Hans van Jaarsveld & Dmitriy Klimov

Sea salt contribution to PM10 levels in the Netherlands

Netherlands Environmental Assessment Agency

Background

PM10 limits (directives 1999/30/EC and 96/62/EC):

- 40 ug/m3 annual average
- 50 ug/m3 daily average not exceeded 35 days / year

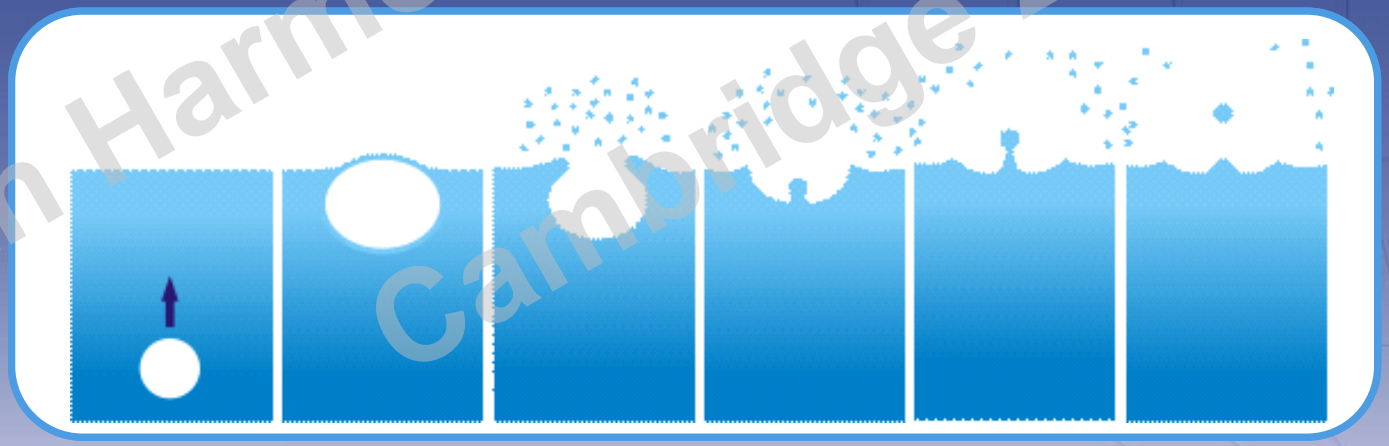
The EC thematic strategy on air pollution provides a possibility to discount contributions of natural substances for compliance purposes if determined with sufficient certainty

This study tries to quantify the contribution of sea-generated particles to annual and daily average PM10 levels in the Netherlands

Sea salt particle generation

- Direct mechanisms
  - aerosol production by spumes at wind speeds higher than 7-11 m/s

- Indirect mechanisms (most important)
  - bursting air bubbles produced by breaking waves



Sea salt generation functions

For *Dp* < 1μm according to Mårtensson *et al.* (2003):

$$\frac{dF_0}{d\log D_P} = 3.84\times10^{-6} U_{10}^{3.41}(A_k T_w + B_k)$$

where *Dp* – dry particle diameter (μm),
U10 – wind speed at 10-m height (m/s),
Tw – water temperature (K),
Ak, *Bk* – polynomials of fourth power as a function of *Dp*.

For 1 ≤ *Dp* ≤ 5 μm according to Monahan *et al.* (1986):

$$\frac{dF_0}{dr} = 1.373 U_{10}^{3.41} r^{-3}(1+0.057 r^{1.05})\times10^{1.19e^{-B^2}}$$

where *r* – particle radius at 80% relative humidity (μm),
U_{10} – wind speed at 10-m height (m/s),

$$B = \frac{0.38 - \log r}{0.65}$$

Sea salt generation functions

For D_p > 5 µm according Smith and Harrison (1998)

$$\frac{dF_0}{dr} = \sum_{i=1}^{2} A_i \exp\left\{-f_i\left[\ln\left(\frac{r}{r_{0i}}\right)\right]^2\right\}$$

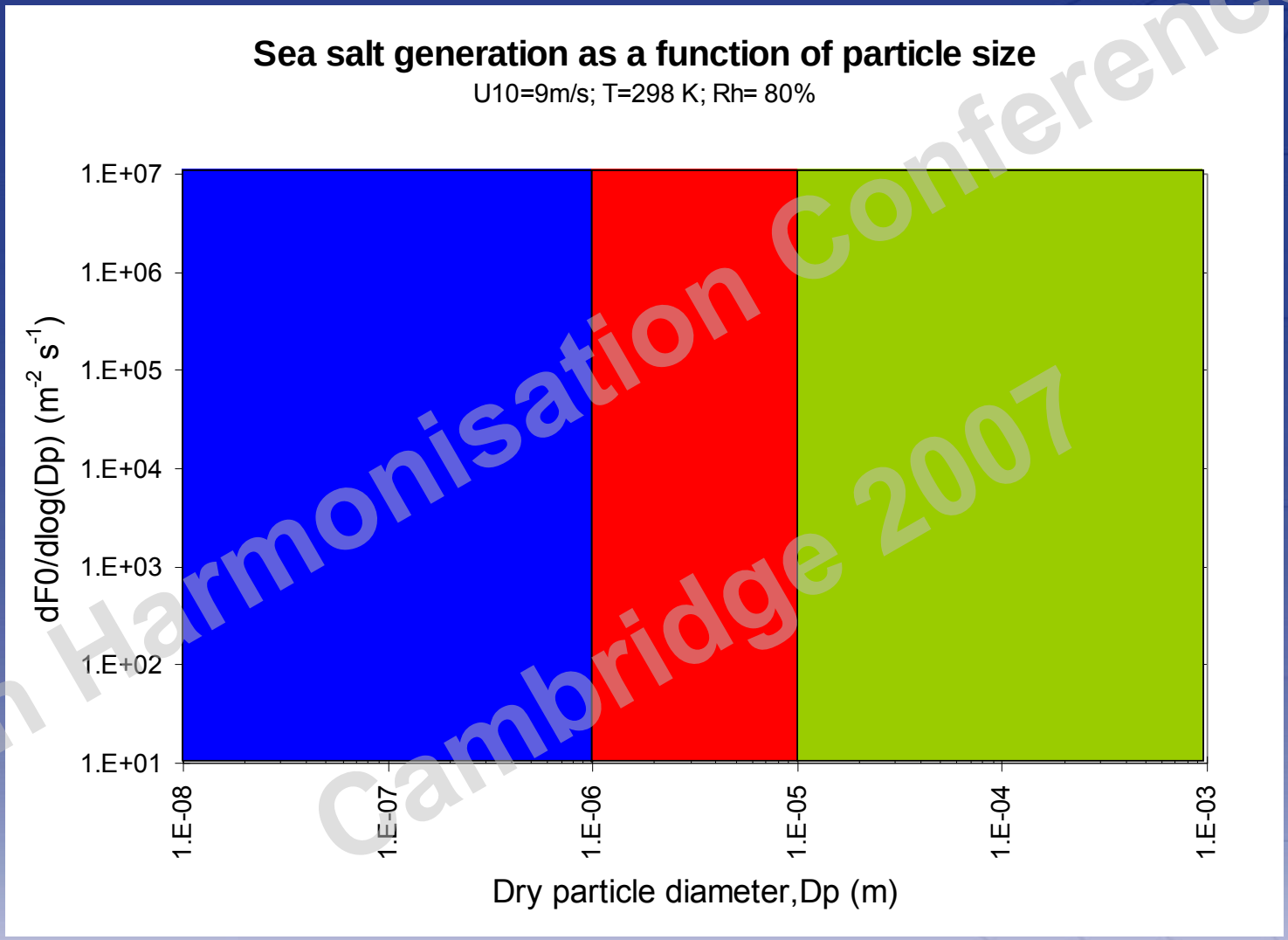
where r = particle radius at 80% relative humidity (µm),

