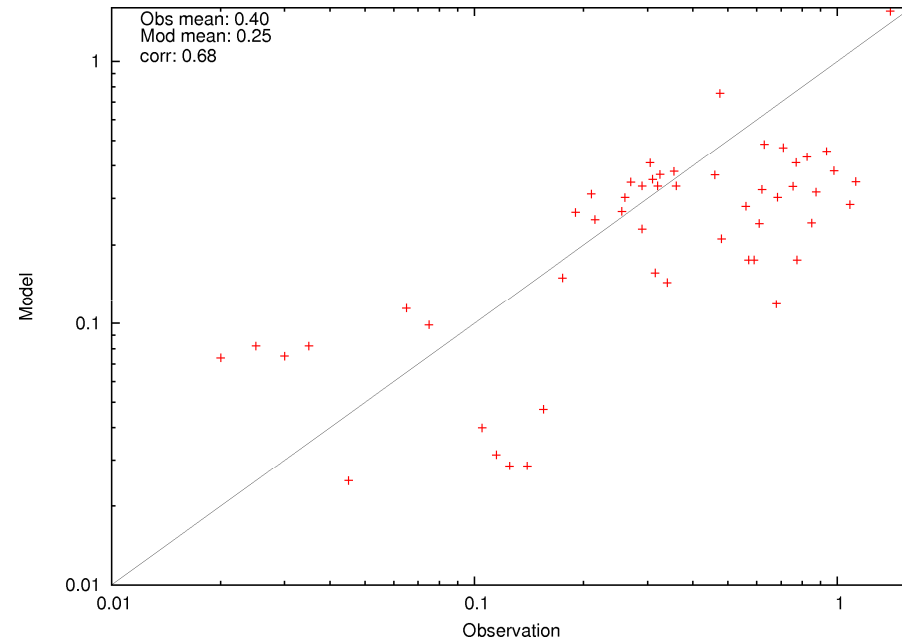


Seasonal performance, nitrate

Summer



Winter

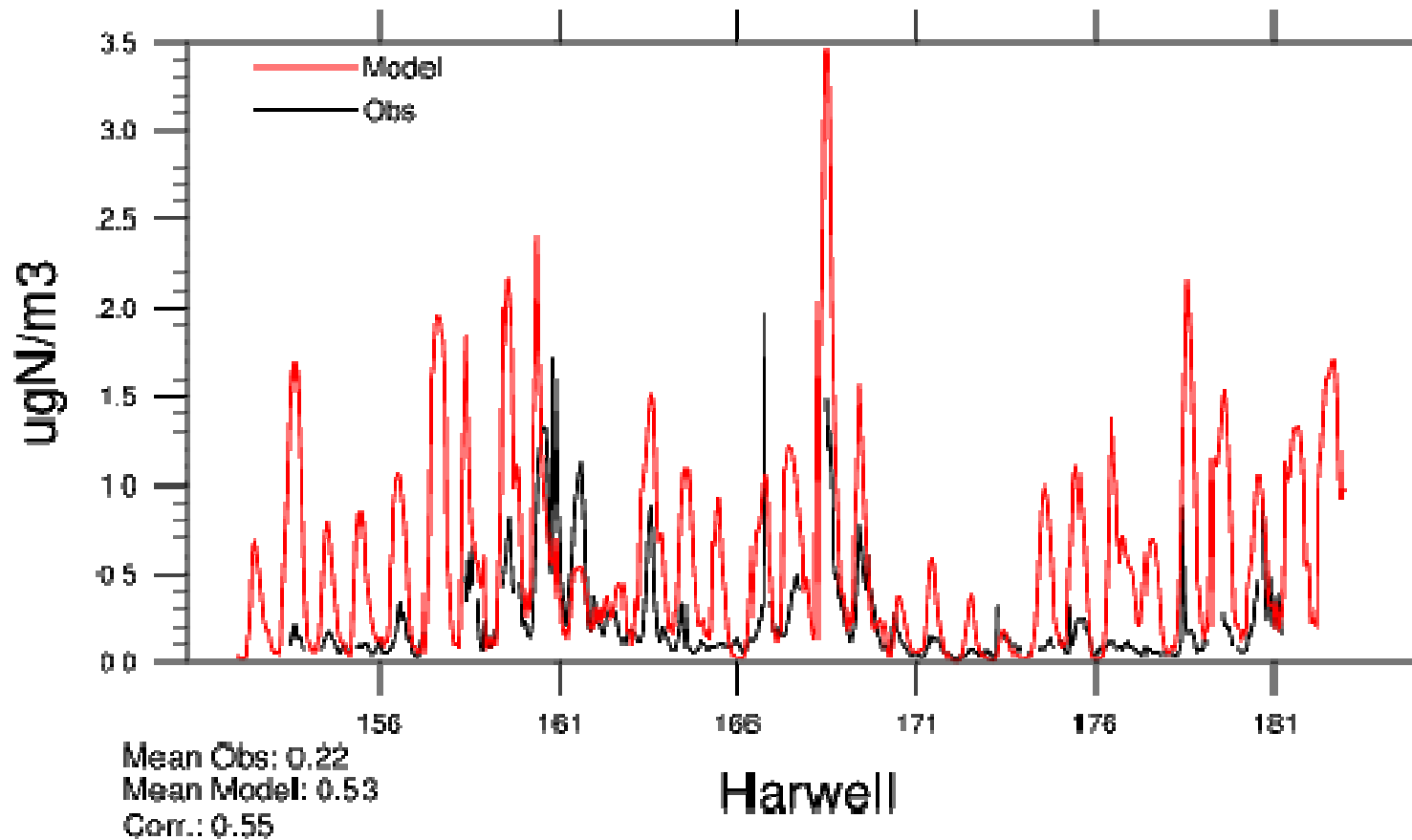


- Summer more under estimated than winter

