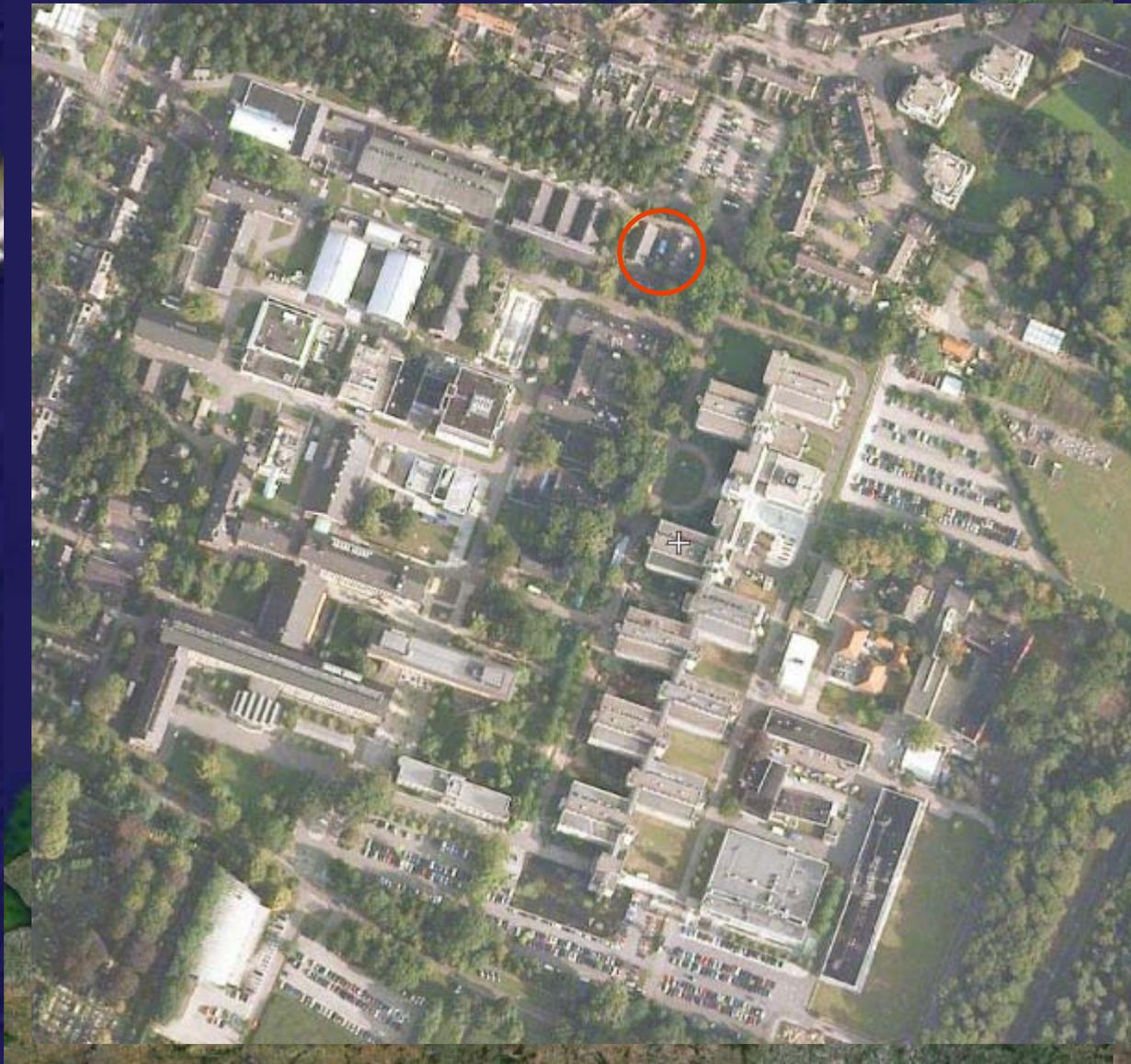




Flemming R. Cassee

## Ambient PM in the Netherlands - Interaction between Science and Policy



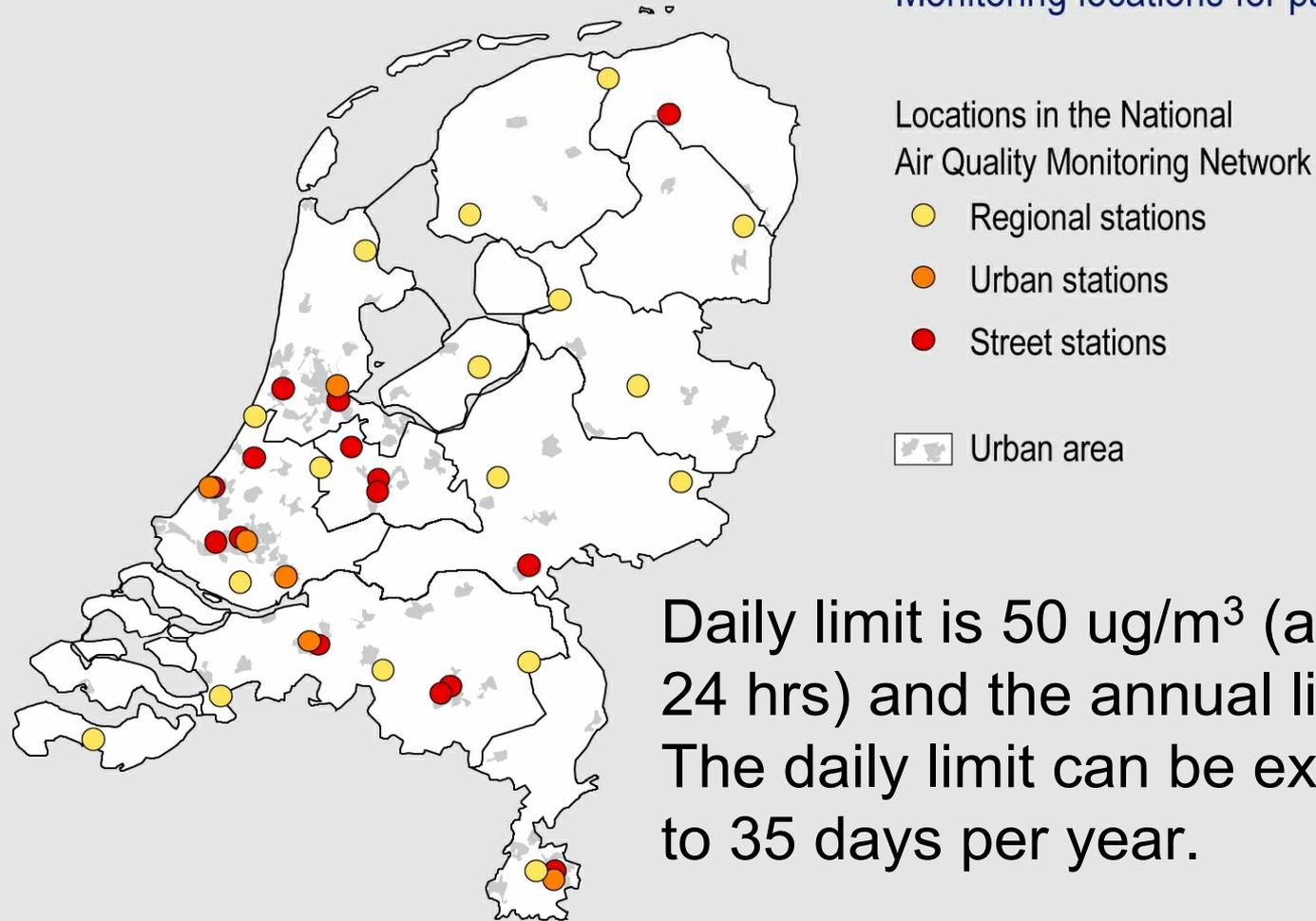


# The state of affairs in the particulate matter dossier from a Dutch perspective

1. What is the problem?
2. Do other countries also have a problem?
3. How high is the emission?
4. How much particulate matter do we measure?
5. How much particulate matter do we calculate?
6. What are the health effects?

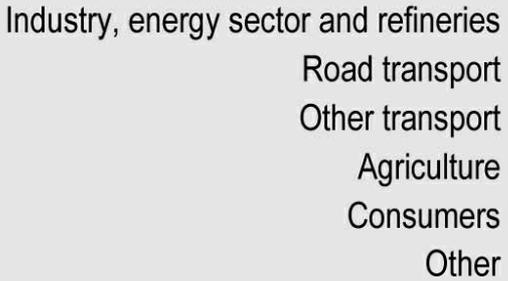
# Monitoring locations for particulate matter (PM10) in the Dutch National Air Quality Monitoring Network

## Monitoring locations for particulate matter

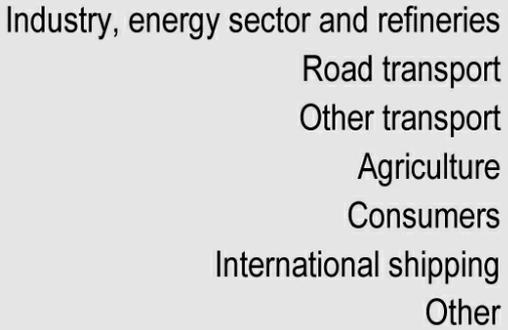


Daily limit is  $50 \text{ ug/m}^3$  (averaged over 24 hrs) and the annual limit  $40 \text{ ug/m}^3$ . The daily limit can be exceeded on up to 35 days per year.

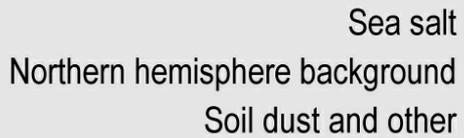
**Sources in the Netherlands (15%)**



**Sources abroad (30%)**



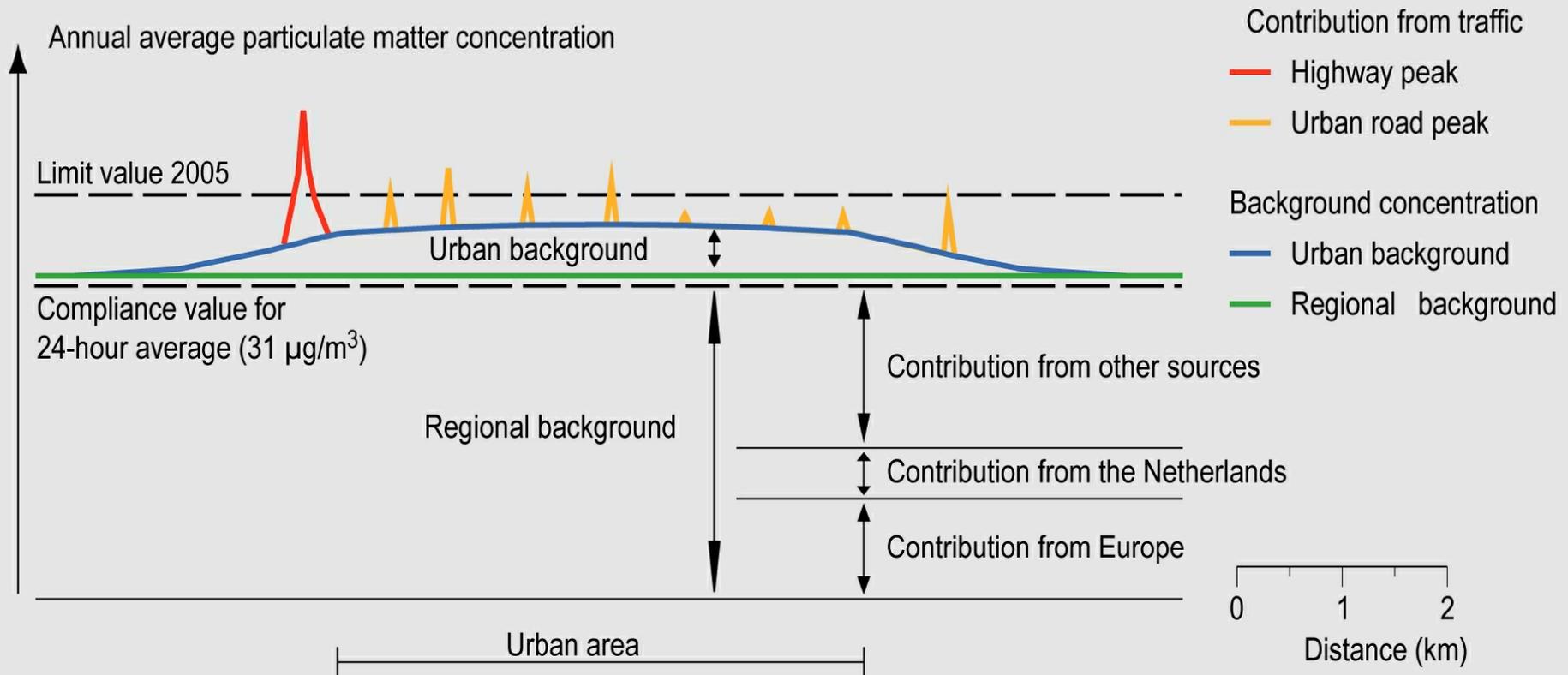
**Other sources (55%)**



0 10 20 30 40  
% of total

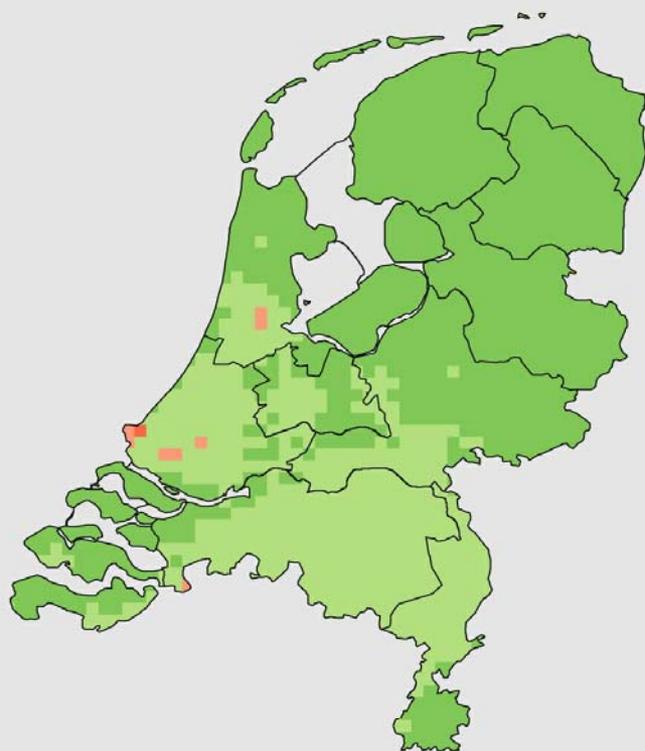
# Composition of particulate matter concentrations in an urban area.

## Example of composition of particulate matter in a cross section of a city

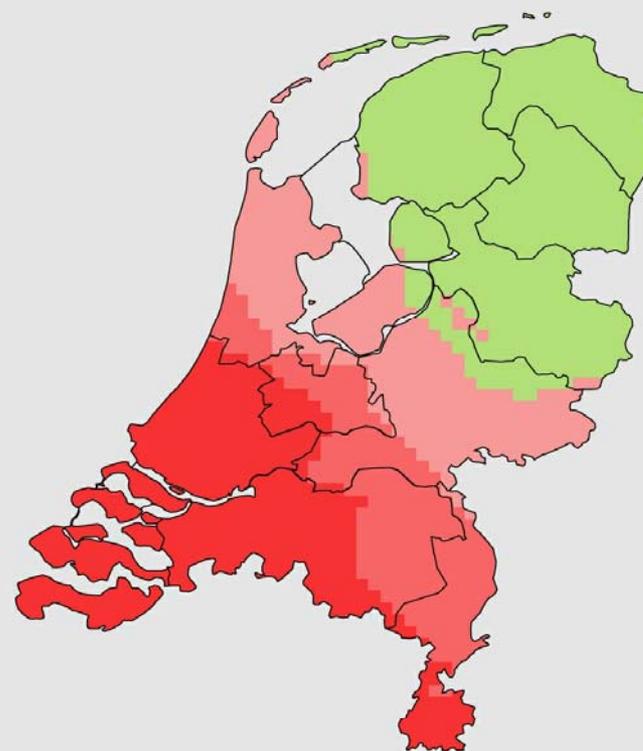
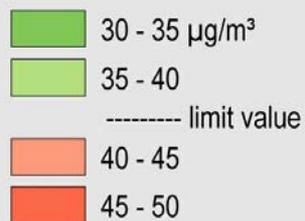


# days with a 24-hour average particulate matter concentration above 50 $\mu\text{g}/\text{m}^3$ (right)

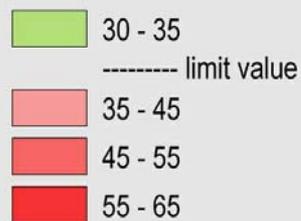
Particulate matter concentration in 2003