r_{01} = 3 µm and r_{02} = 30 µm,

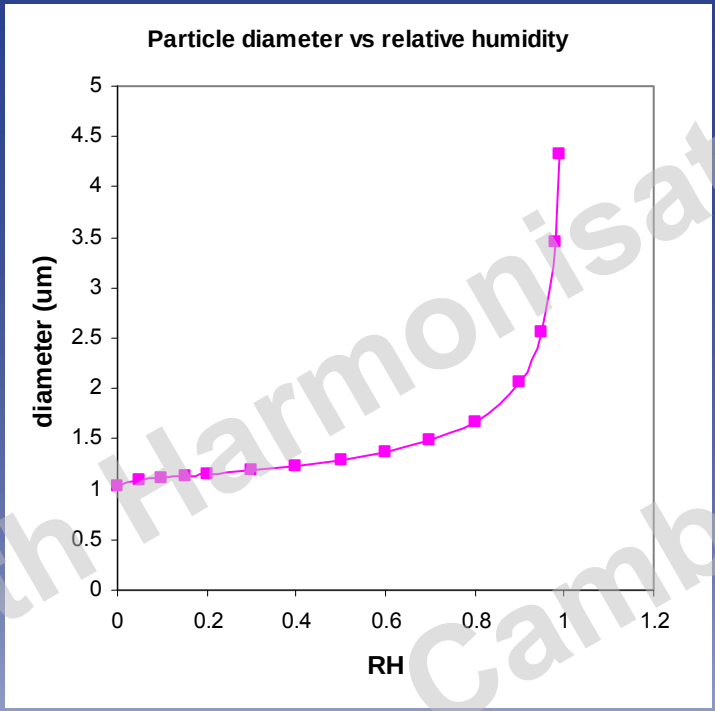
f_1 = 1.5 and f_2 = 1.0,

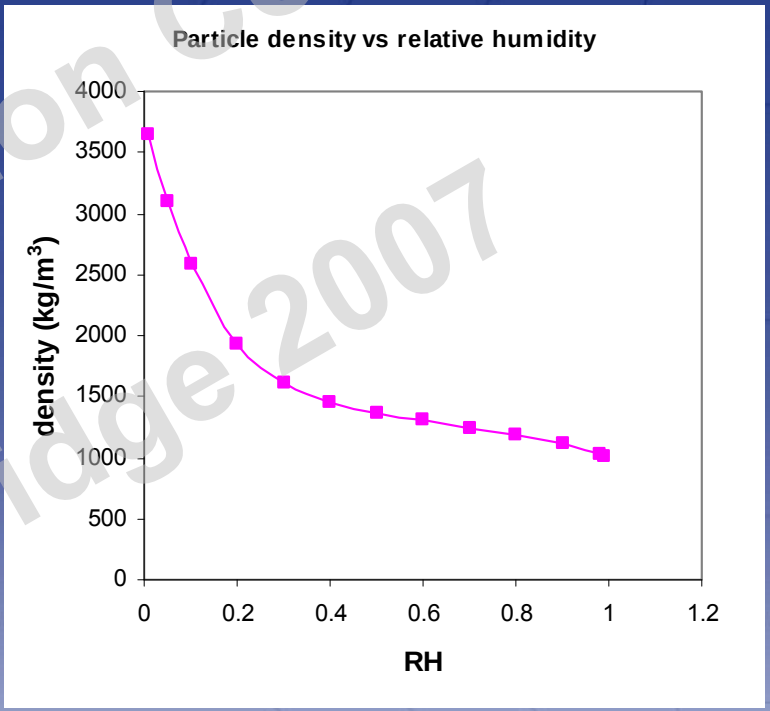
A_1 = 0.2 $U_{10}^{3.5}$ and A_2 = 6.8 × 10^{-3} U_{10}^{3},

U_{10} = wind speed at 10m height (m s^{-1}).



Properties of hygroscopic NaCl particles





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OPS-ShortTerm model

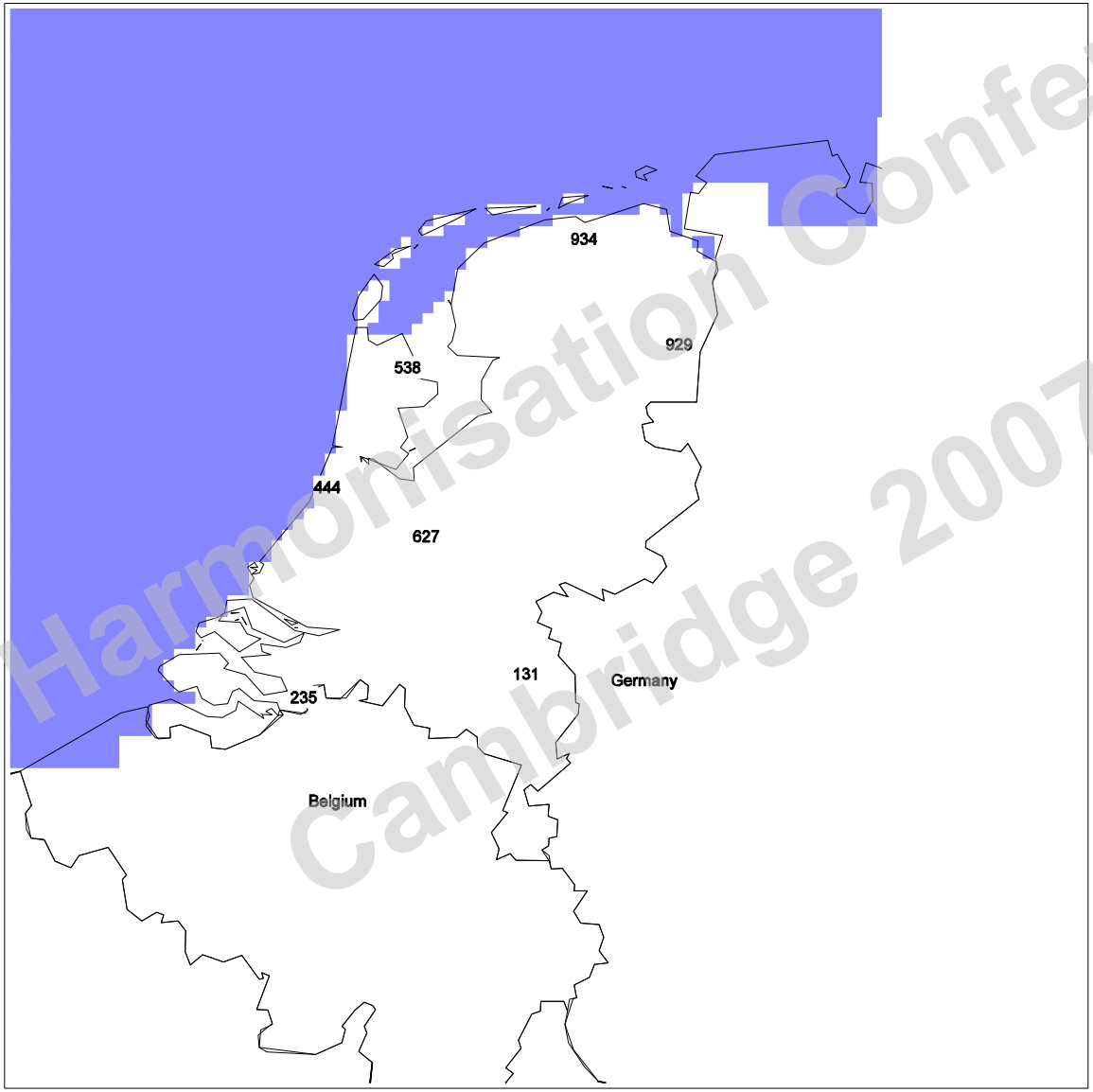
- The model is derived from the OPS long term model.
- The model combines a Gaussian type model for local dispersion and a trajectory model for transport over longer distances.
- Meteorological data from both land- and sea-based stations are used. This short term version is successfully used in the 'VELD' project to describe hourly and daily NH3 concentrations
- The model can handle the decending of plumes due to large particles
- The model uses up to 20 size-bins to describe the particle size distribution
- The model has been tested against Prairie grass, Kincaid and other data sets.