Evaluation of diurnal variations

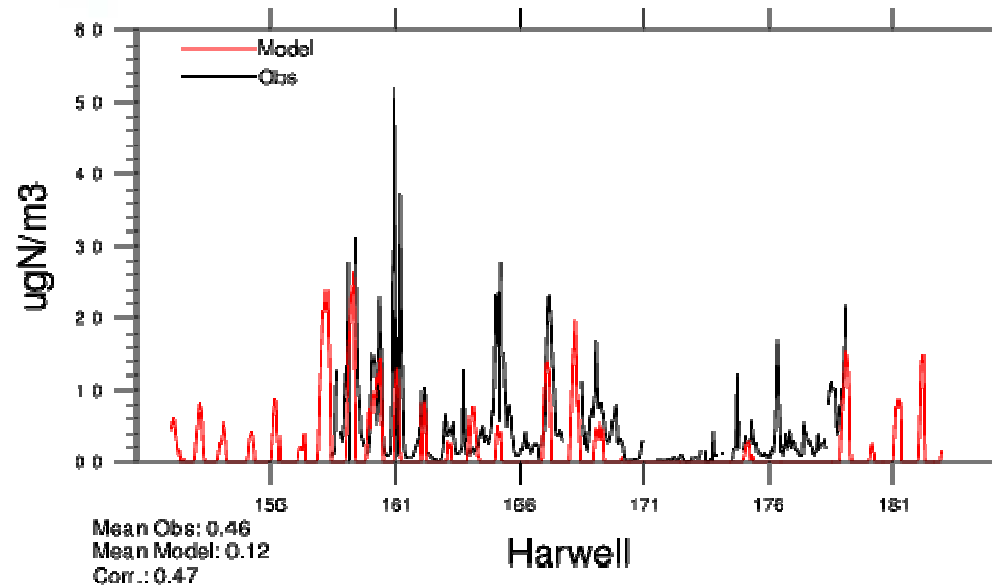


(4-5 sites)
HNO₃ in Air

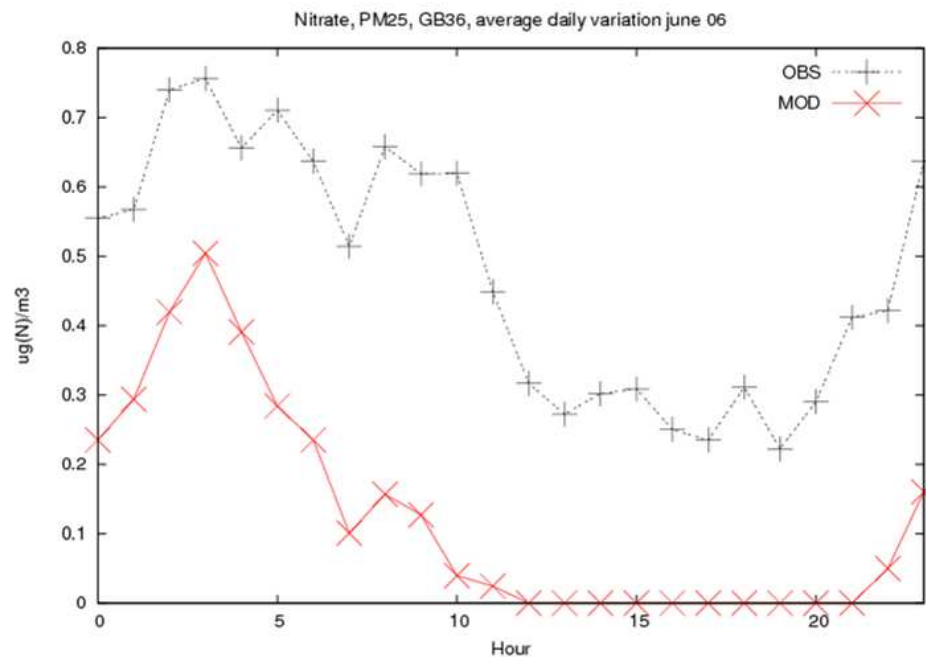


- The EMEP model reproduce the peak around noon