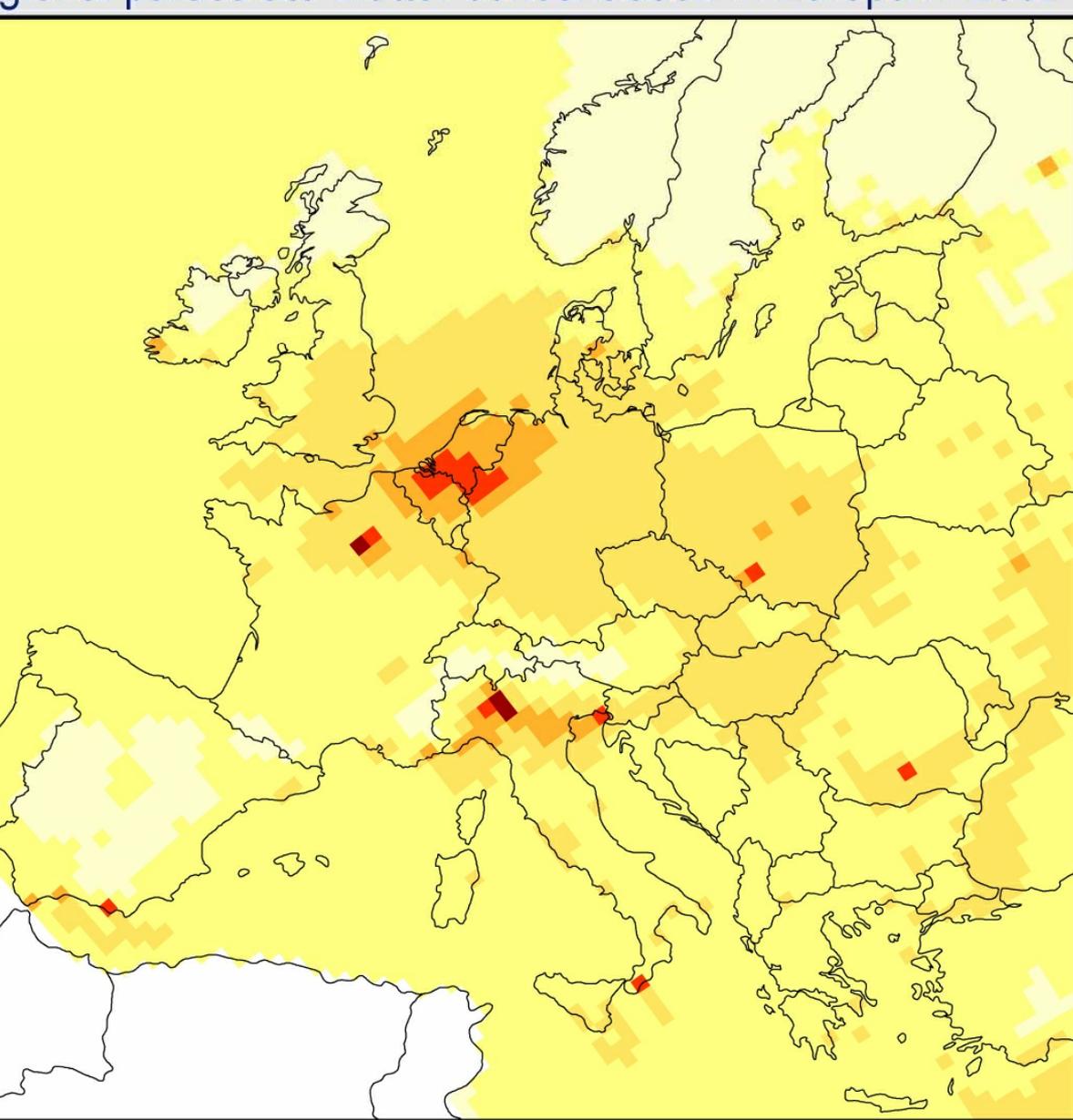


Annual average

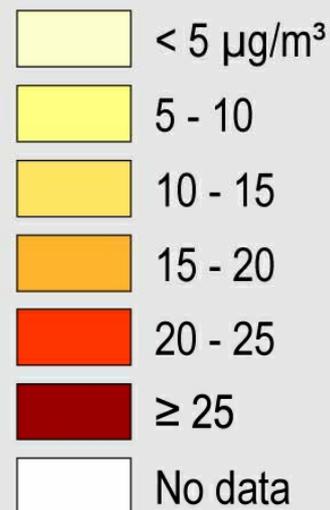


Number of days with 24-hour average above 50  $\mu\text{g}/\text{m}^3$





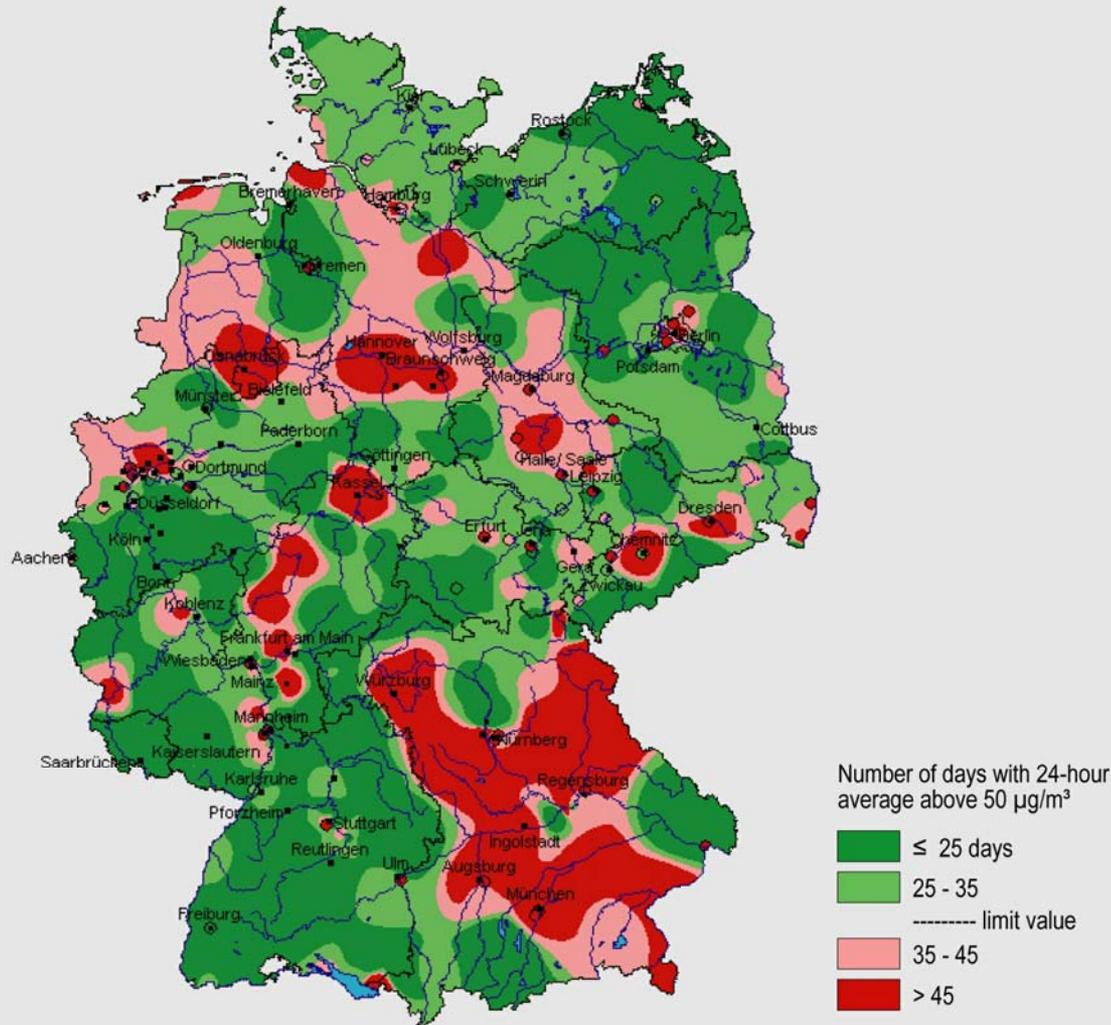
Annual average



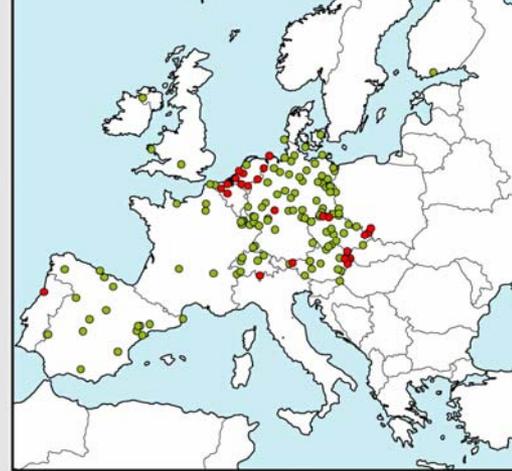
Annual average  
background concentration  
of particulate matter in 200  
modeled at ground level in  
the EU

# Number of days in Germany in 2003 that the 24-hour particulate matter concentration was higher than $50 \mu\text{g}/\text{m}^3$ .

Particulate matter concentration in Germany in 2003

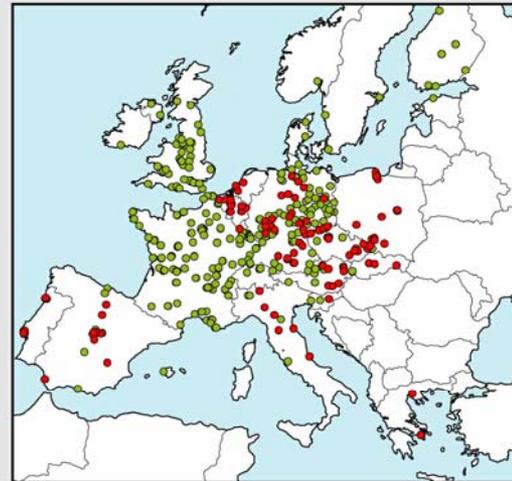


Occurrences where the limit value of the 24-hour average concentration of particulate matter in Europe was exceeded in 2002.

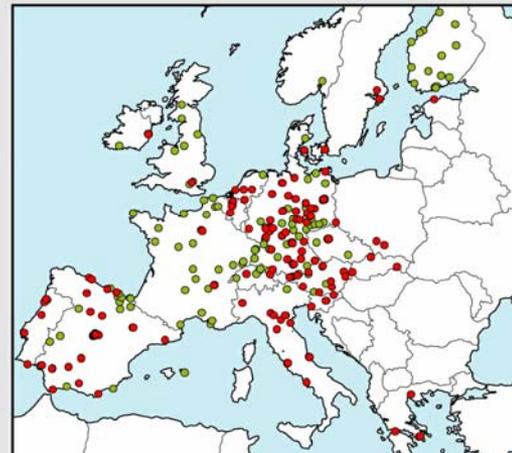


Number of days with 24-hour average above  $50 \mu\text{g}/\text{m}^3$

- > 35
- $\leq 35$



Urban background stations



Street stations

Table 3.1 Emission of primary particulate matter in the Netherlands, 1990-2003 <sup>a</sup>.

Emission per sector	1990	1995	2000	2002	2003	2010
<i>Millions of kg</i>						
Industry, energy sector and refineries	38	23	13	13	12	12
Traffic	23	20	17	16	16	13
Of which road traffic	18	14	12	11	11	
of which diesel vehicles <sup>b</sup>	14	10	8	7	6	
Of which wear <sup>c</sup>	3	3	3	3	3	
Consumers	4	4	4	4	4	9 <sup>d</sup>
Trade, services, government and construction	4	3	4	4	3	
Agriculture	9	10	10	9	8	10
<b>Total PM<sub>10</sub></b>	<b>78</b>	<b>59</b>	<b>49</b>	<b>45</b>	<b>42</b>	<b>44</b>

The emissions from shipping are not included in this table. In 2000 these amounted to 2 million kg for emissions in ports and 8 million kg for emissions on the continental portion of the Netherlands. (Emission Inventory, 2005).

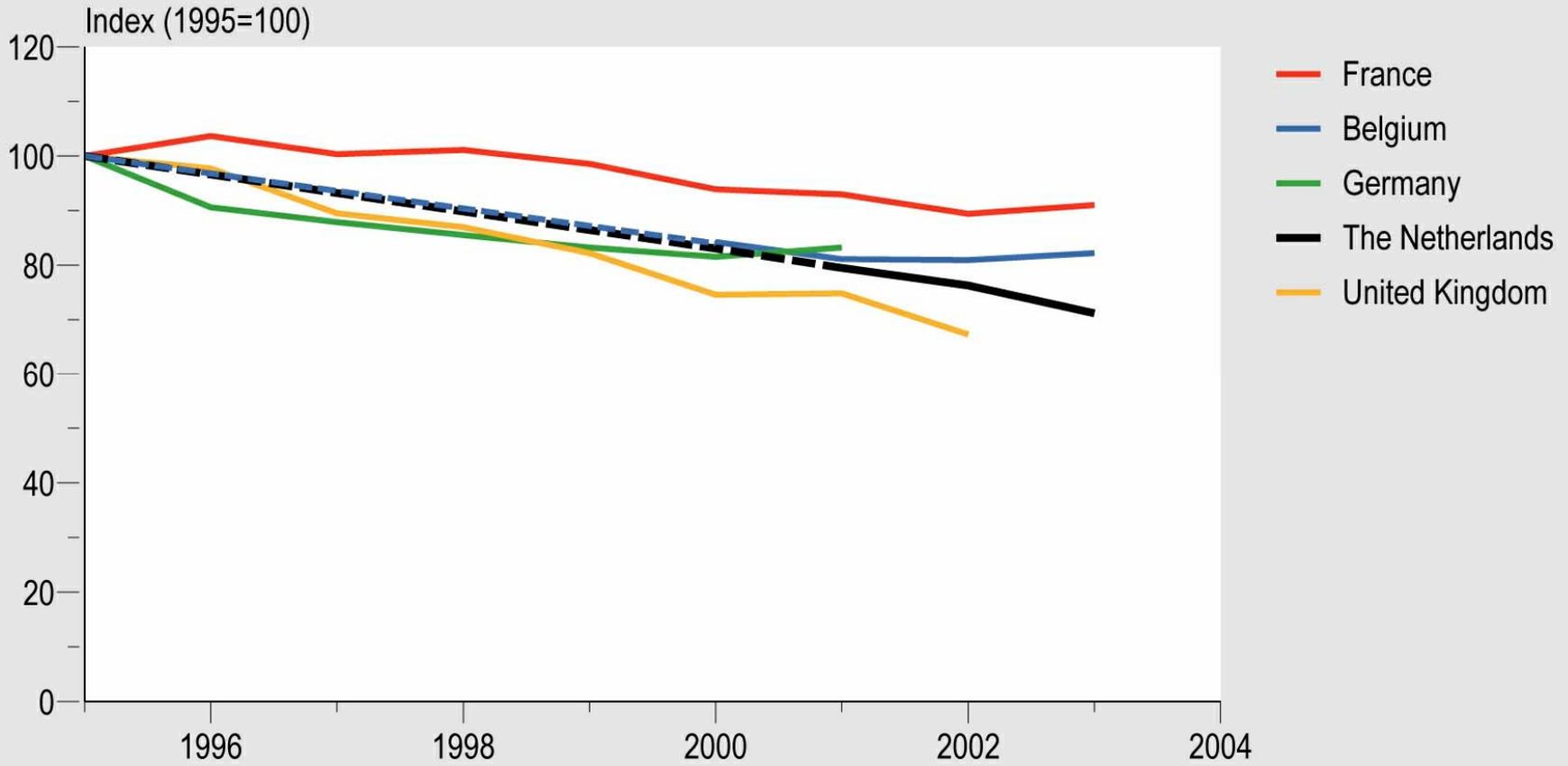
<sup>b</sup> 30% originates from automobiles, 70% from trucks (including delivery vans and busses).

<sup>c</sup> Wear from tyres, road surfaces and brakes.

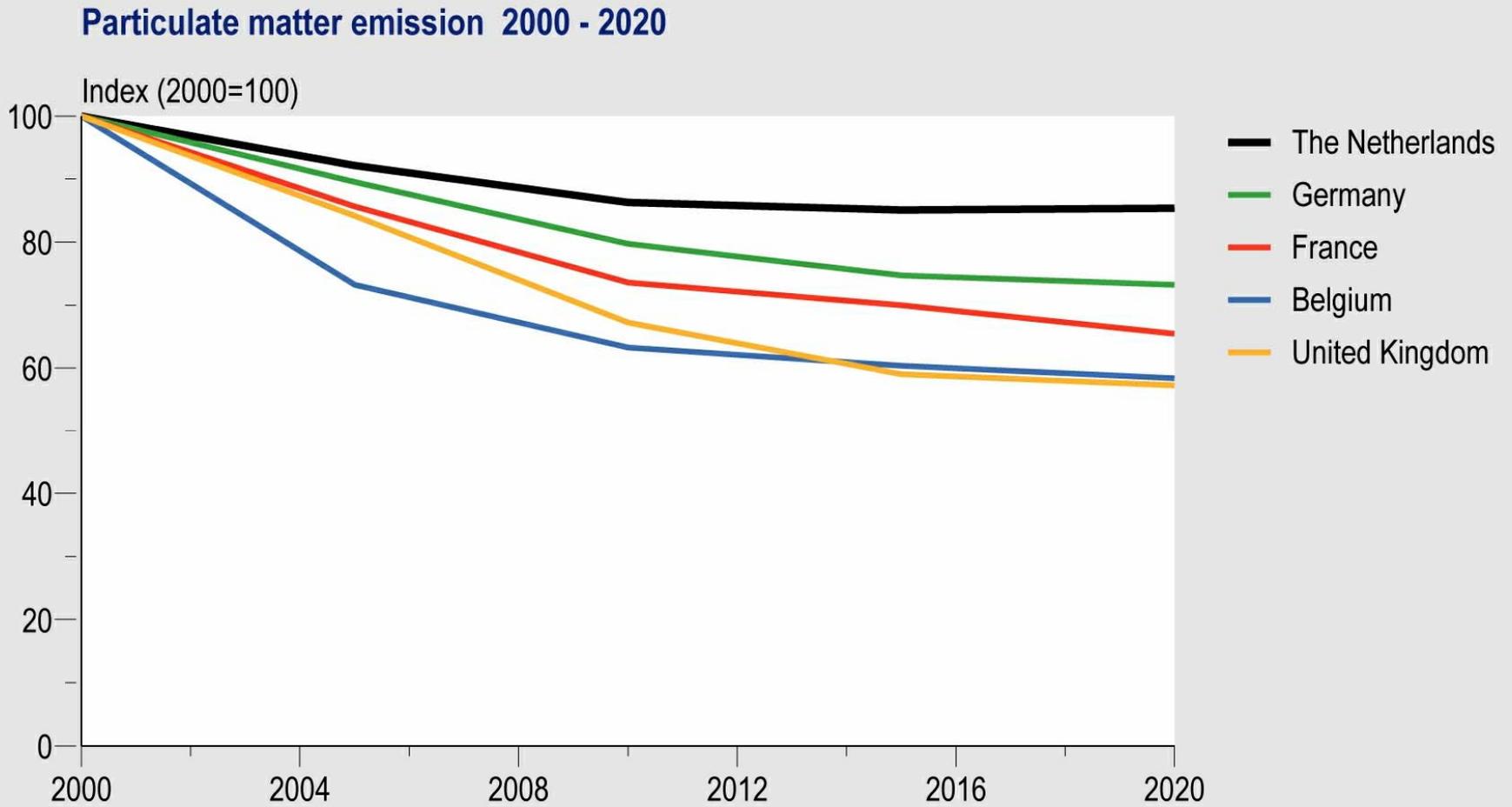
<sup>d</sup> Total consumers, trade, services, government and construction.