Measurements of sea-salt related compounds used as a reference for model calculations

- Low Volume Sampler (LVS) system measuring CL^- at 7 locations since 1993 on a daily basis (from the Dutch LML network)
  - CL^- is depleted from sea salt by reactions with HNO_3 and H_2SO_4 forming gaseous HCl
  - Unknown cut-off size (probably between 2.5-10 µm)
  - Many observations below detection limit

- Wet deposition measurements of CL^- and Na^+ at 15 locations since 1988 on a 4-weekly basis (LML network)
  - Wet deposition concerns all particle sizes

Stations with Cl- measurements



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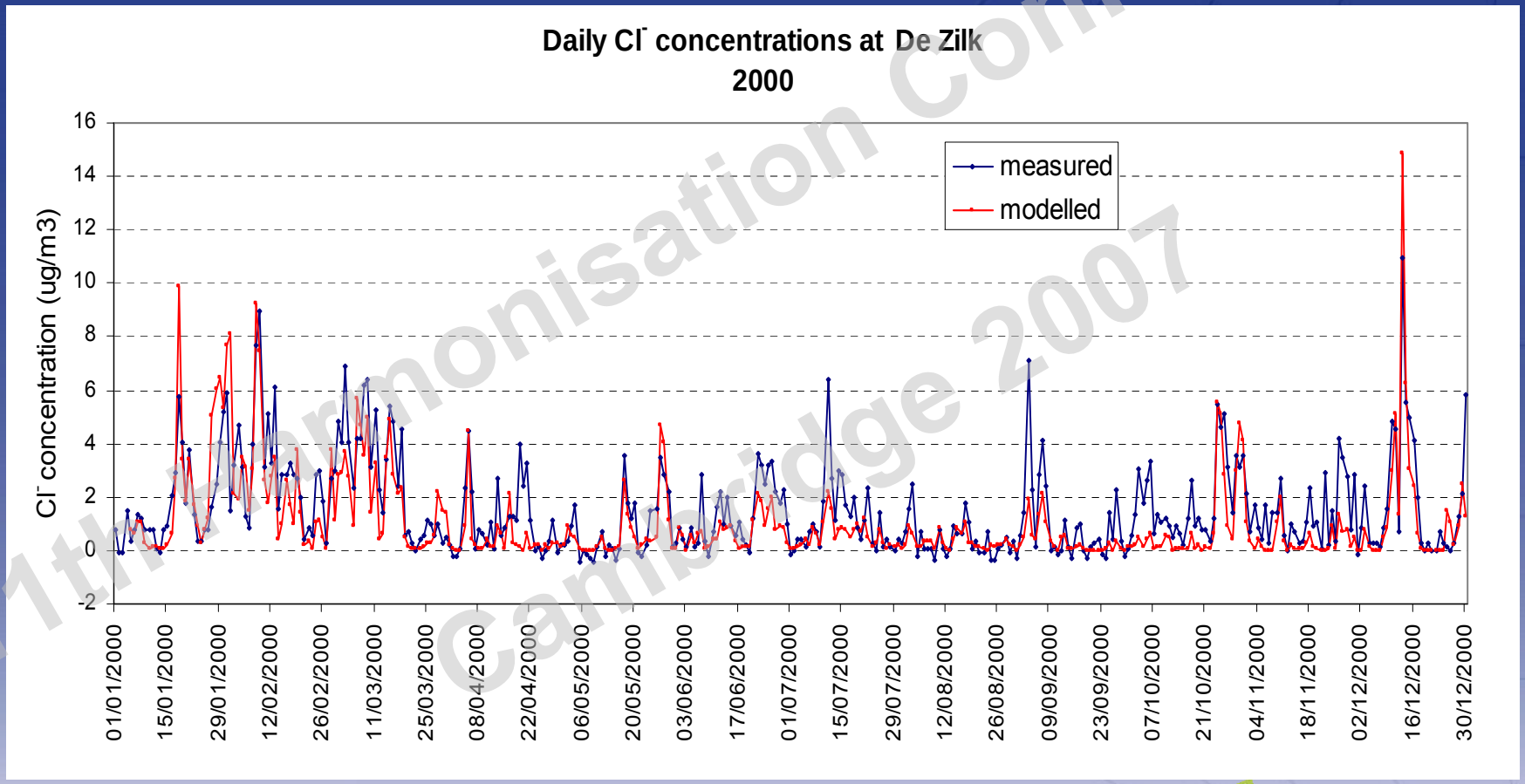
Measured and modelled Cl^- concentrations:
daily averages at a coastal location