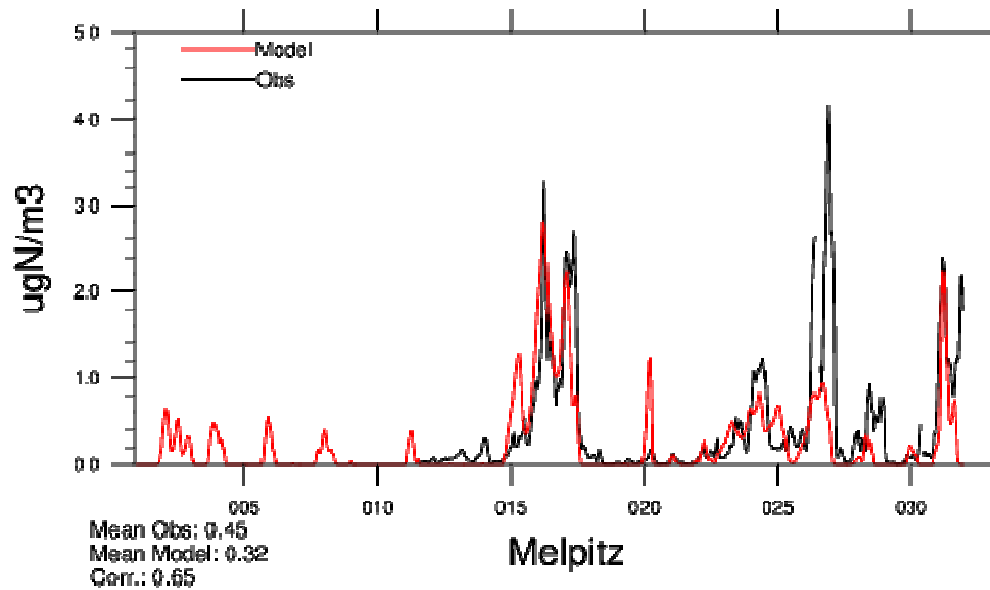
Fine (PM25) nitrate in air



- Fine nitrate, summer:**
- NH_4NO_3 completely evaporates during day
 - Absolute levels low