# Emission of primary particulate matter 1995-2003.

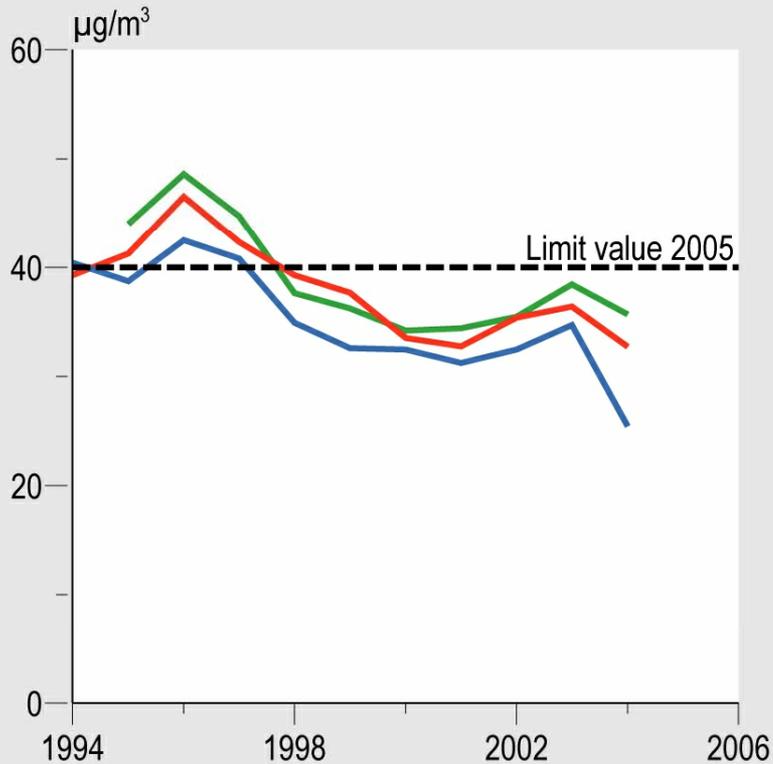
Particulate matter emission 1995 - 2003



# Emission of primary particulate matter between 2000-2020

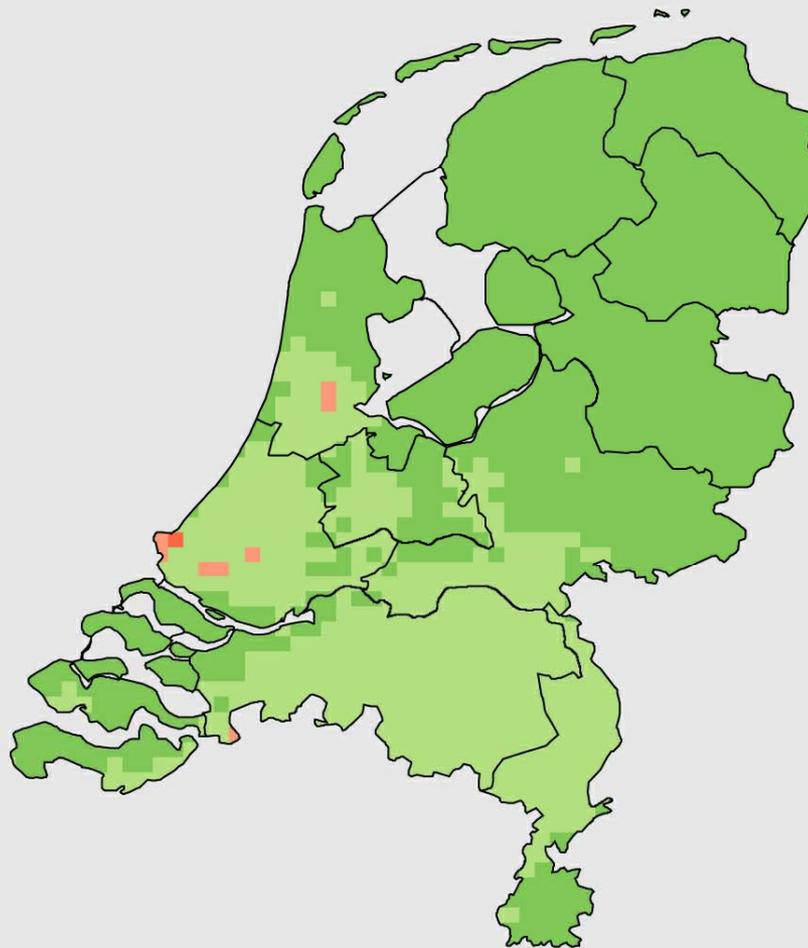


# Trends



- Street
- Urban
- Regional

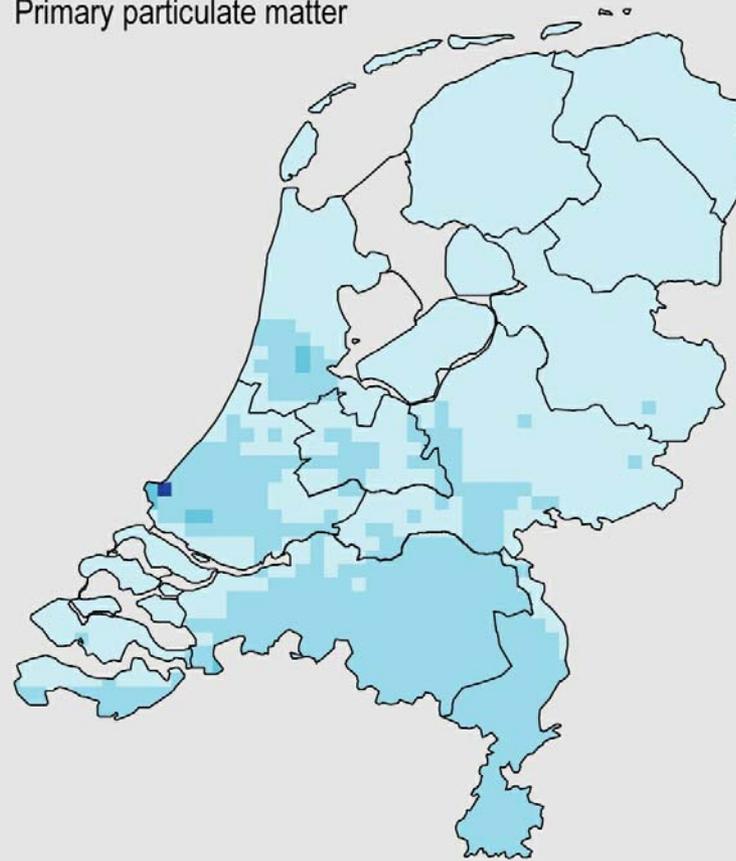
# 2003



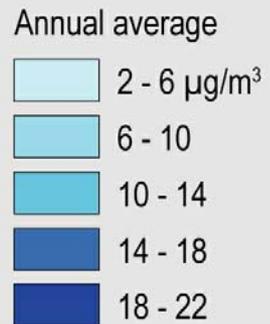
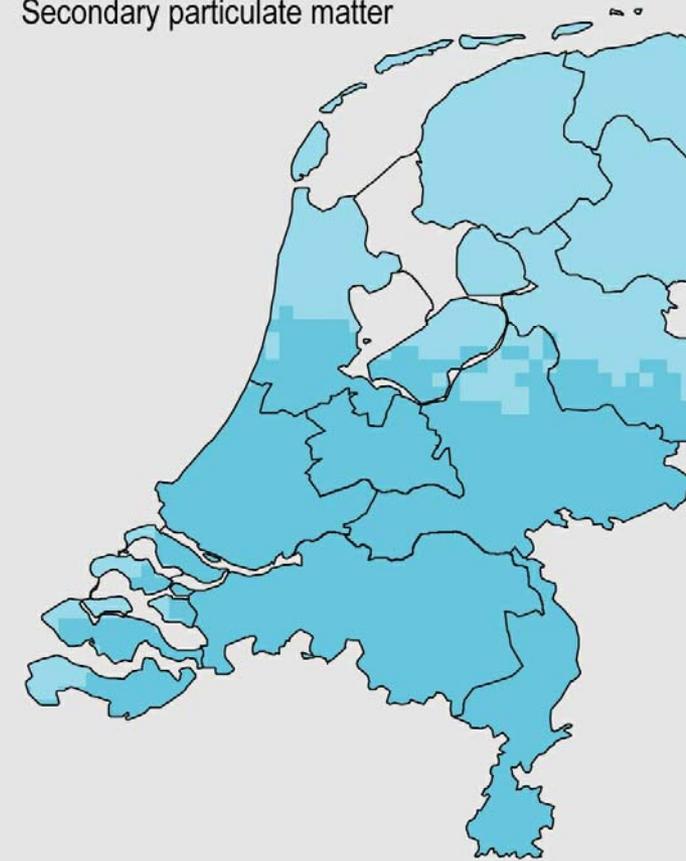
- Annual average
- 30 - 35  $\mu\text{g}/\text{m}^3$
  - 35 - 40
  - limit value 2005
  - 40 - 45
  - 45 - 50

Measured annual average PM concentrations in the Netherlands in 2003

Primary particulate matter



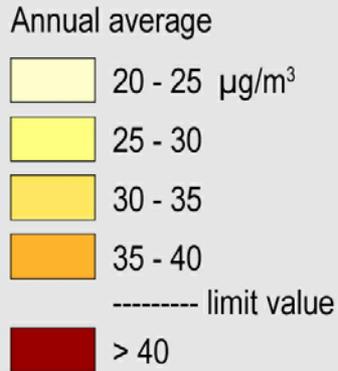
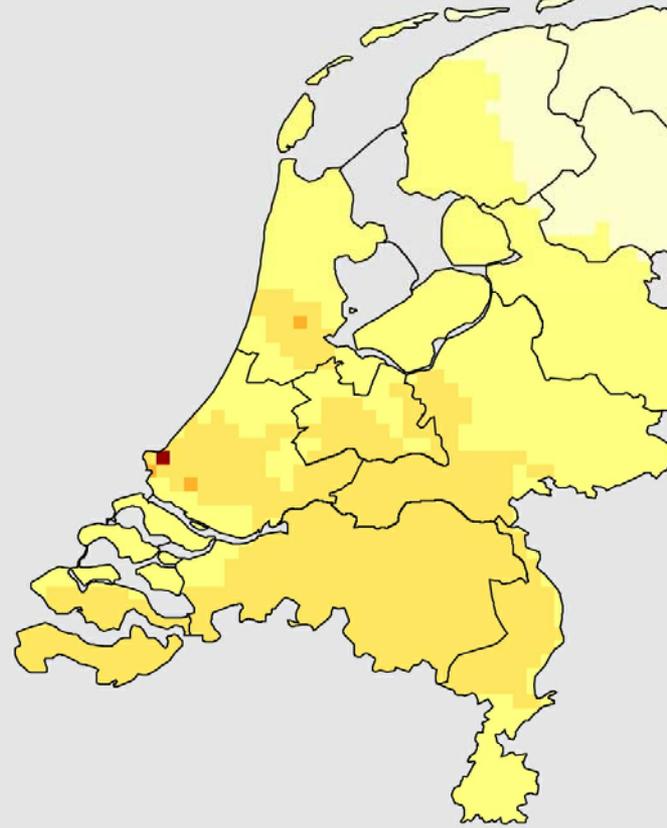
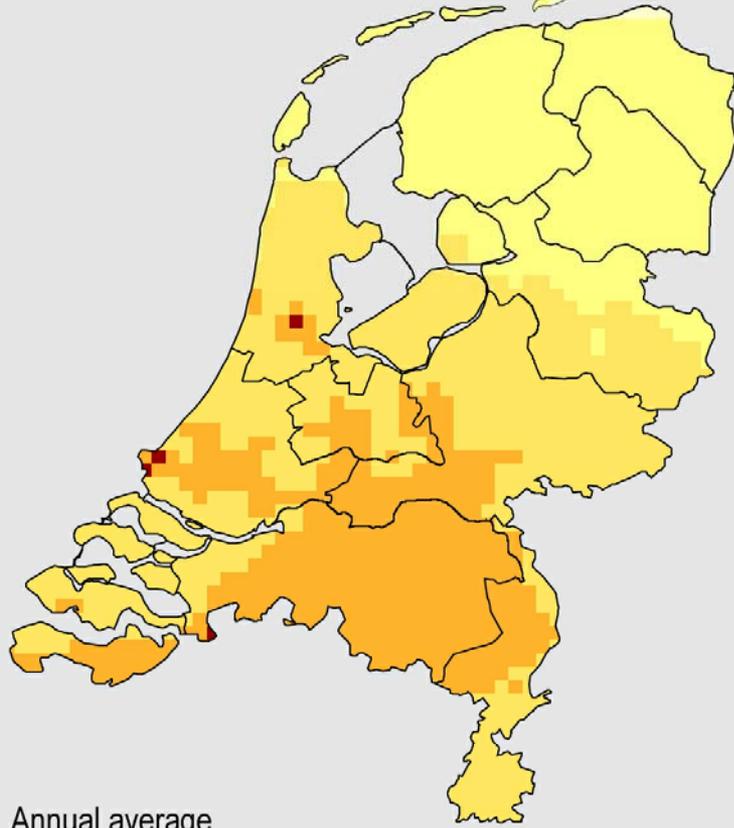
Secondary particulate matter



Calculated concentrations of primary and secondary particulate matter in 2003

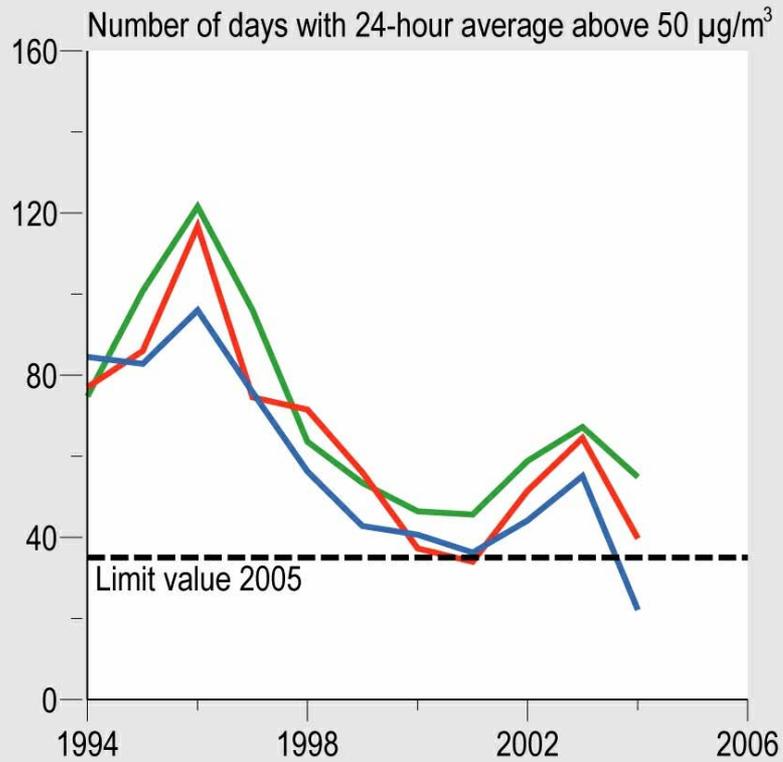
2000

2010



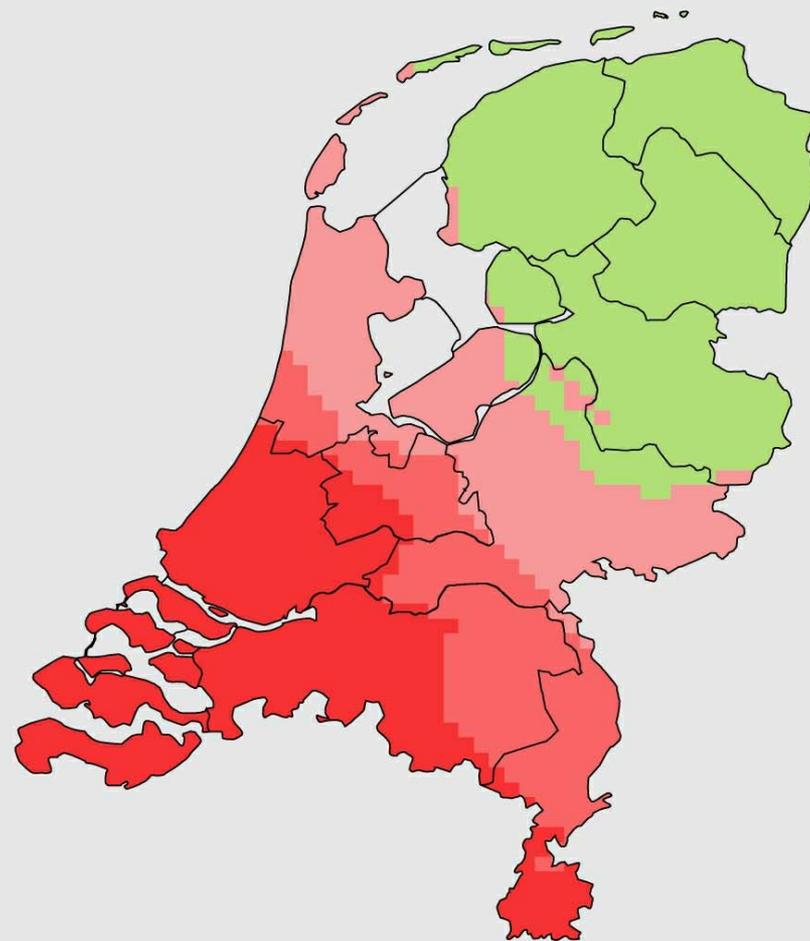
Calculated annual average concentration of particulate matter in 2000 (left) and 2010 (right)