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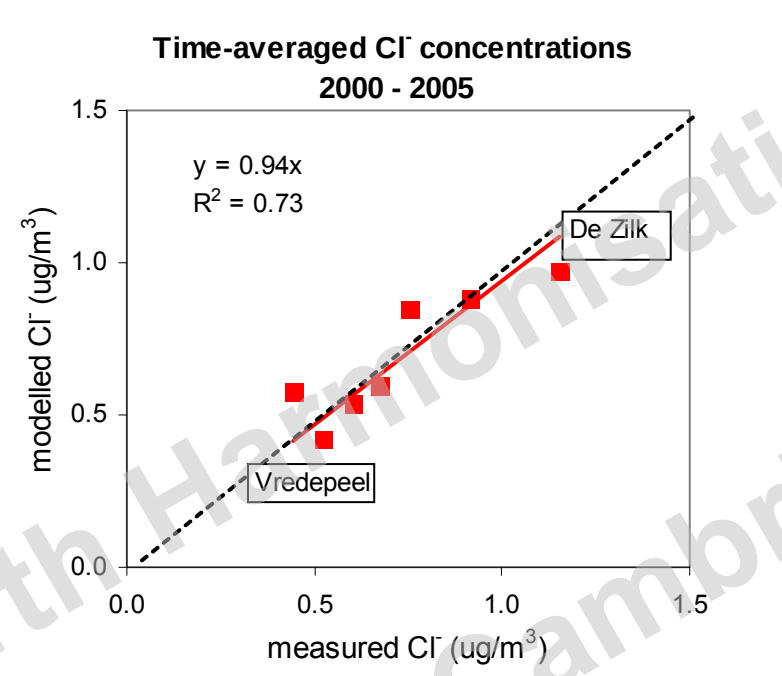
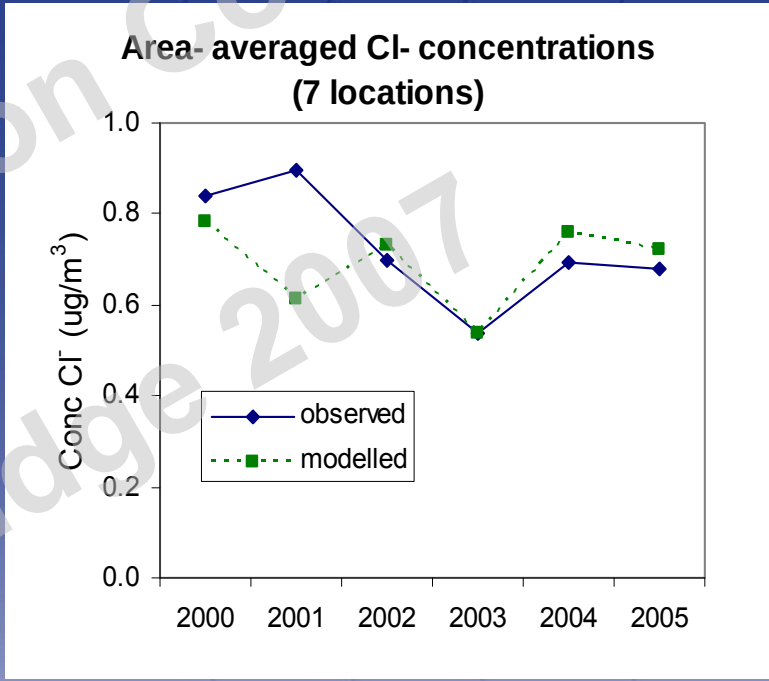
Comparison with measurements

air concentrations of chloride





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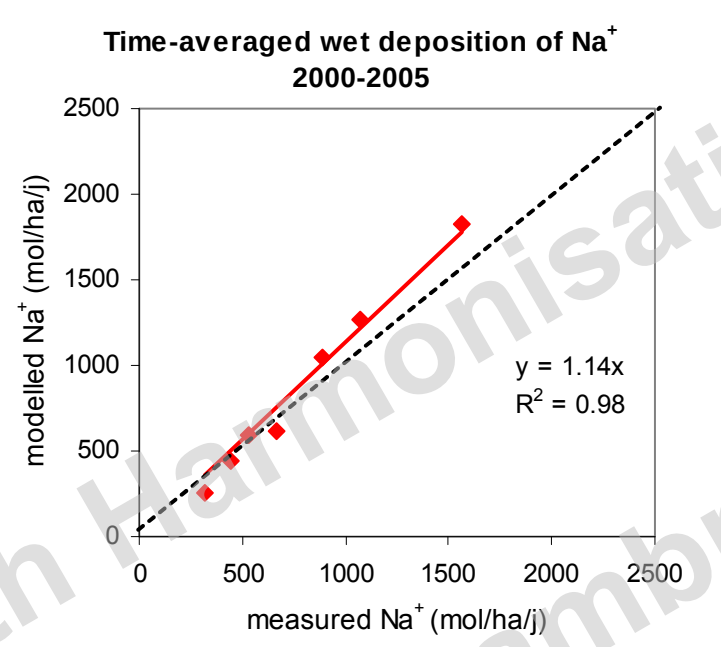
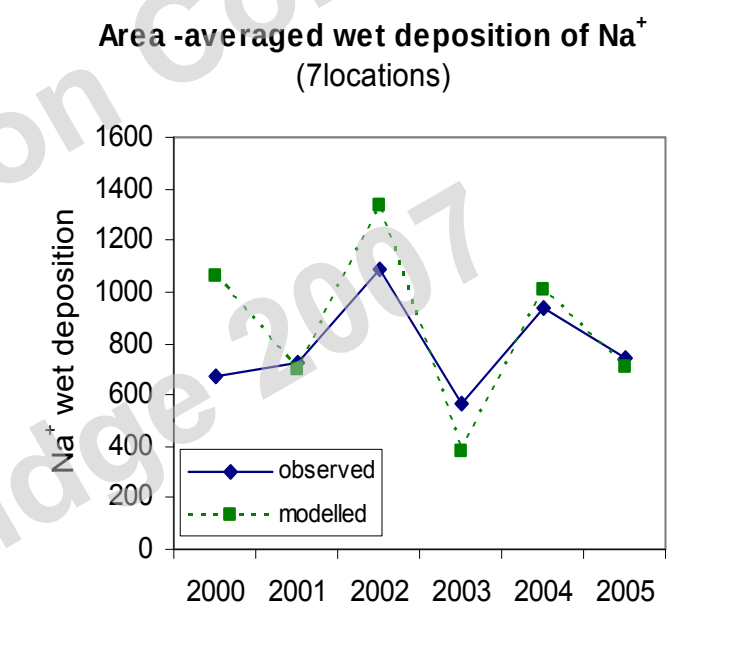
Comparison with measurements