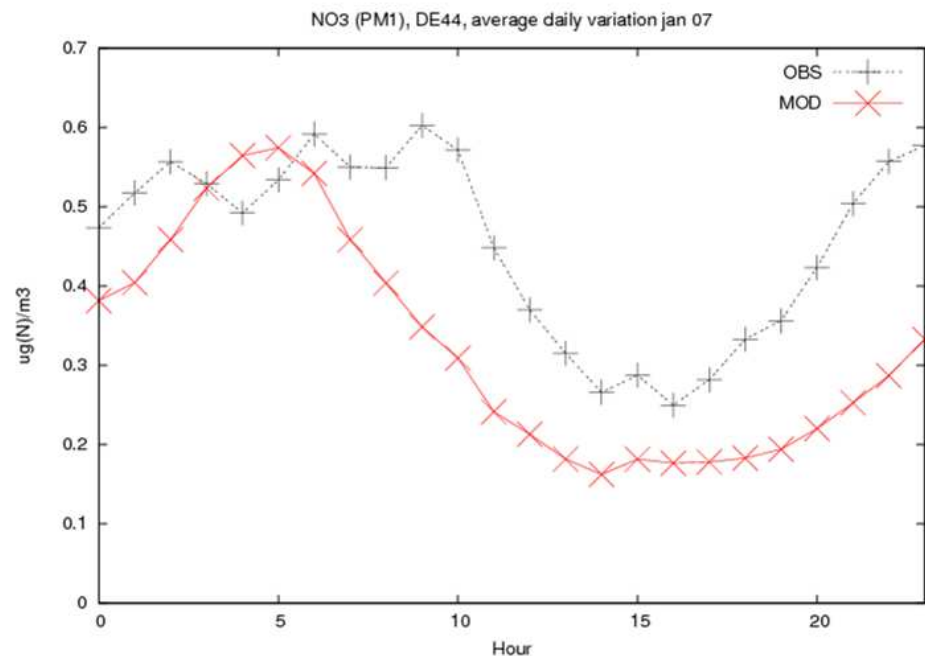


Fine (PM1-obs, PM25-mod) nitrate in air

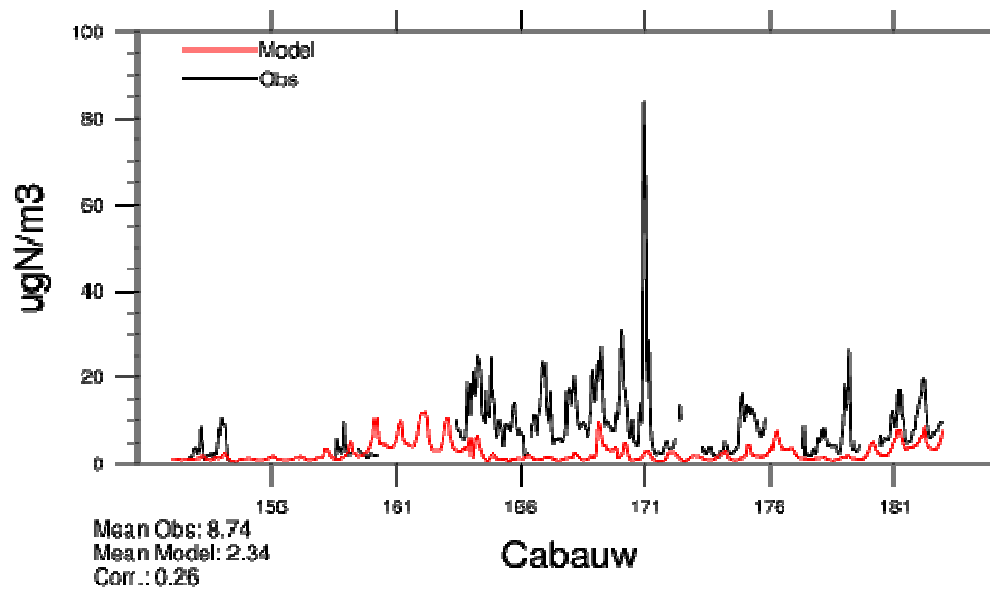


Fine nitrate, winter:

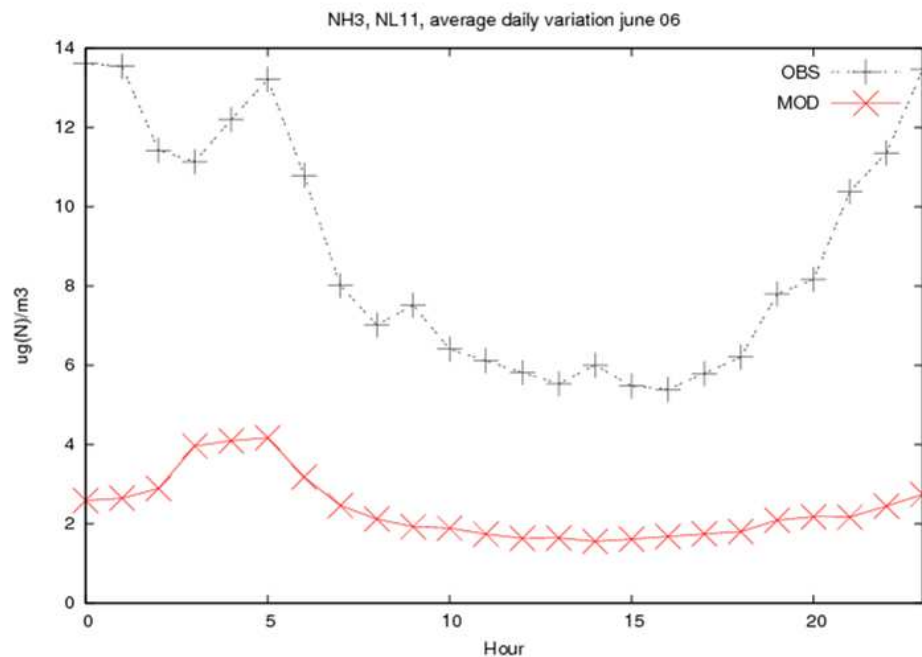
- The EMEP model reproduces morning peak
- Absolute values ok



Ammonia in Air



- Diurnal variation NH₃ determined by:
- Emissions
 - Stability/dry deposition
 - Conversion to ammonium