## Trends



- Street
- Urban
- Regional

## 2003



Number of days with 24-hour average above  $50 \mu\text{g}/\text{m}^3$

30 - 35

----- limit value 2005

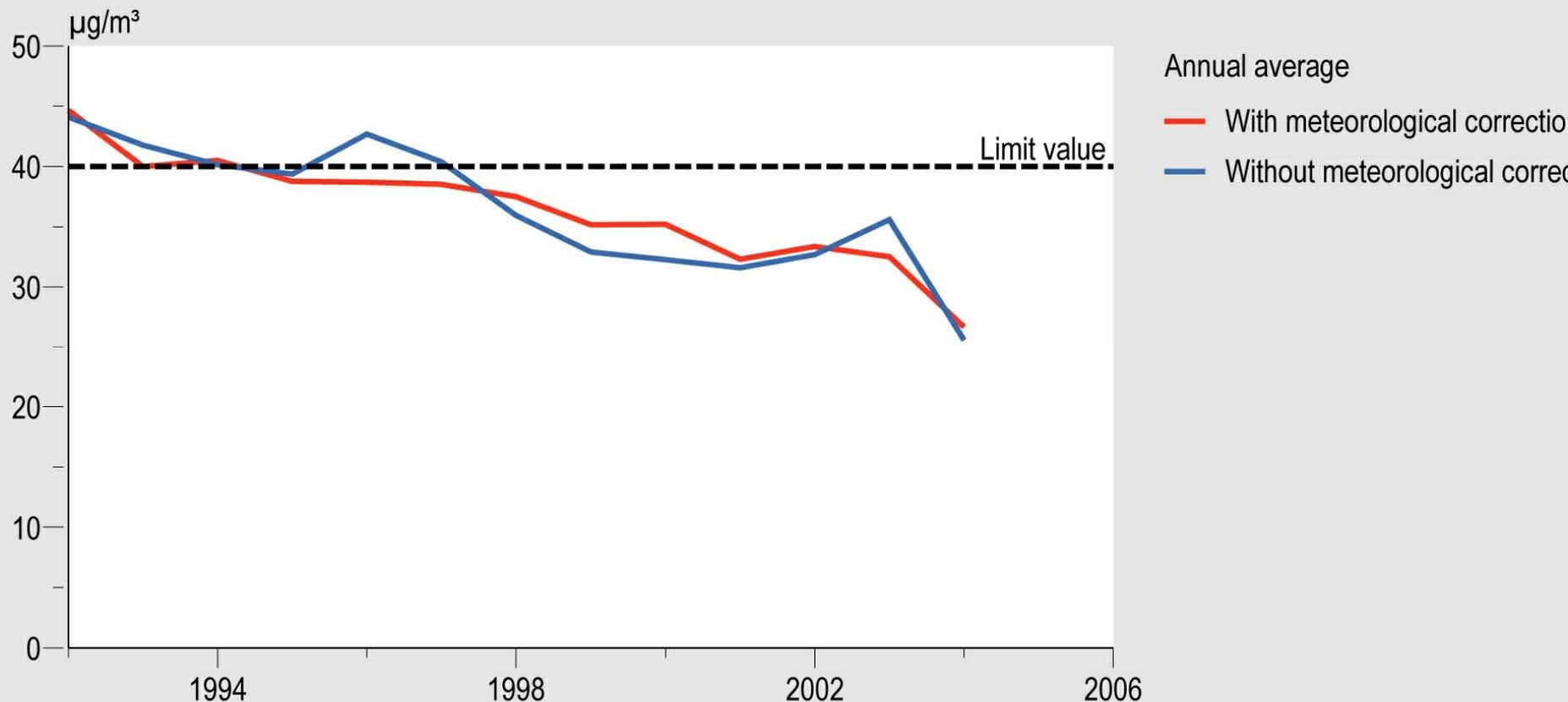
35 - 45

45 - 55

55 - 65

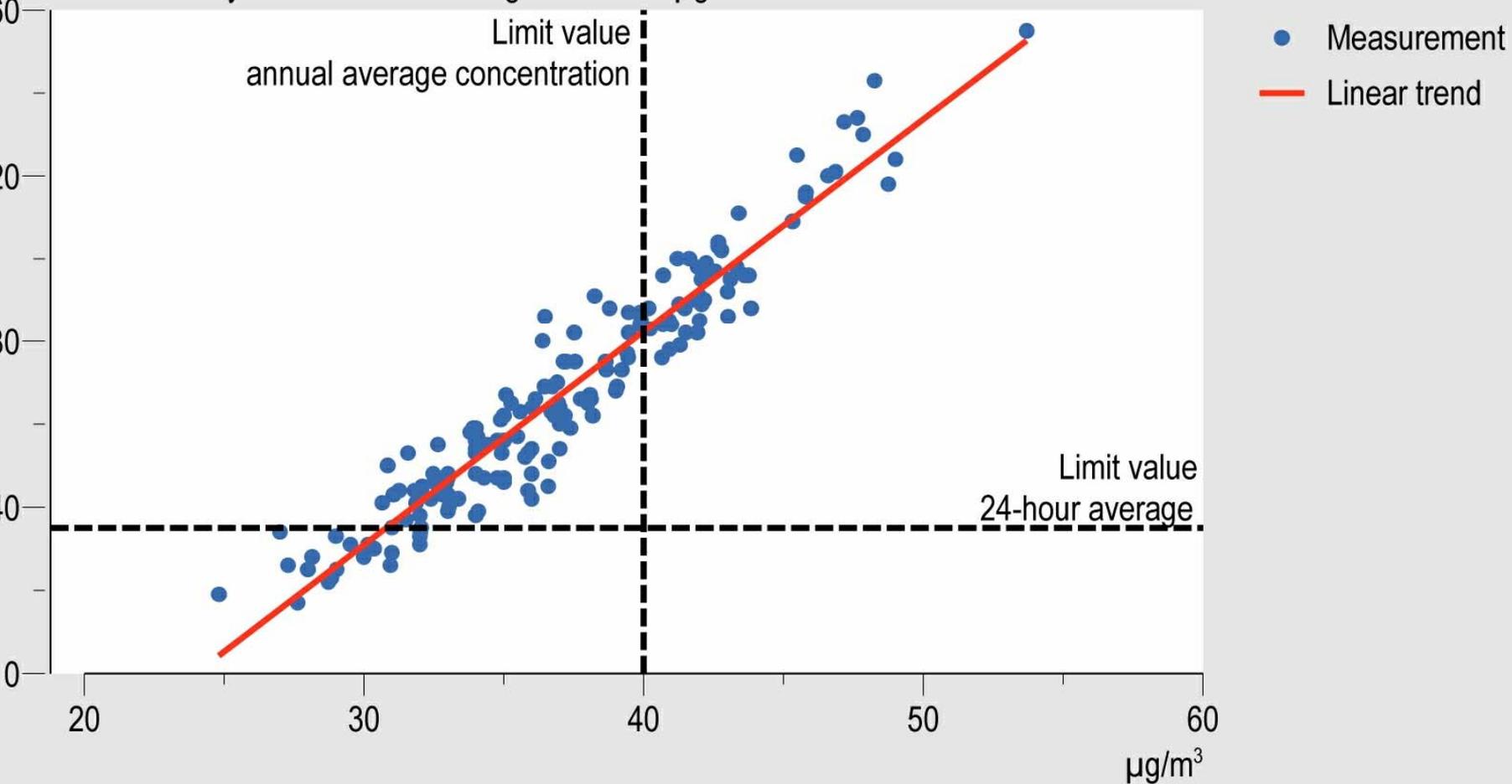
# Measured trend in the annual average particulate matter concentration at the regional stations

Regional particulate matter concentration



## Relationship of annual average particulate matter concentration and number of days exceeding a 24-hour average concentration of $50 \mu\text{g}/\text{m}^3$

Number of days with 24-hour average above  $50 \mu\text{g}/\text{m}^3$



# What's the problem?

- Exceedences of daily standard
- Derogation options allow subtraction of 'natural' PM from the measurements
- This can be sea salt, Sahara dust or other types of natural dust

# Composition of particulate matter in model terms

- Modeled portion

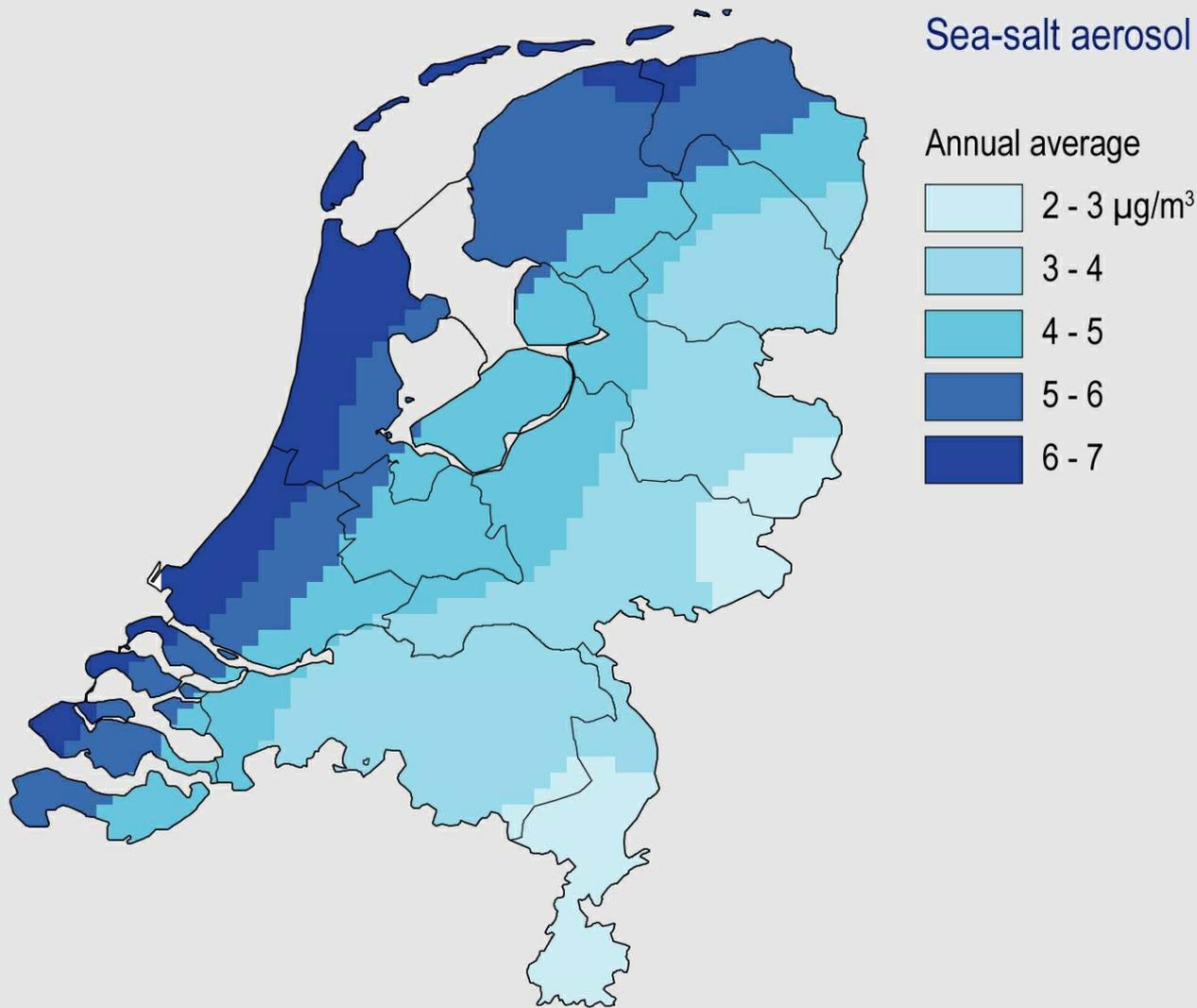
- Primary sources in the Netherlands 8%
- Primary emissions from abroad 10%
- Secondary emissions in the Netherlands 7%
- Secondary emissions from abroad 20%

- Non-modeled portion

- **Sea salt 14%**
- Soil dust 12%
- Northern hemispherical background 3%
- Other 27%

The non-modeled portion is - by definition – the difference between the measured and the calculated concentration.

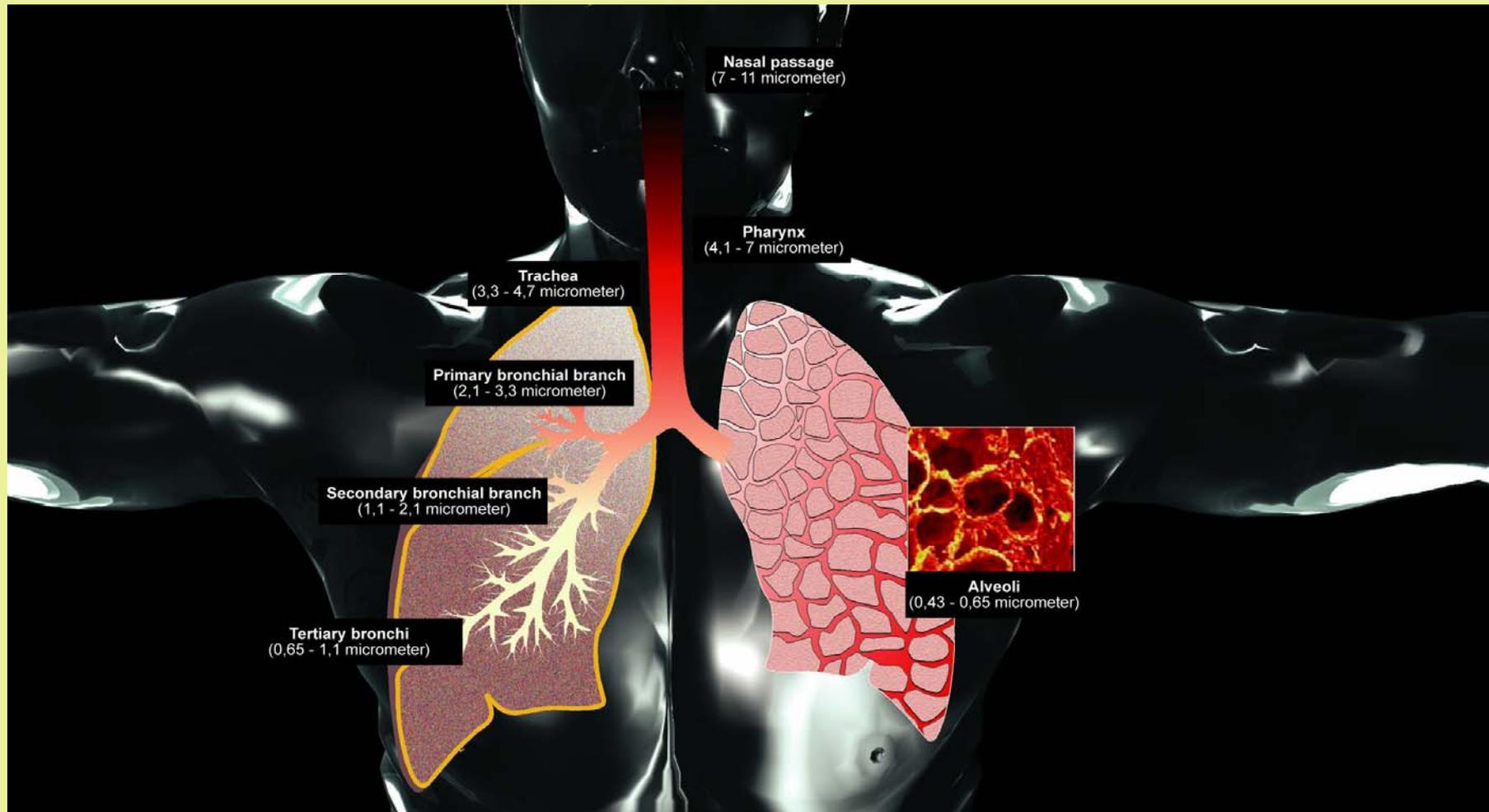
# Annual average contribution of sea salt aerosol to the particulate matter concentration



# What's the problem?

- Subtraction of 'natural' PM from the measurements is scientifically not sound
- Most natural dust is in the air at high wind speeds; these are typically the conditions that NL is in compliance with the standard
- It weakens the standard and allows the gap between the actual levels and the standard to be filled up with anthropogenic PM

# What are the health effects?



# What are the health effects?

- Population: 17 mil
- Associated with short-term exposure to particulate matter, it is estimated that 2300-3500 people die prematurely each year in the Netherlands, with 3000 as an average
- Associated with long-term exposure estimated that the magnitude of these effects in the Netherlands would be between 12,000 and 24,000 premature deaths per year with 18,000 as an average, at an annual average particulate matter concentration of 35  $\mu\text{g}/\text{m}^3$ .