wet deposition of Na^+





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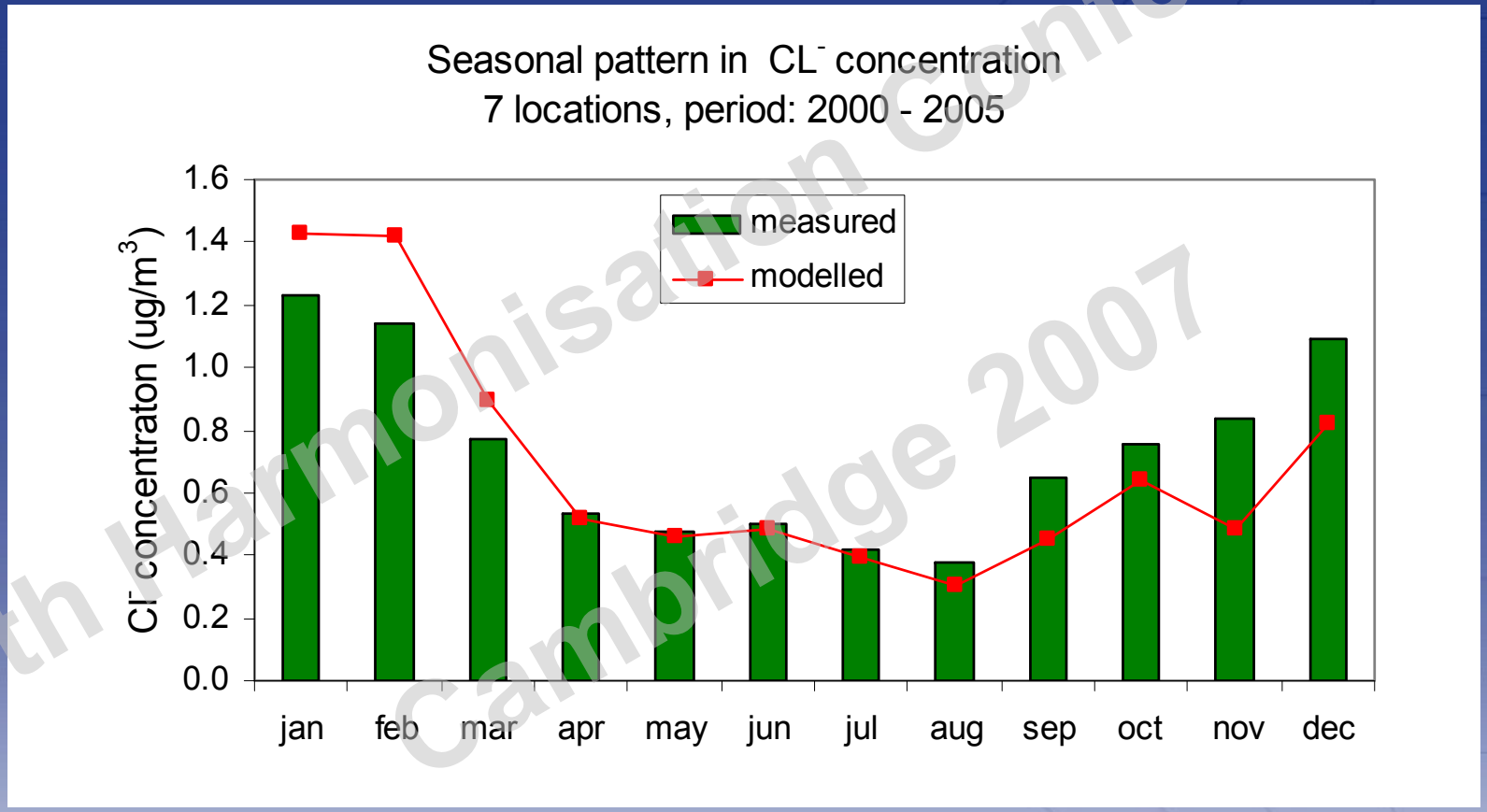
Long term average seasonal pattern of Cl^- aerosol



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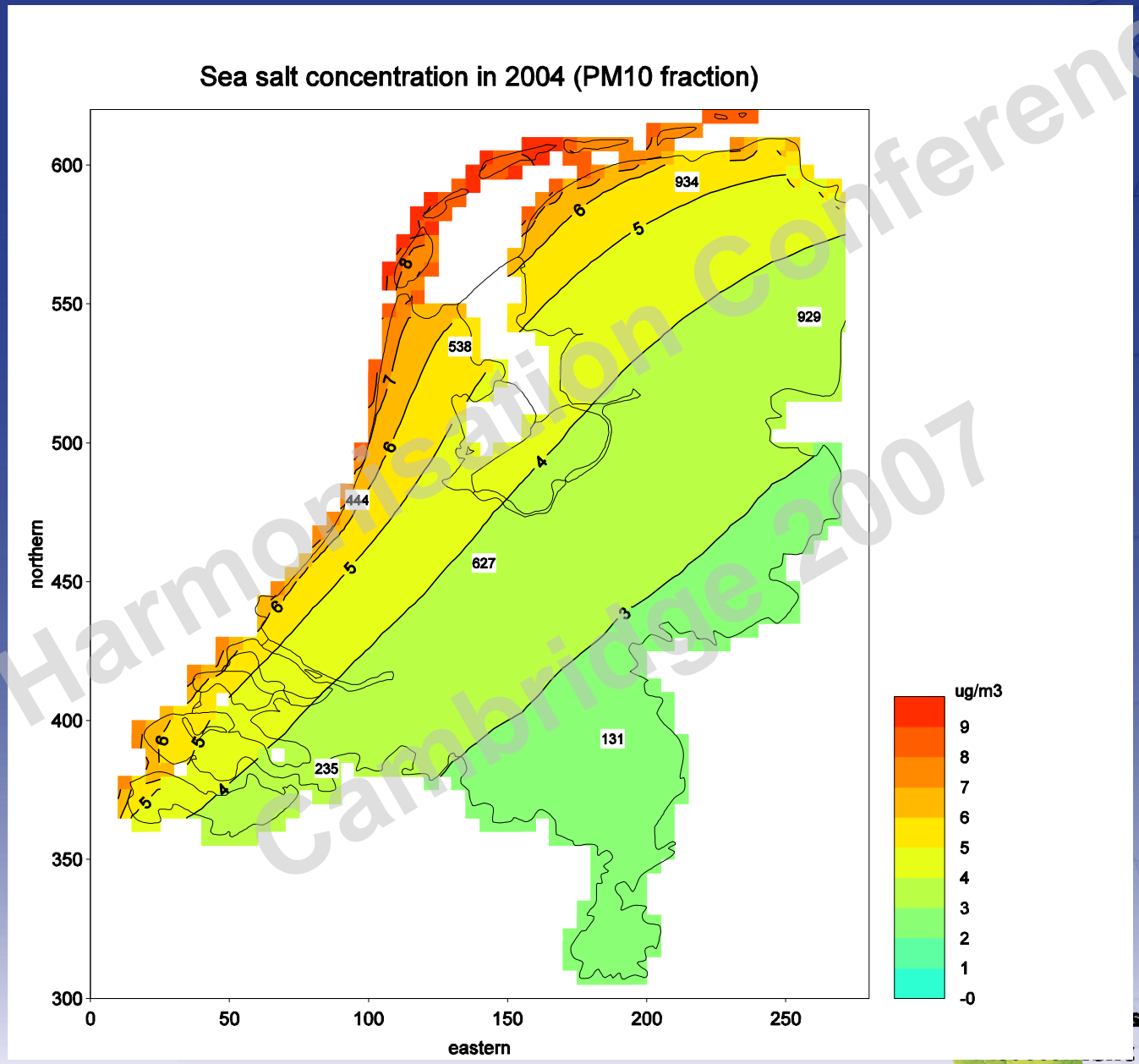