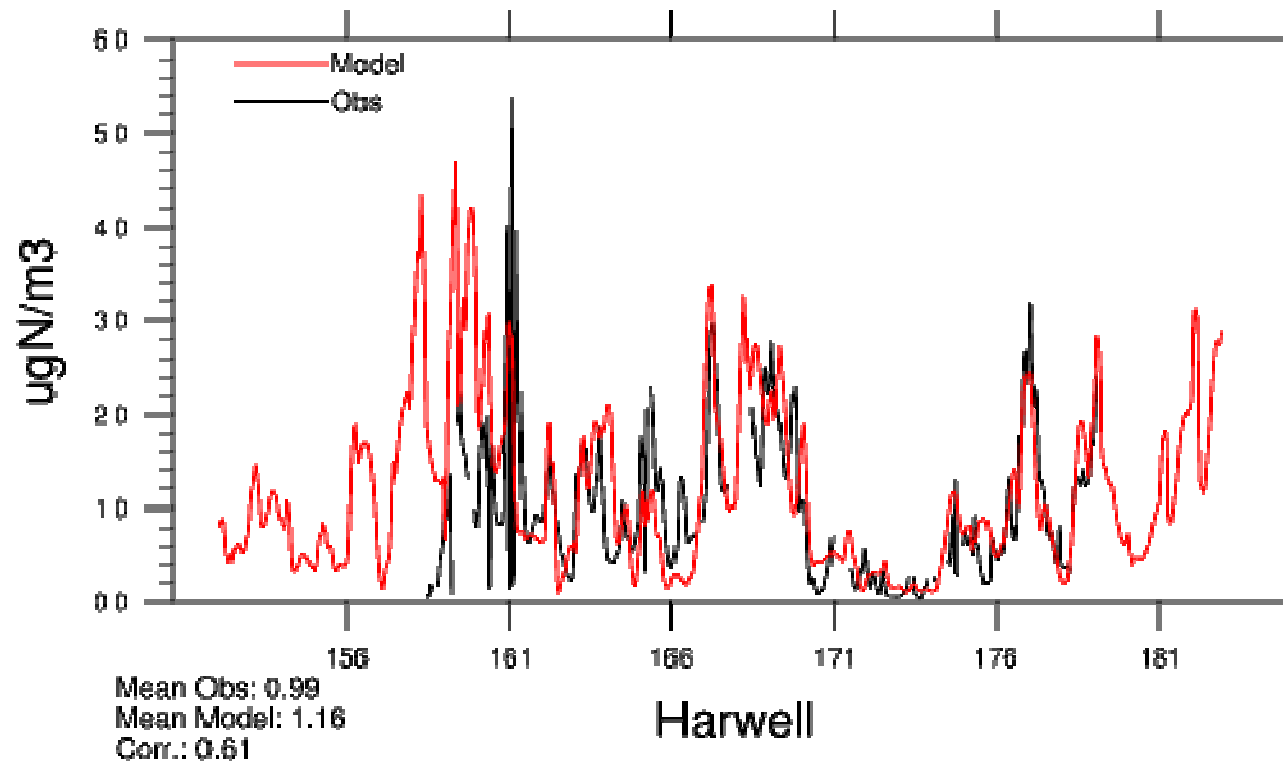


Cabauw June 06: model and obs gives qualitatively same diurnal variation

The EMEP model always gives night time maximum:
-diurnal variation of NH₃ emissions
-need dynamical NH₃emission module



NH₄⁺ (PM₂₅) in Air



- Ammonium modelled well both in summer and winter

Conclusions



- The campaign data have been very valuable for evaluating the (missing) PM mass closure – missing carbonaceous matter..
- Evaluation of N gas-particle: problem in summer formation of fine nitrate
- Diurnal profile of NH_3 in many cases wrong; because of the lack of emission coupling to meteorology?

Model – measurements comparison for PM2.5



			PM_{2.5}	SO₄²⁻	NO₃⁻	NH₄⁺	EC	OC^{*)}	Na⁺	Miner.
IT01	2006	Bias	-45	-53	-92	-39	-76	-94	-79	25
		R	0.71	0.63	-0.03	0.69	0.55	0.56	0.30	0.67
	2007	Bias	-78	-62	57	71	-53	-95	-6	-18
		R	0.30	0.10	0.11	-0.19	0.35	0.59	0.42	0.16
IT04	2006	Bias	-10	-27	-21	-15	-34	-81	-82	
		R	0.75	0.63	0.74	0.59	0.85	0.84	-0.21	
	2007	Bias	-90	-79	-92	-89	-86	-94	-80	0
		R	0.42	0.60	0.55	0.67	0.04	0.25	-0.11	0.24
DE44	2006	Bias	-62	-36	-70	-32	-81	-92	-40	
		R	0.55	0.79	0.03	0.44	0.60	0.54	0.80	
	2007	Bias	-66	-64	-64	-57	-55	-49	-35	
		R	0.35	0.59	0.58	0.58	0.63	0.50	0.65	
NO01	2006	Bias	-70	-55	-100	-51	-22	-86	-33	
		R	0.72	0.88	-0.06	0.48	0.89	0.79	0.51	
	2007	Bias	-48	-36	-	0	20	-69	-41	
		R	0.08	-0.09	-	0.36	0.25	0.14	0.46	