## Particulate matter considered: What do we know for certain in the particulate matter dossier?

- Airborne particulate matter can lead to a wide range of detrimental health effects, including premature mortality.
- To reduce these health risks, the European Union established air quality norms in the form of limit values.
- Particulate matter concentrations are measured in the Netherlands according to the method prescribed by European legislation.
- Between 1992 and 2003, the concentration of airborne particulate matter declined by  $1 \mu\text{g}/\text{m}^3$  per year on average. The total decline in particulate matter concentration since 1994 has been 25%.

## Particulate matter considered: What do we know for certain in the particulate matter dossier?

- Between 1990 and 2003, the emissions in the Netherlands from known sources of particulate matter and gases from which particulate matter can be formed in the air have declined sharply.
- Measurements and model calculations show that the limit value for the annual average concentration ( $40 \mu\text{g}/\text{m}^3$ ) is exceeded in the Netherlands, but only to a limited extent.
- The limit value for the 24-hour average concentration (no more than 35 days per year exceeding a 24-hour average concentration of  $50 \mu\text{g}/\text{m}^3$ ) is exceeded in large areas of the Netherlands.

## Particulate matter considered: What do we know for certain in the particulate matter dossier?

- Violations of the limit values have been observed in nearly all European cities.
- At least 45% of the average particulate matter concentration in the Netherlands is of anthropogenic origin.
- It is estimated that two-thirds of the anthropogenic particulate matter originates from sources outside the Netherlands and that one-third originates from within the Netherlands.
- On busy streets the concentration originating from within the Netherlands can rise to 30-45%.

# Particulate matter considered: What are the uncertainties in the particulate matter dossier?

- Not all detrimental health effects are known.
- In Europe there is no level playing field regarding the protection of public health.
- The modeling method uncertainty margin of 50% are not considered in the judicial analysis.
- The average estimate is used to determine compliance with the limit values, and measurements and model results are used as if they were absolute values.
- There is a risk that building projects will be suspended in areas where the estimated concentration lies just above the limit value, and the actual concentration lies just below the same limit value, or the other way around.

# Particulate matter considered: How do we proceed in the Netherlands?

- Due to further reductions of particulate matter emissions the air quality will continue to improve.
- The policy is based on the combination of measurements and calculation models. In many other countries, interpretations based only on measurements are thought to be sufficient.
- By disregarding non-hazardous particulate matter fractions of natural origin, such as sea salt, it is easier to comply with limit values and spatial planning limitations can be partly eliminated.
- However, this does not reduce the health risks of particulate matter

# Particulate matter considered: How do we proceed in the Netherlands?

- The particulate matter problem cannot be solved by the Netherlands alone. A European-wide approach is required.
- A policy that focuses on the soot fraction of particulate matter is sensible from a health point of view and appears to be most probably a 'no regret' approach.

# Will air quality standards be relaxed under the proposed new EU Directive (Commissions view)

- No, on the contrary, the Ambient Air Quality Directive would for the first time introduce controls on human exposure to PM2.5 to complement the existing limits on coarse particulate matter
- The proposed approach would establish a concentration cap for PM2.5 in ambient air in the most polluted areas at a level that would prevent unduly high risks to the population (= 25 ug/m<sup>3</sup> ??).
- This would be coupled with an obligation on Member States to reduce average human exposure to urban background levels of PM2.5 over the period 2010- 2020. As far as possible, they would have to aim for a 20% reduction.
-

Table 1: Comparison of the different positions taken by the Commission, Parliament and Council compared to existing legislation

PM10	Adopted legislation	Commission's proposal	Parliament's proposal	Council's approach
<i>Daily limit value for the protection of human health</i>				
A limit value of 50µg/m <sup>3</sup> PM <sub>10</sub> , not to be exceeded ...	... more than 35 times a calendar year since 2005, and, as an indicative limit value, from 1 January 2010, more than 7 times a calendar year	... more than 35 times a calendar year since 2005. So same as adopted legislation requires from 2005.	... more than 35 times a calendar year (unless this cannot be achieved through site-specific dispersal characteristics, or adverse meteorological or geographical reasons in which case value can be exceeded on no more than 55 days)	... more than 35 times a calendar year since 2005. So same as adopted legislation requires from 2005.
<i>Annual limit value for the protection of human health</i>				
A limit value of ...	... 40µg/m <sup>3</sup> PM <sub>10</sub> since 2005, although a stricter limit value of 20µg/m <sup>3</sup> was proposed from 1 January 2010 as an indicative limit value	... 40µg/m <sup>3</sup> PM <sub>10</sub> (so same as adopted legislation, although without the further reduction from 2010)	... 40µg/m <sup>3</sup> PM <sub>10</sub> (in each year up to 2009) and 30µg/m <sup>3</sup> PM <sub>10</sub> (from 2010)	... 40µg/m <sup>3</sup> PM <sub>10</sub> (so same as adopted legislation, although without the further reduction from 2010)

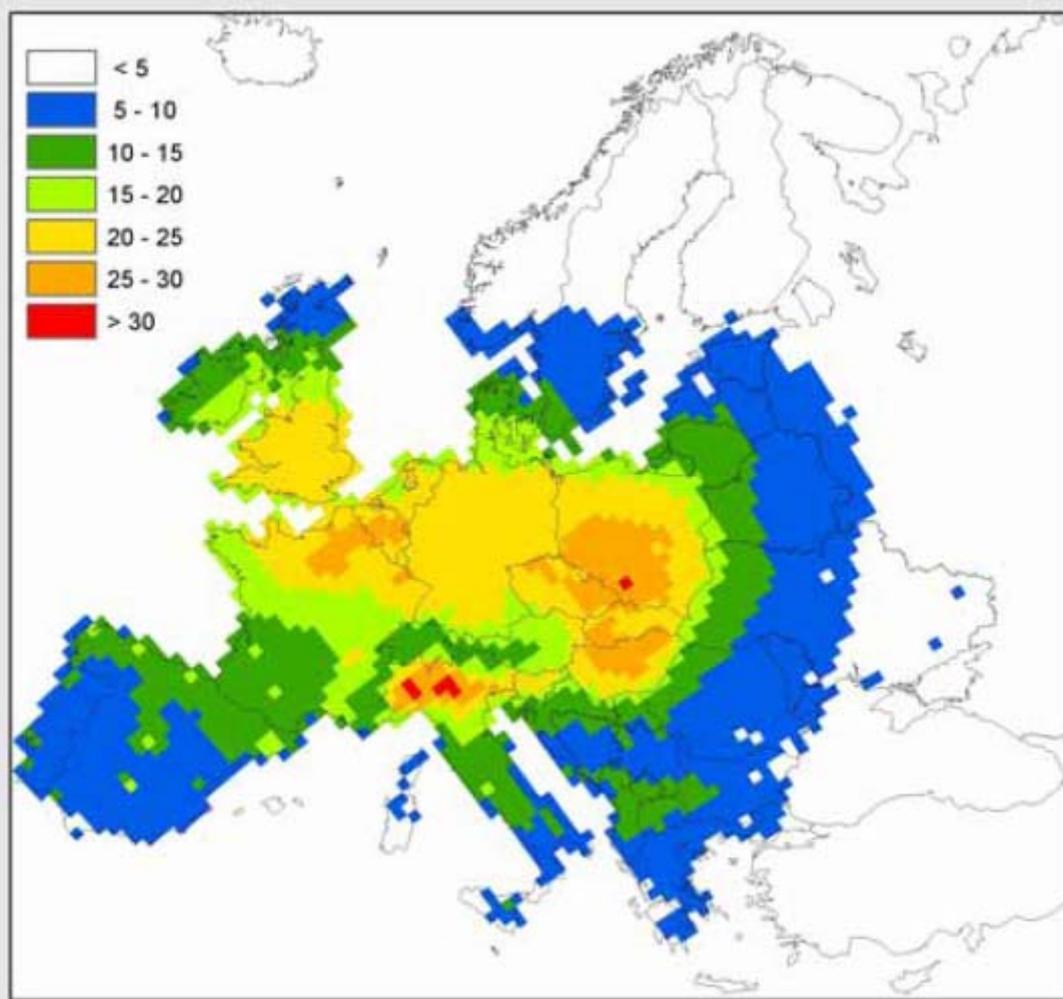
The **European Council** is a meeting of the heads of state or government of the European Union, and the President of the European Commission.

The **European Commission** is the executive body of the European Union.



Table 2: Air quality standards relating to PM<sub>2.5</sub>, as set out in the proposals of the Commission and the Parliament and the approach of the Council

Commission's proposal	Parliament's proposal	Council's approach
<i>Exposure reduction target (relative to Average Exposure Indicator for 2010)</i>		
20% (by 2020)	<p style="text-align: center;"><i>Initial concentration in <math>\mu\text{g}/\text{m}^3</math> Reduction target in percent</i></p> <p style="text-align: center;">&lt; 10 0 %</p> <p style="text-align: center;">= 10 – &lt;15 10 %</p> <p style="text-align: center;">= 15 – &lt;20 15 %</p> <p style="text-align: center;">= 20 – &lt; 25 20 %</p> <p style="text-align: center;">&gt;25 <i>All appropriate measures to achieve the target of 20 <math>\mu\text{g}/\text{m}^3</math></i></p> <p>To be met by 2020</p>	<p style="text-align: center;"><i>Initial concentration in <math>\mu\text{g}/\text{m}^3</math> Reduction target in percent</i></p> <p style="text-align: center;">&lt; 7 0 %</p> <p style="text-align: center;">= 7 – &lt;13 <i>AEI x 1.5%</i></p> <p style="text-align: center;">&gt;13 20 %</p> <p>To be met by 2020</p>
<i>Annual concentration cap</i>		
25 $\mu\text{g}/\text{m}^3$ PM <sub>2.5</sub> (to be met by 1 January 2010)	Deleted	Deleted
<i>Annual target value</i>		
None	20 $\mu\text{g}/\text{m}^3$ PM <sub>2.5</sub> (to be met by 1 January 2010)	25 $\mu\text{g}/\text{m}^3$ PM <sub>2.5</sub> (to be met by 1 January 2010)
<i>Annual limit value</i>		
None	20 $\mu\text{g}/\text{m}^3$ PM <sub>2.5</sub> (to be met by 1 January 2015)	25 $\mu\text{g}/\text{m}^3$ PM <sub>2.5</sub> (to be met by 1 January 2015)



The concentration reduction between 2010 (with current policy) to 2020 with the emission reductions proposed in the Thematic Strategy. The urban scale is not captured in this figure.

## The Commission is concerned at two of the Parliament's amendments in particular

- Extend the extra time allowed for compliance with the PM10 limits beyond the deadline of 1 January 2010 proposed by the Commission
- Weaken the existing daily limit on concentrations of PM10 by allowing it to be exceeded on up to 55 days per year instead of 35 now.
- At the same time, the Parliament has voted to tighten the annual limit on PM10 by reducing it from 40  $\mu\text{g}/\text{m}^3$  to 30 in 2010

# Evaluation new proposal EU

- Of the different limit values for PM10 and PM2.5, the Parliament's proposed annual limit value for PM10 and the equivalent current daily limit value for PM10 seem to be the most binding constraints in densely populated and polluted areas.
- However, if natural contributions to the concentration of particulate matter may be subtracted, there could be a fifty-fifty chance that the PM2.5 limit value proposed by the Parliament becomes the most stringent.

# Evaluation new proposal EU

- The Parliament proposal to tighten the annual limit for PM10 to  $30\mu\text{g}/\text{m}^3$  will ensure better equivalence with the current daily limit value.
- However, at the same time it allows - in specific circumstances - an extension of the number of days that this daily limit value can be exceeded.
- The latter seems to be redundant and makes the system more complex. Additionally, this approach weakens the daily limit value, thus making the annual limit value for PM10 the stronger limit value.



## Toxicological research



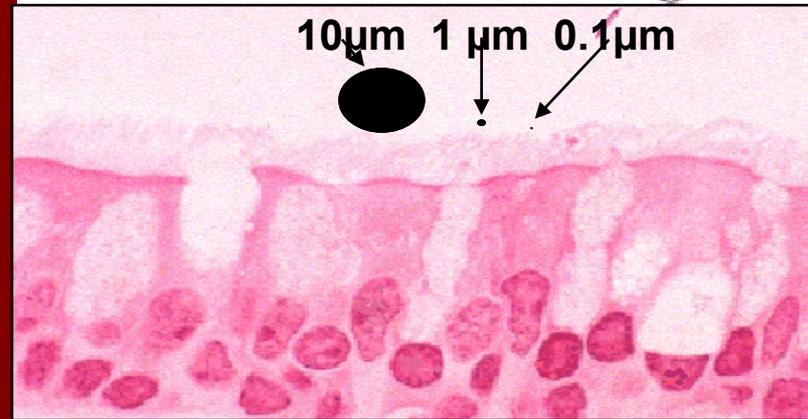
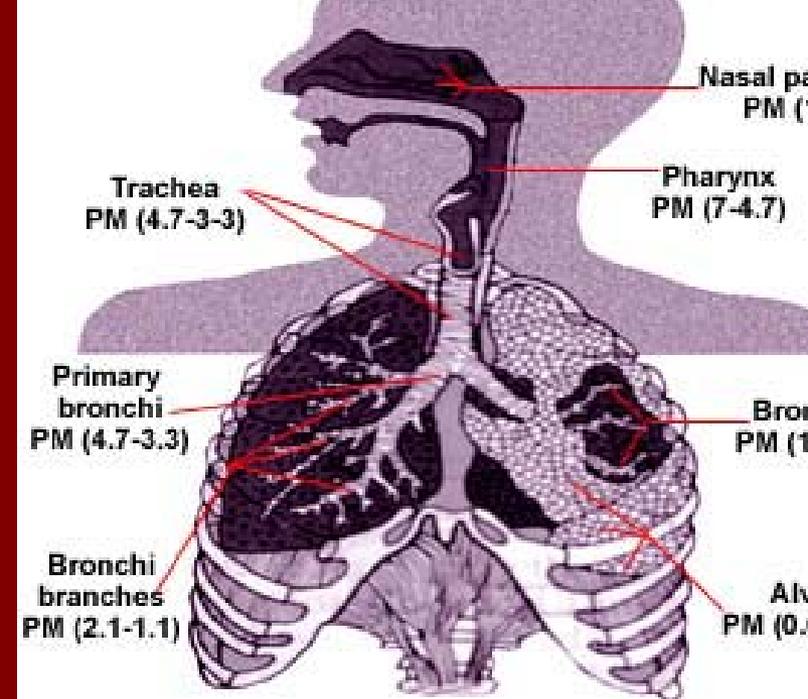
# deposition