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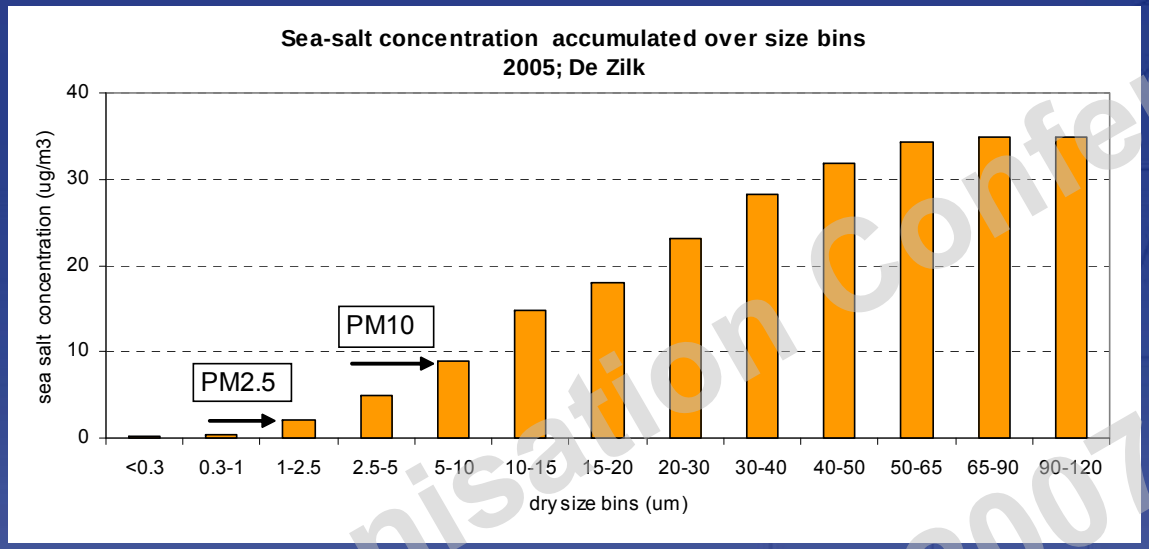
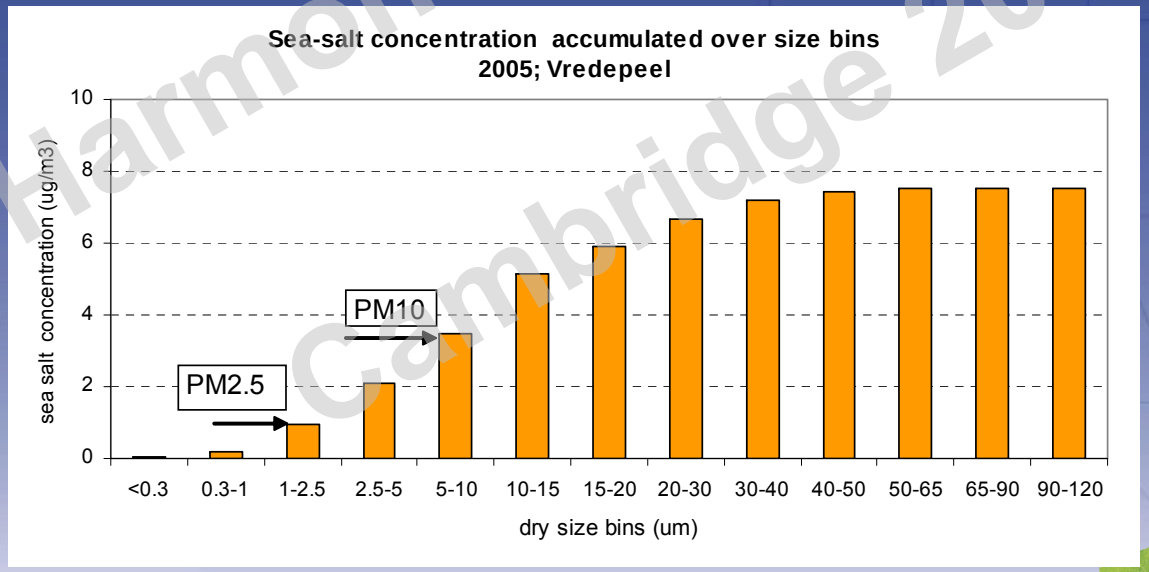
Average sea salt concentrations at 2 locations

	De Zilk (coast)	Vredepeel (inland)
size µm	conc. µg/m3	conc. µg/m3
0-2.5	0.8	0.4
0-10	5.5	2.3
0-120	35.0	7.6





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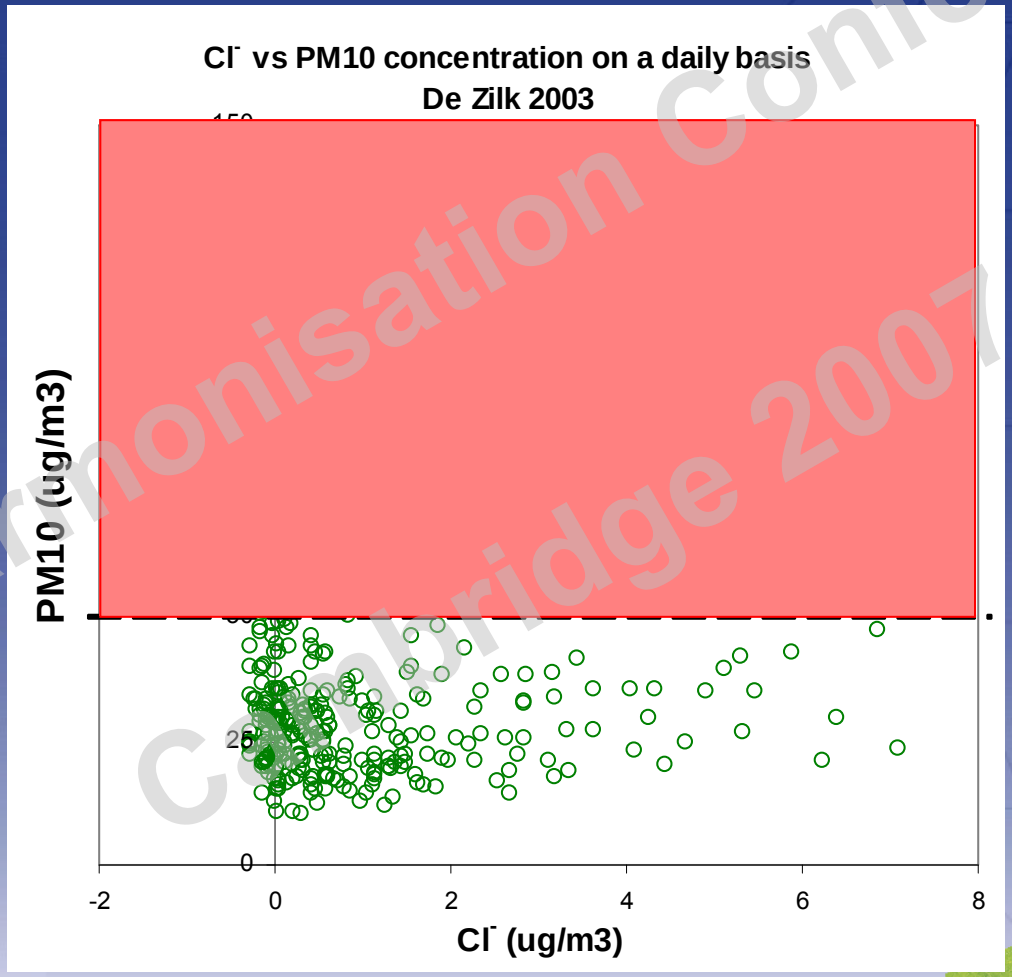
Impact of seasalt on exceeding the 50 ug/m3 daily limit