Results for different components



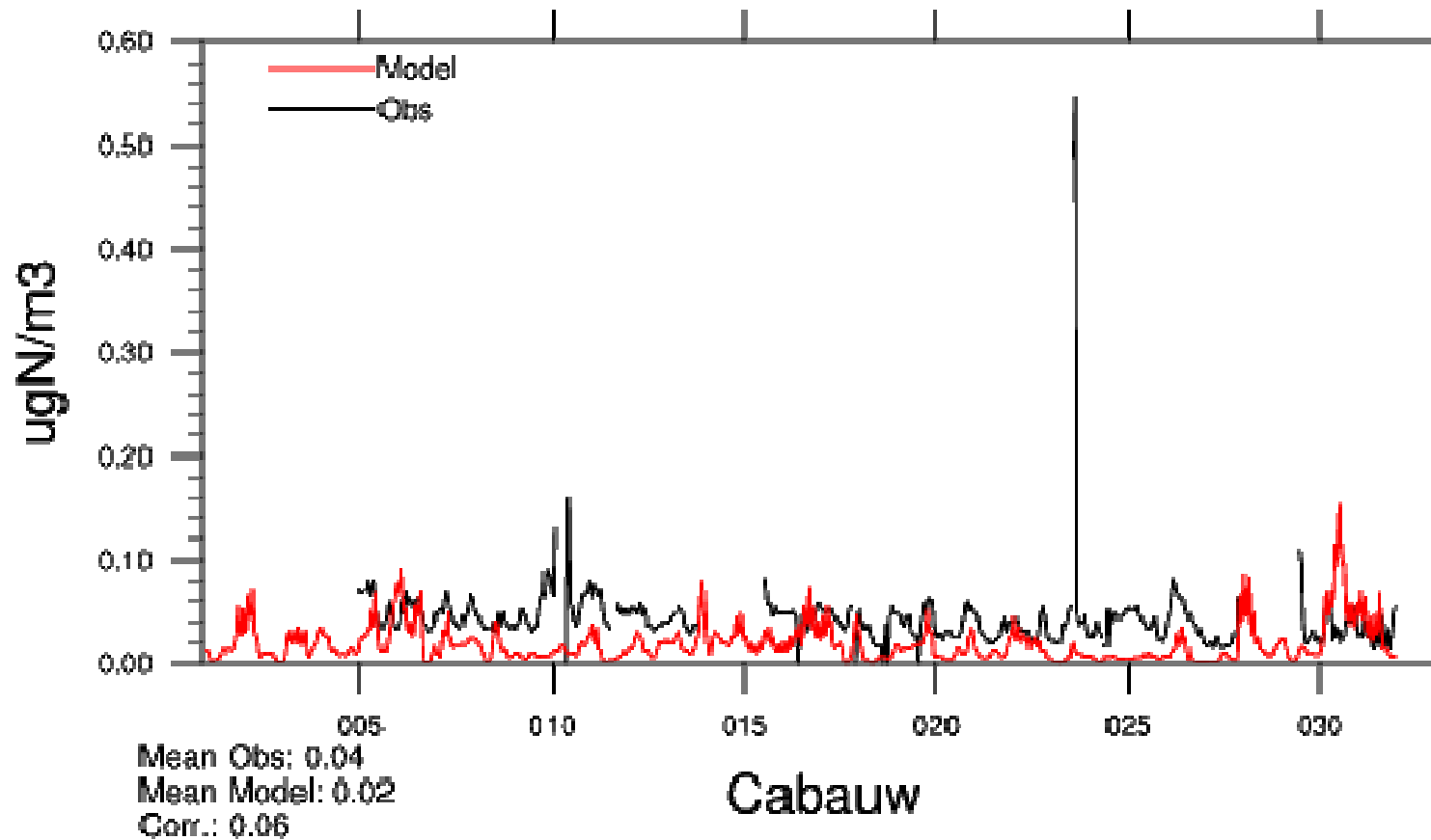
- **Carbonaceous matter: main reason for underestimation of C (especially in June 06), SOA + biogenic missing**
- **Results for EC/OC consistent with comparison against EMEP EC/OC data: underst. EC in central/south (especially summer), overestimate in northern Europe in summer**
- **Na+ underestimated, mostly in summer**
- **Mineral dust, mixed results, mostly within +/-35%**