← 10  $\mu\text{m}$

← 1  $\mu\text{m}$

← 0.1  $\mu\text{m}$

← 0.01  $\mu\text{m}$



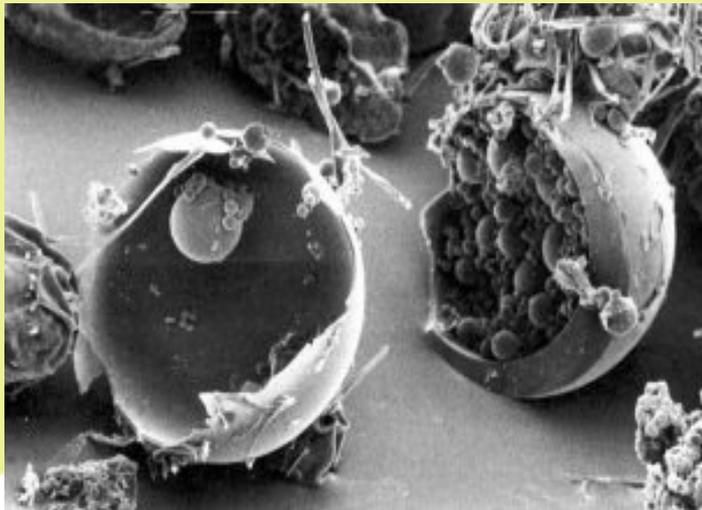
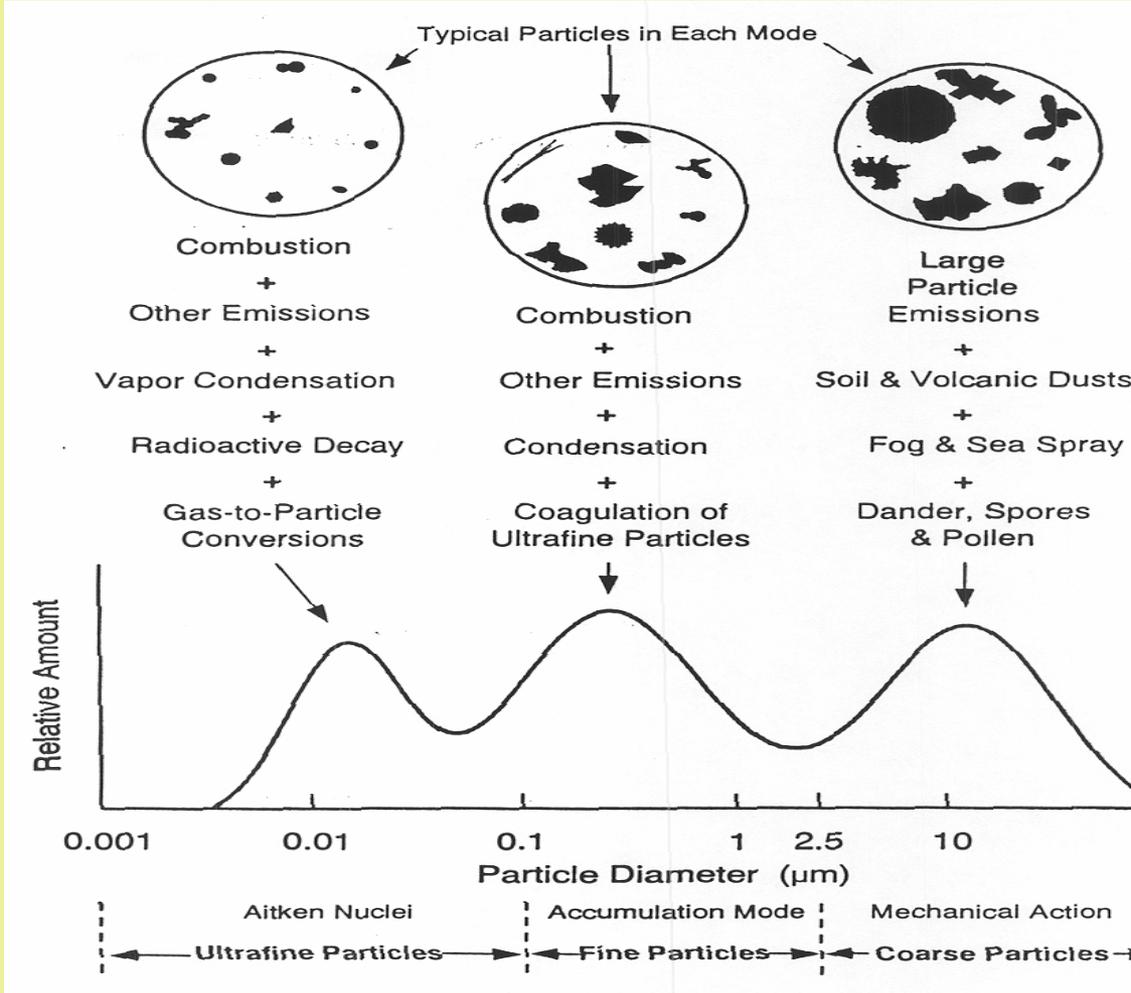
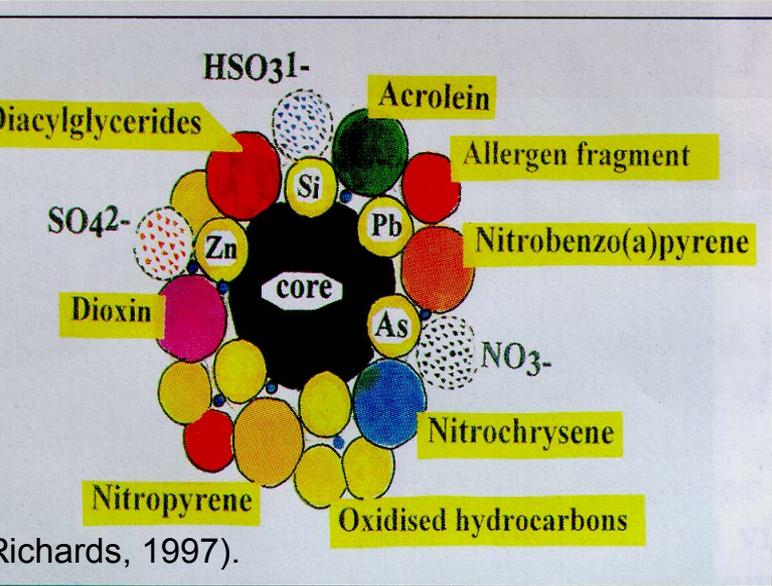
(K. Donaldson)

Size matters

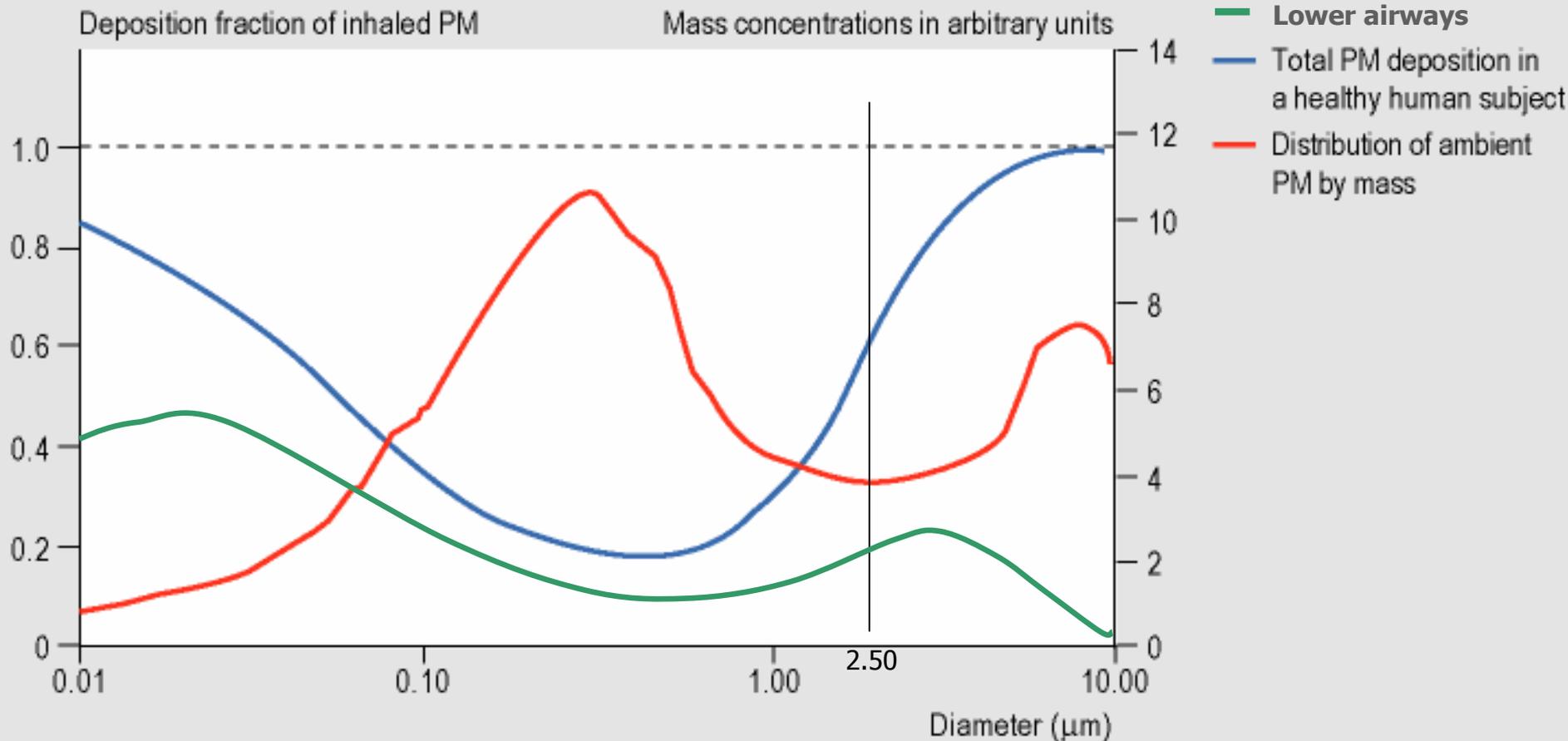
# Why toxicological PM research?

- To increase our understanding on how exposure to PM can result in health effects
  - Causality (support for epidemiological findings)
  - Identification of risk groups (asthmatics?)
  - Effect of policy measures
- To identify
  - the mechanism of action at a biological level
  - the role of particle chemistry and physical properties
  - the role of sources of emission

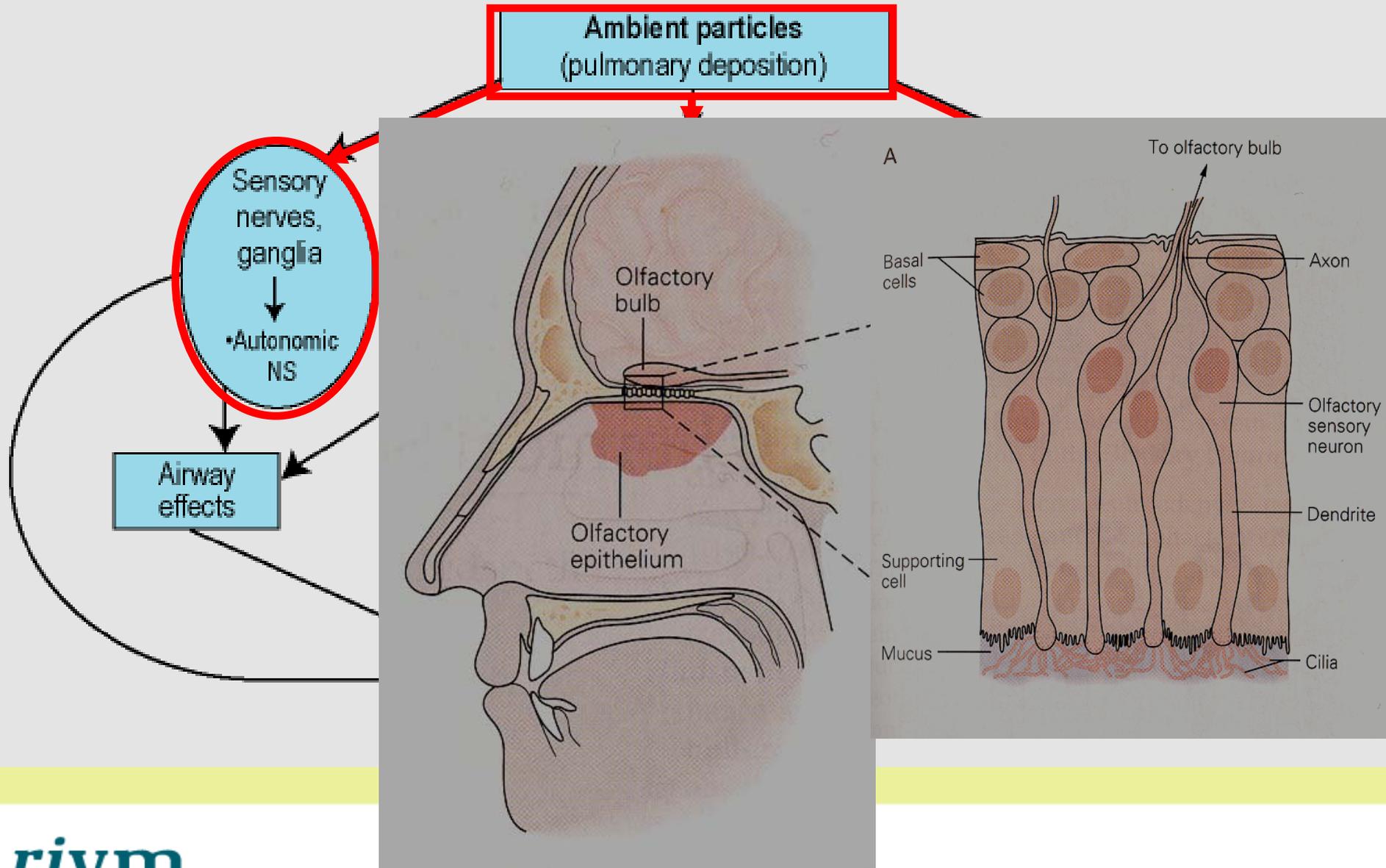
# Physical, chemical or biological characteristics of PM



## Deposition of particles in human respiratory tract



# Biological mechanisms of PM from exposure to effects



# Tools for toxicological research



- Inhalation chamber
- On site exposure



techniques

# Examples of studies with collected PM



- Respiratory Allergy and Inflammation Due to Ambient Particles (RAIAP)

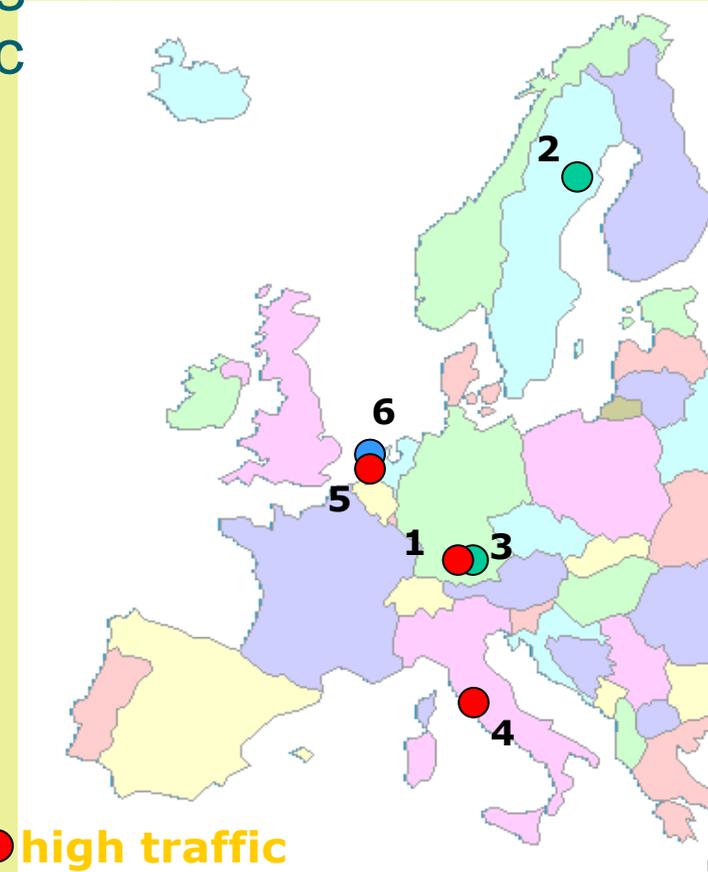


- Health effects of particles from motor engine exhaust and ambient air pollution (HEPMEAP).

# HEPMEAP

- Compare the toxicity of various ambient derived PM samples collected throughout Europe at sites with contrast in traffic density
  - Coarse and fine PM fraction
  - Six PM samples from six different locations with contrast in traffic contribution
  - In vitro tests guide for selection

PM samples	Abbreviation	Traffic intensity
Munich Ost Bahnhof	MOB	high
Hendrik-Ido-Ambacht	HIA	high
Rome	ROM	high
Dordrecht	DOR	medium
Munich Grosshadern hospital	MGH	low
Lycksele	LYC	low