- Count number of 50 ug/m3 exceedances in measured daily mean PM10 data
- Calculate sea salt (10 um fraction) on a daily basis
- Substract sea salt concentration from PM10
- Count number of exceedances again

Impact of Cl^- on exceedances of 50 ug/m3 daily PM10 limit



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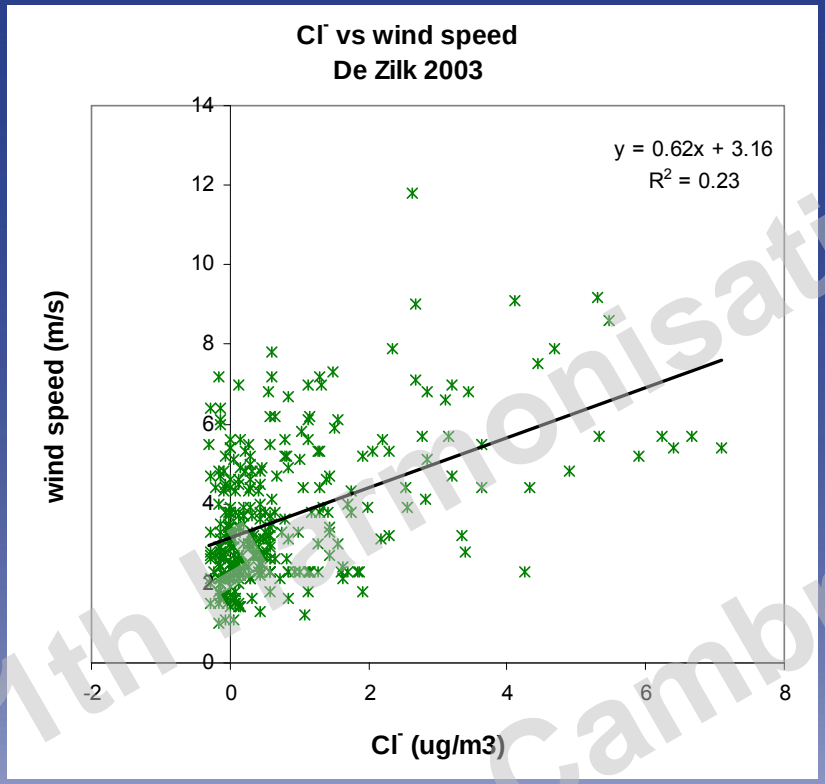
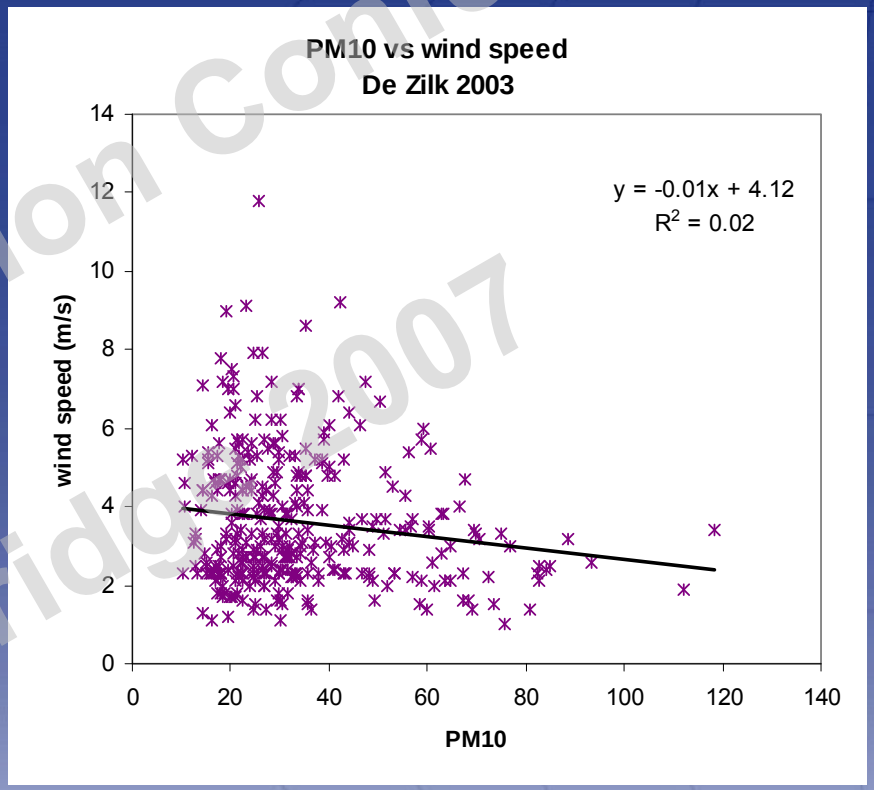
Wind speed dependence of Cl^- and PM10





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Contribution of sea salt to PM10 levels (coastal station)

	Annual average			Number of exceedence days		
	PM_{10} ($\mu g/m^3$) measured	Sea-salt PM_{10} ($\mu g/m^3$) modelled	Contribution to annual average (%)	PM10 measured	Due to sea-salt modelled	Sea-salt contribution (%)
1999	27.2	5.4	19.7	20	3	15
2000	28.4	5.8	20.5	25	5	20
2001	26.3	5.0	18.9	19	0	0.0
2002	30.1	5.5	18.4	31	1	3.2
2003	33.8	4.2	12.5	58	4	6.9
2004	28.6	5.8	20.4	23	0	0.0
2005	25.1	5.5	22.0	11	0	0.0
average	**28.5**	**5.3**	**18.9**	**27**	**1.8**	**6.9**

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Contribution of sea salt to PM10 levels (rural station)

	Annual average			Number of exceedence days		
	PM10 ($\mu g/m^3$) measured	Sea-salt PM10 ($\mu g/m^3$) modelled	Contribution to annual average (%)	PM10 measured	Due to sea-salt modelled	Sea-salt contribution (%)
1999	31.6	2.2	6.8	34	1	2.9
2000	31.9	2.4	7.7	50	2	4.0
2001	32.5	2.0	6.0	39	1	2.6
2002	31.3	2.4	7.5	46	2	4.3
2003	40.9	1.7	4.1	98	4	4.1
2004	25.6	2.4	9.4	25	0	0.0
2005	24.8	2.3	9.4	16	0	0.0
average	**31.2**	**2.2**	**7.3**	**44**	**1.4**	**3.3**

Uncertainties

- Emissions are very sensitive to the windspeed data used
- Reference measurements have a not well defined cut-off size. Modelled values increase 25% if cut-off size is taken 1 µm higher

Conclusions

- Sea salt contributes significantly to average PM10 concentrations in the Netherlands:

  2-6 ug/m^3 on an annual basis (7-20 % of PM10)
- Sea salt contributes hardly to exceedances of the 50 ug/m^3 daily PM10 limit (0-5 days/yr)
- Current approach underestimates CL^- concentrations by 5% and overestimates wet deposition of Na^+ by 15%
- Spatial gradients in air and precipitation are well reproduced
- The ill-defined cut-off size of the Low Volume Sampler system is a major uncertainty in the current approach