Model – measurements comparison for PM10



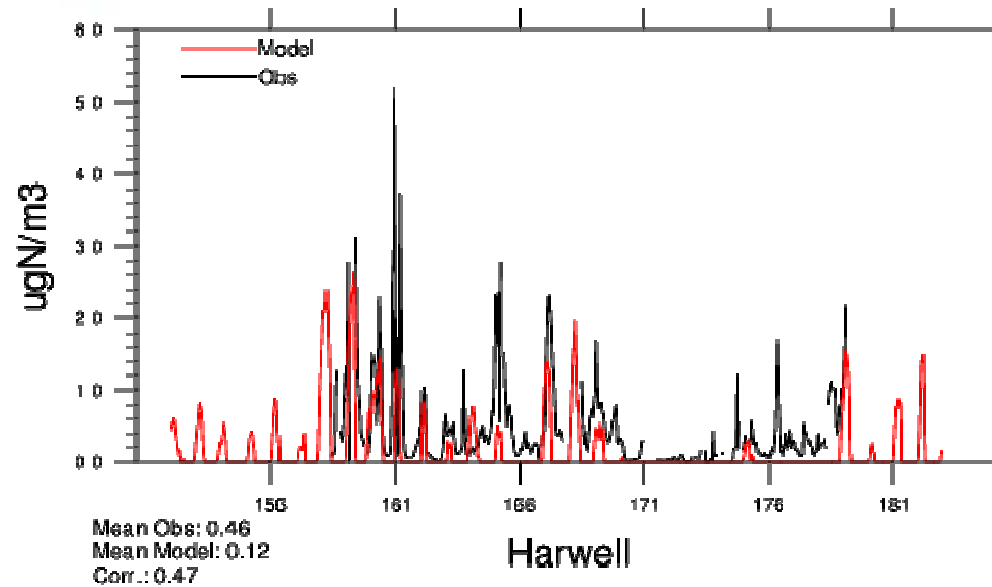
			PM₁₀	SO₄²⁻	NO₃⁻	NH₄⁺	EC	OC[*]	Na⁺	Miner.
IT01	2006	Bias	-58	-51	-67	-32	-78	-94	-82	-34
		R	0.85	0.63	0.33	0.62	0.60	0.49	0.02	0.84
	2007	Bias	-75	-65	-22	57	-59	-94	-27	-57
		R	-0.03	0.10	-0.11	0.10	0.50	0.62	0.53	0.21
DE44	2006	Bias	-66	-40	-49	-32	-88	-92	-40	
		R	0.37	0.77	0.06	0.33	0.73	0.45	0.87	
	2007	Bias	-62	-68	-68	-60	-72	-75	-25	
		R	0.37	0.58	0.62	0.56	0.55	0.48	0.66	
NO01	2006	Bias	-72	-57	-62	-48	-42	-88	-44	
		R	0.78	0.83	0.45	0.76	0.87	0.65	0.79	
	2007	Bias	-26	-42	-50	-33	20	-68	29	
		R	0.01	-0.08	-0.06	-0.05	0.65	0.16	0.31	
ES17	2006	Bias	-10	44	19	269	182	-69	-64	21
		R	0.32	0.72	-0.66	0.92	-0.24	-0.24	0.53	0.54
	2007	Bias	-3	-42	-6	51			-45	-1
		R	0.66	0.77	0.54	0.45			0.57	0.4