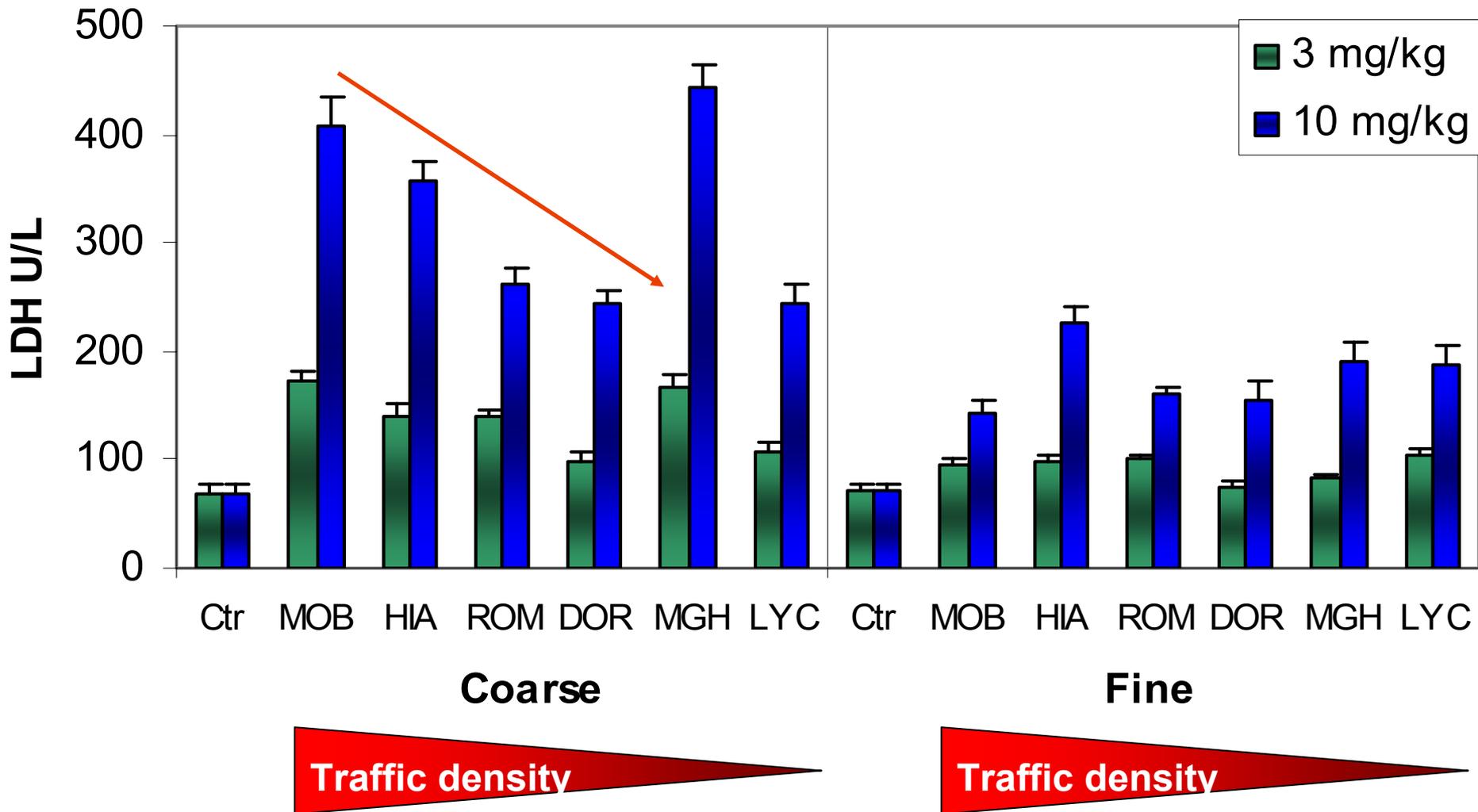


- high traffic
- medium traffic
- low traffic

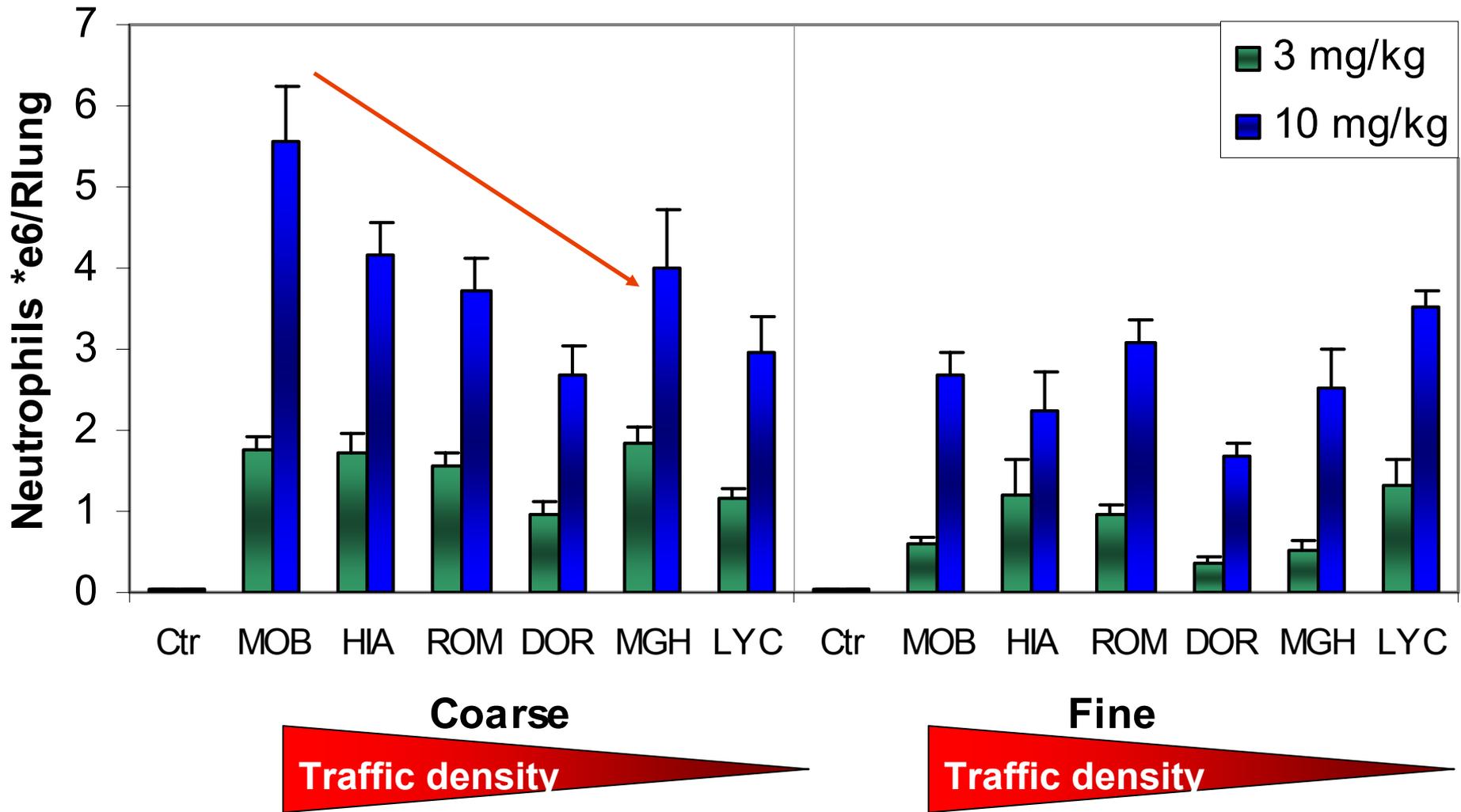
# Methods

- Spontaneously hypertensive rats (male, 12 wk old)
- A single PM dose (3 or 10 mg PM/kg of body weight) by intra-tracheal instillation
- Biological effects were investigated 24 hours after exposure.
  - **Pathology** (including cell proliferation)
  - **Bronchoalveolar lavage fluid** analysis for cytotoxicity, inflammation and oxidative stress (LDH, protein, ALP, CC16, UA, TNF- $\alpha$ , Cell counts, ascorbate, GSH, NAG etc)
  - **Blood** (Fibrinogen, vWF)

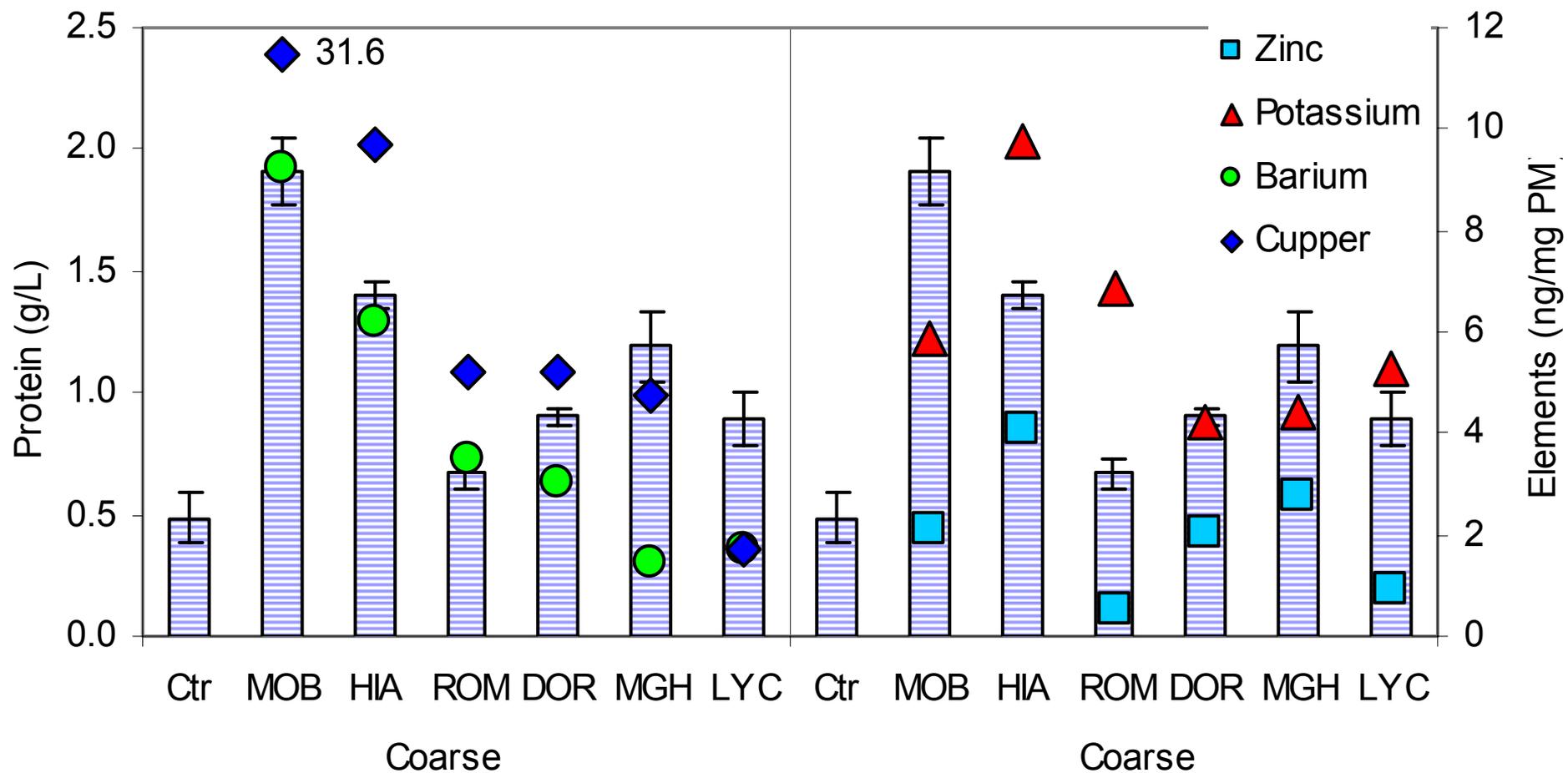
# Results: PM induced cytotoxicity



# Results: PM induced inflammation



# Results: effects in relation to components



**Traffic density**

**Traffic density**

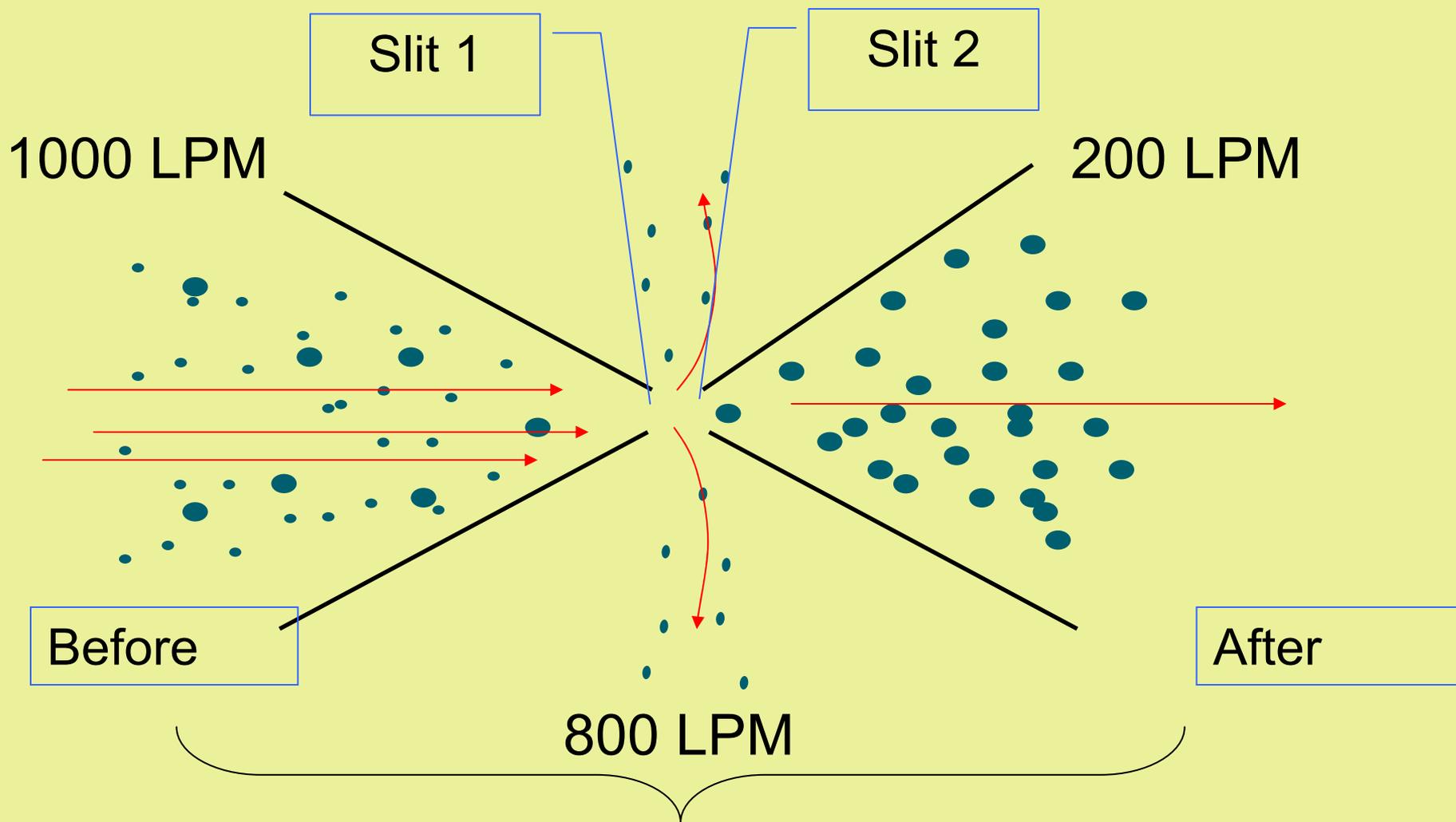
# Summary

- As a general rule samples from locations with high traffic densities induced greater responses than those from low traffic sites. However that two low traffic density sites also appeared to induce relatively large toxic responses
- Ba and Cu (*brake ware*) were associated with cytotoxicity in the coarse fraction and inflammation in the fine fraction; no positive correlation with *tailpipe* emissions
- Zn was associated with pulmonary toxicity (not pathology) in the fine fraction; K (wood smoke?) in fine fraction was linked with the overall pathology score

# RAIAP conclusions

- Differences among locations suggesting role of local sources
- Fine PM stronger allergic response than coarse PM
- PM collected in winter more potent than PM from summer or spring (data not shown)

# Principal of concentrators for particulate matter



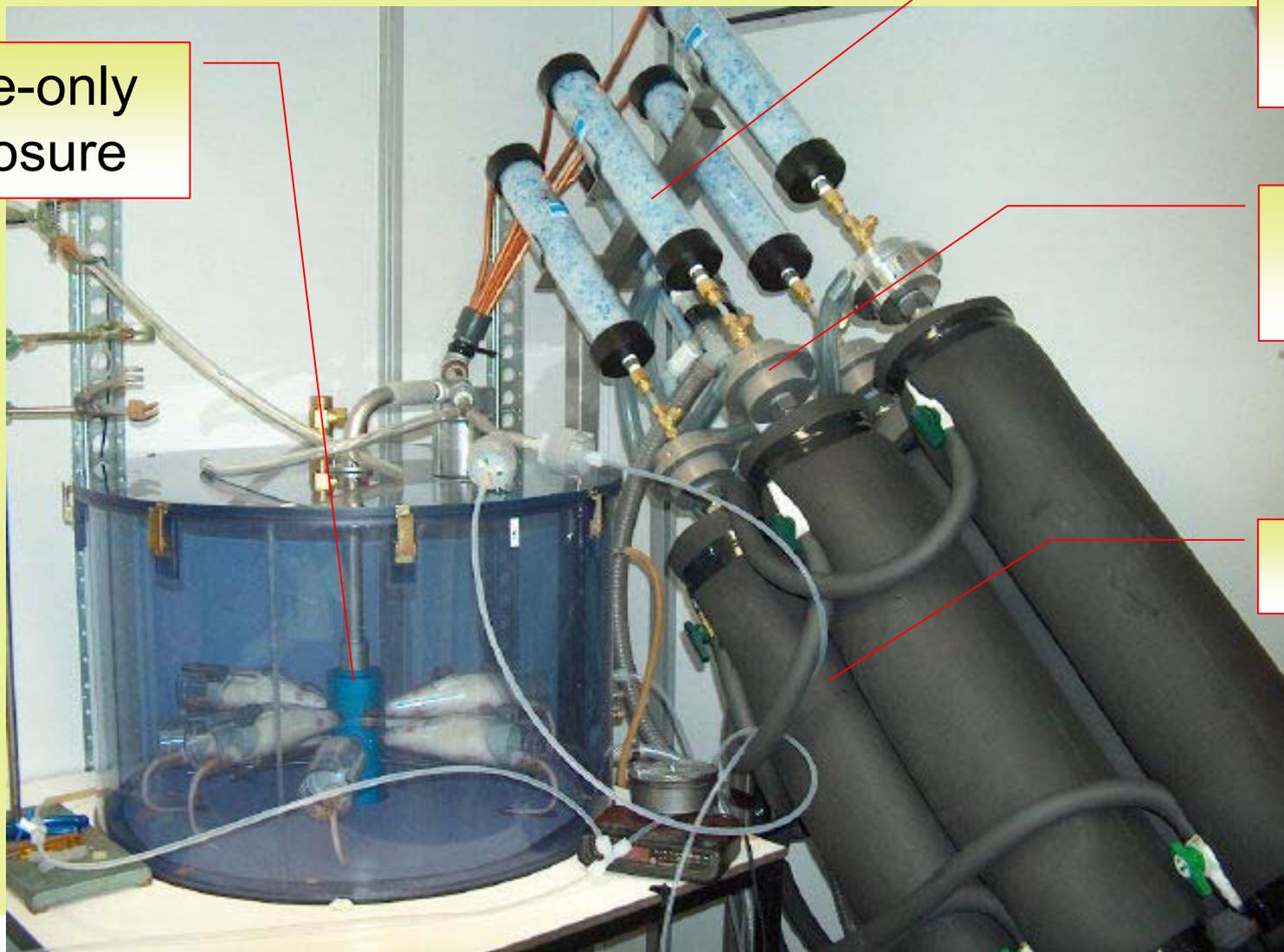
# Example of a concentrator system used for nose-only exposures