11th Harmonisation Conference Cambridge 2007

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Outline
Background
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The model
Validation
Results
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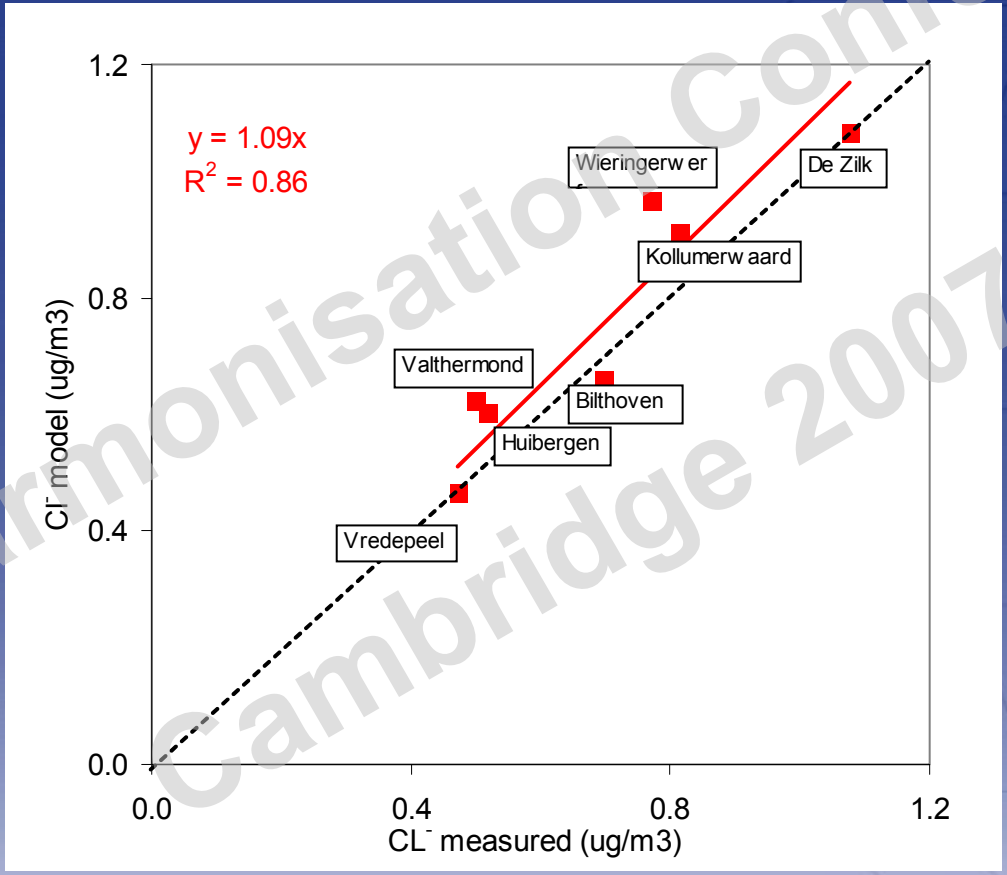
11th Annual Emission Inventory Conference
Cambridge 2007

Measured vs modeled Cl^- concentrations (2004)



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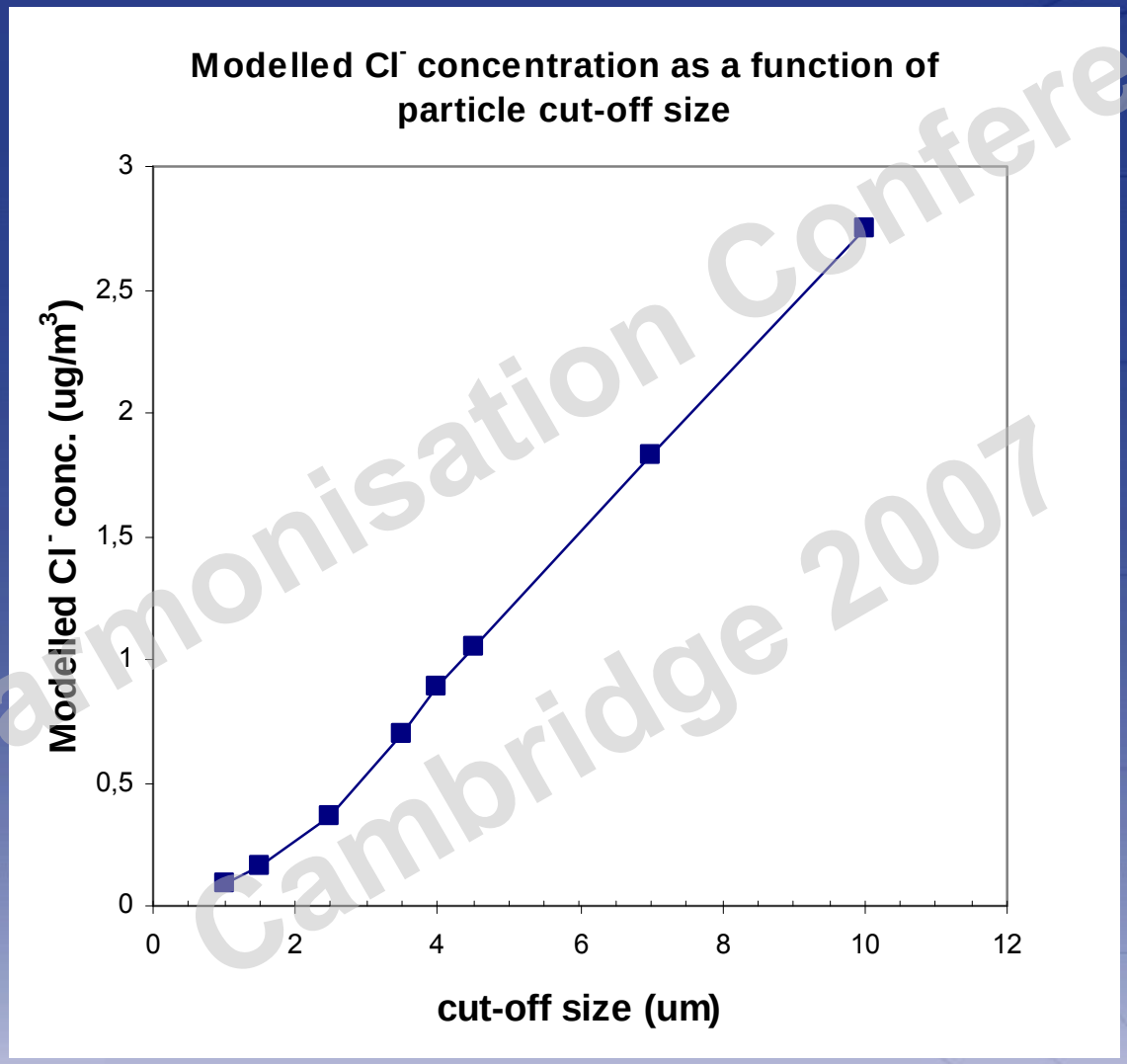

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