HNO₃ in Air

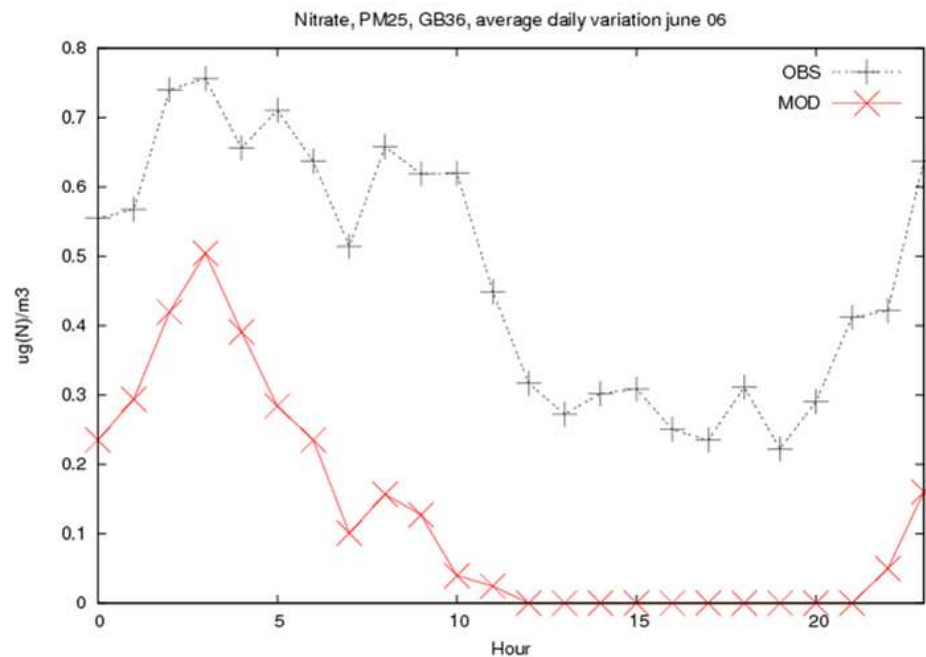


Winter: Much less pronounced diurnal variation. Weaker correlation between obs and model

Fine (PM25) nitrate in air

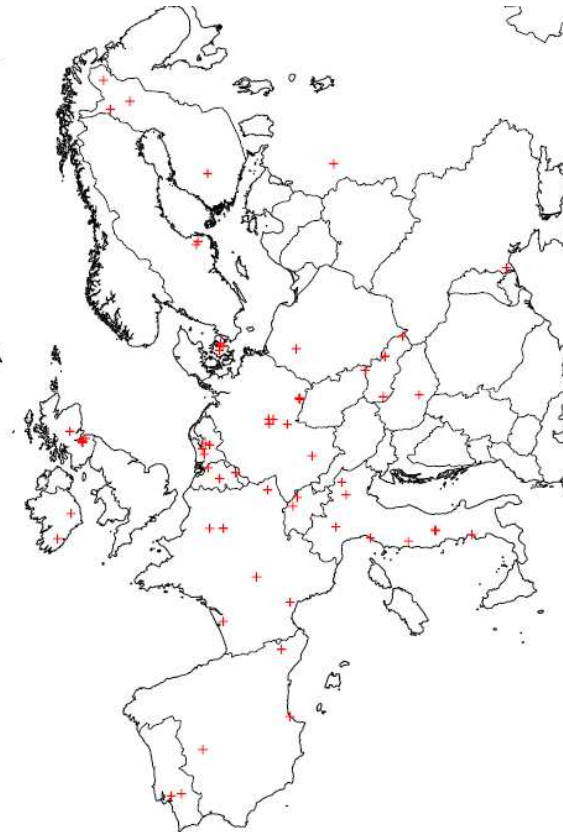


- Fine nitrate, summer:**
- NH_4NO_3 completely evaporates during day
 - Absolute levels low



NitroEurope data versus EMEP data, 2007

Component	Ns	Obs	Bias (%)	Corr
NEU data				
NH ₃ (μg(N) m ⁻³)	58	1.64	-13	0.47
NH ₄ ⁺ (μg(N) m ⁻³)	57	0.91	-26	0.67
HNO ₃ (μg(N) m ⁻³)	57	0.25	-46	0.66
NO ₃ ⁻ (μg(N) m ⁻³)	57	0.48	-27	0.79
EMEP data				
NH ₃ (μg(N) m ⁻³)	16	1.78	-34	0.93
NH ₄ ⁺ (μg(N) m ⁻³)	30	0.78	-29	0.63
HNO ₃ (μg(N) m ⁻³)	13	0.17	-29	0.49
NO ₃ ⁻ (μg(N) m ⁻³)	25	0.41	-29	0.73



Stations from NitroEurope

Mass distribution between fine and coarse aerosols



- In observations, a smaller NO₃- mass occurs on fine aerosol in summer (between 10 and 50%) than in winter (between 30 and 80%) - reflected in the model results, but discrepancies can be significant for the individual sites.
- Too little coarse EC in the model results compared to observation, which is due to large uncertainties (underestimation and missing sources) in emission data.
- Na⁺ size distribution is relatively well described by the model, with between 10 and 40% mass in the fine mode (probable measurement problems in January 2007 at NO01).
- For mineral dust, the model tends to underestimate the mass of coarse particles

Analysis is made of



- **Geographical distribution (northern/central/southern Europe)**
- **Temporal variability (summer/winter)**
- **Comparison between model calculated and measured concentrations of the PM components**
- **The distribution of aerosol mass between fine and coarse particles**