Nose-only exposure

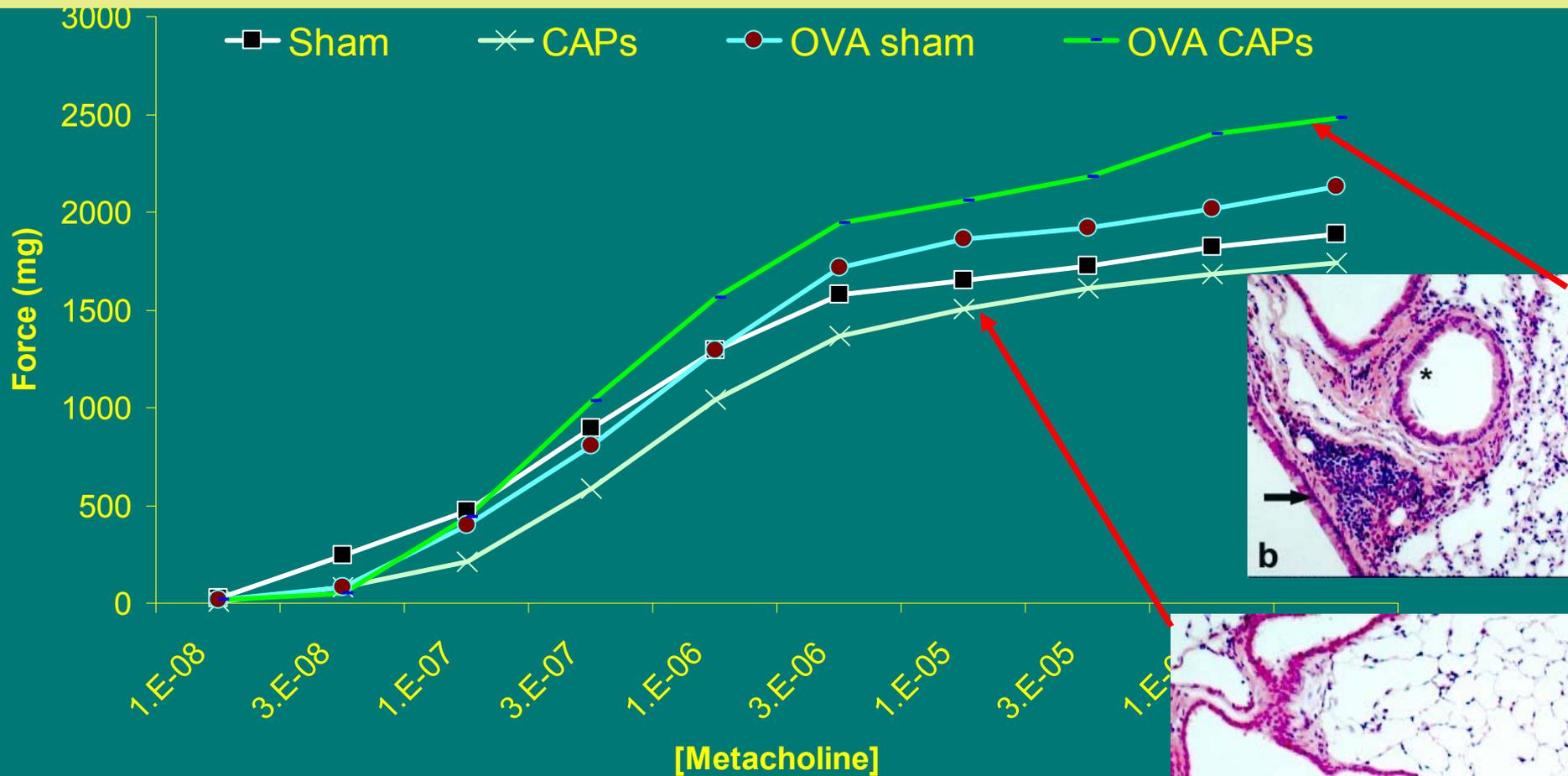
Dryers

Virtual Impactor

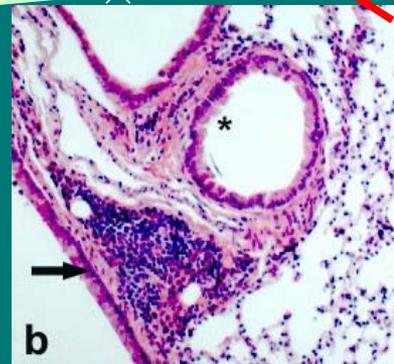
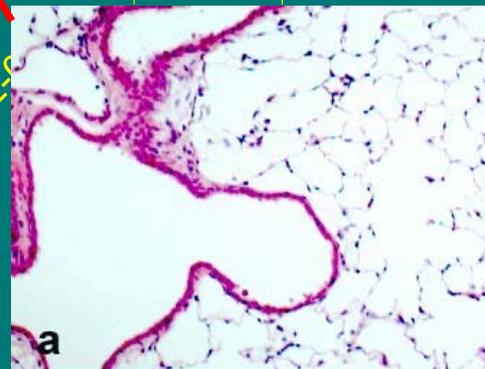
Coolers



are sensitized and challenged with ovalbumin and subsequently exposed to PM

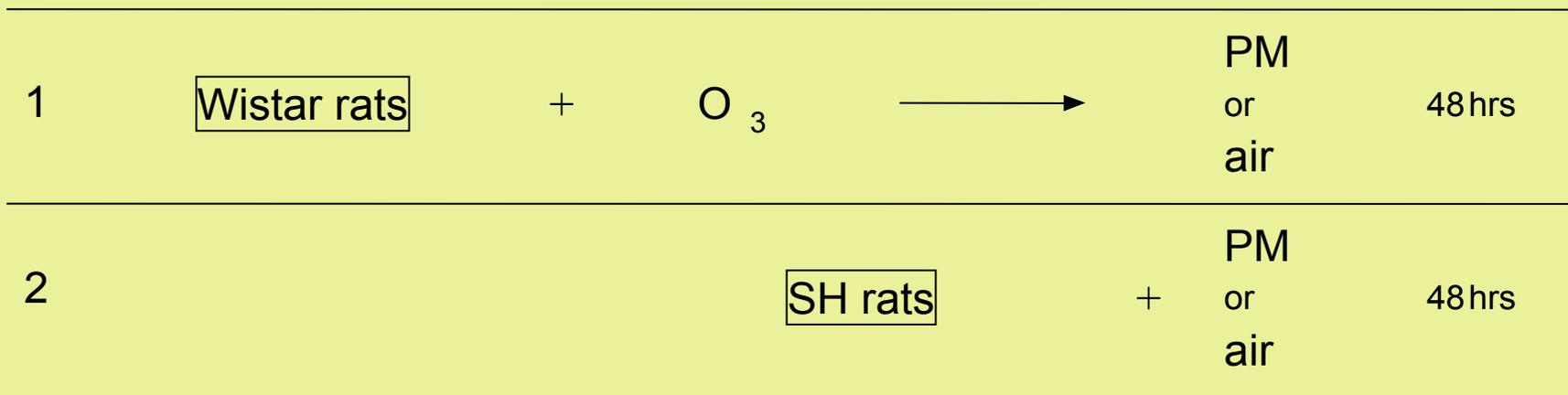


Concentration:

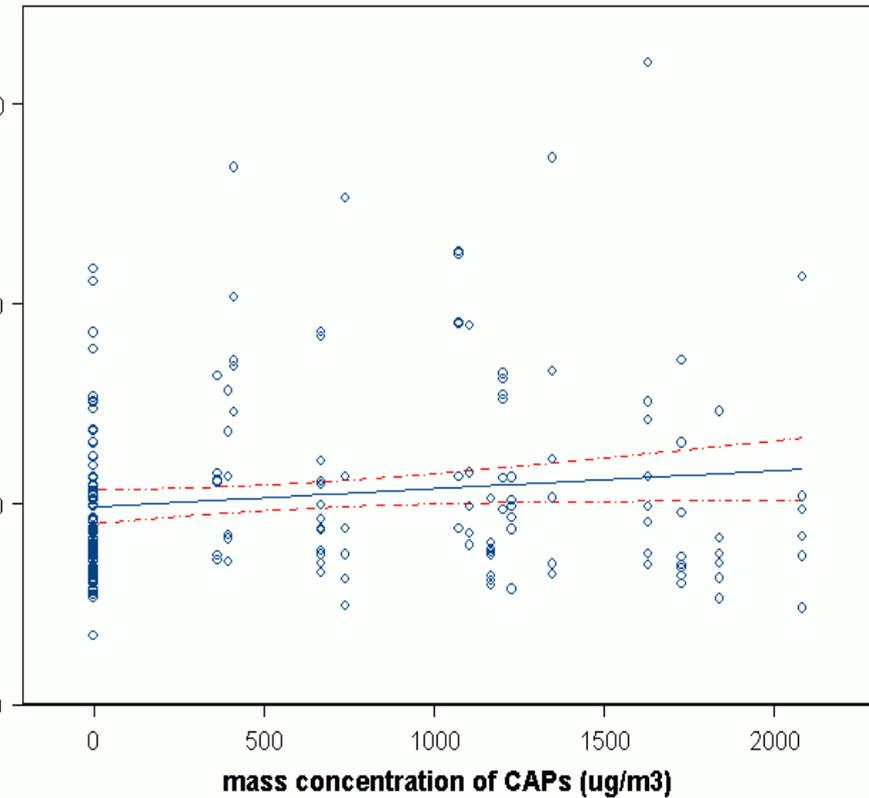


# Exposure to ambient ultrafine+ fine or fine-only PM

- Spontaneously hypertensive rats and ozone-exposed Wistar rats
- Single 6 hr nose-only exposure
- autopsy: lung fluids, pathology and blood analysis
- 3 locations: RIVM, industrial area and motorway/tunnel (HIA)

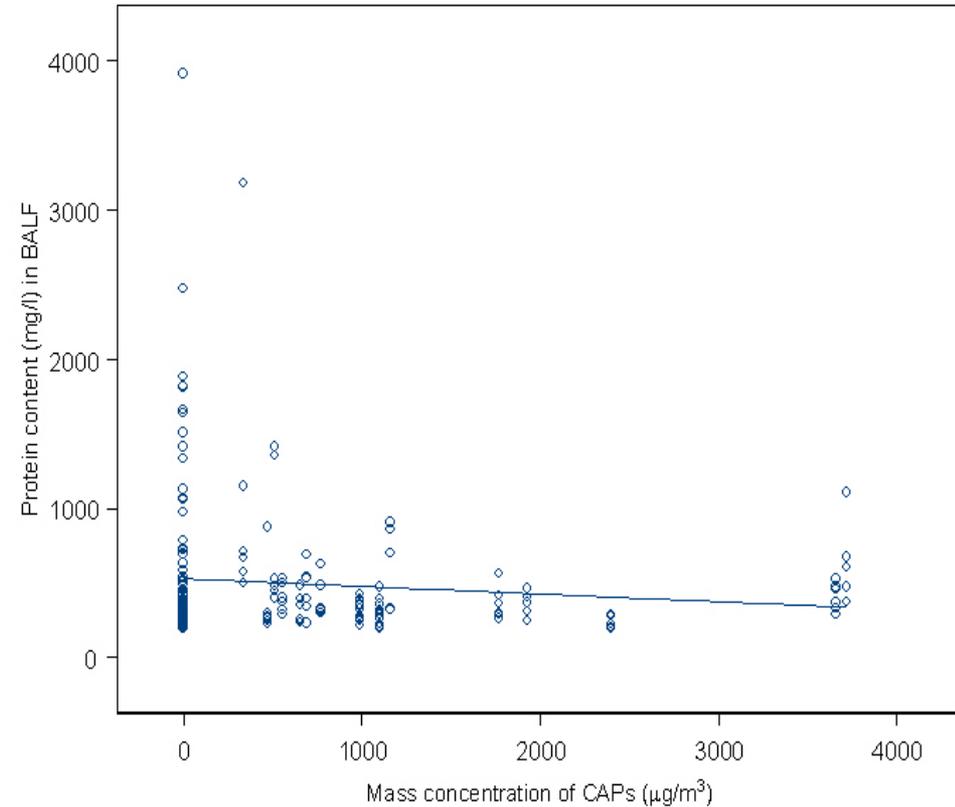


# Concentration-effect relationship protein leakage in the lung due to a 6 hr CAPs exposure



(A) ozone pre-exposed Wistar rats

(8h, 1600  $\mu\text{g}/\text{m}^3$ )



(B) SH rats

# Conclusions 1-day CAPs exposure

- There is no PM mass concentration-effect relationship for any of the investigated parameters
- Inhalation up to 3700  $\mu\text{g CAPs}/\text{m}^3$  did not induce severe toxic or pathological effects in the lung

# Summary of the BALF analysis

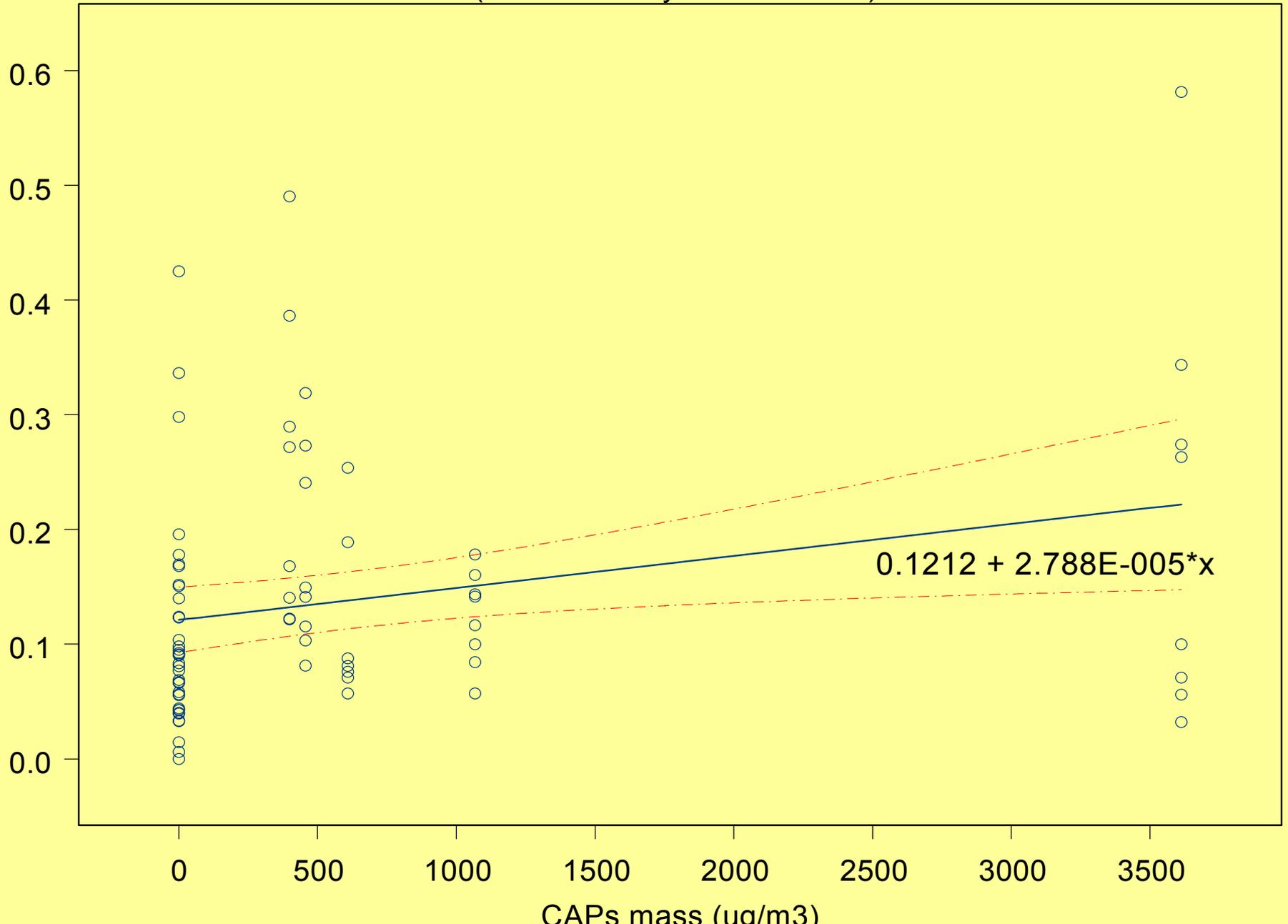
Parameter	Units	Fine - Utrecht		sign.	Ultrafine + Fine - HIA		sign.
		CTRL (n=40)	CAPs (n=40)		CTRL (n=32)	CAPs (n=32)	
		Mean	Mean		Mean	Mean	
ALP	U/L	57.0	51.6		32.7	30.1	
LDH	U/L	66.3	67.6		53.1	52.8	
Protein	mg/L	314	349		266.7	265.6	

On average, no effect of PM on any of the health effect indicators in the lung, however...

GSSG	umol/L	0.155	0.244		0.39	0.31	
GSSG:GSH	ratio	2.61	1.70		4.8	2.7	
TNF- $\alpha$	ng/ml	46.4	47.7		48.7	42.1	
IL-6	ng/ml	0.97	1.30		60.1	64.0	
IL-8	ng/ml	333	311		193.4	193.9	

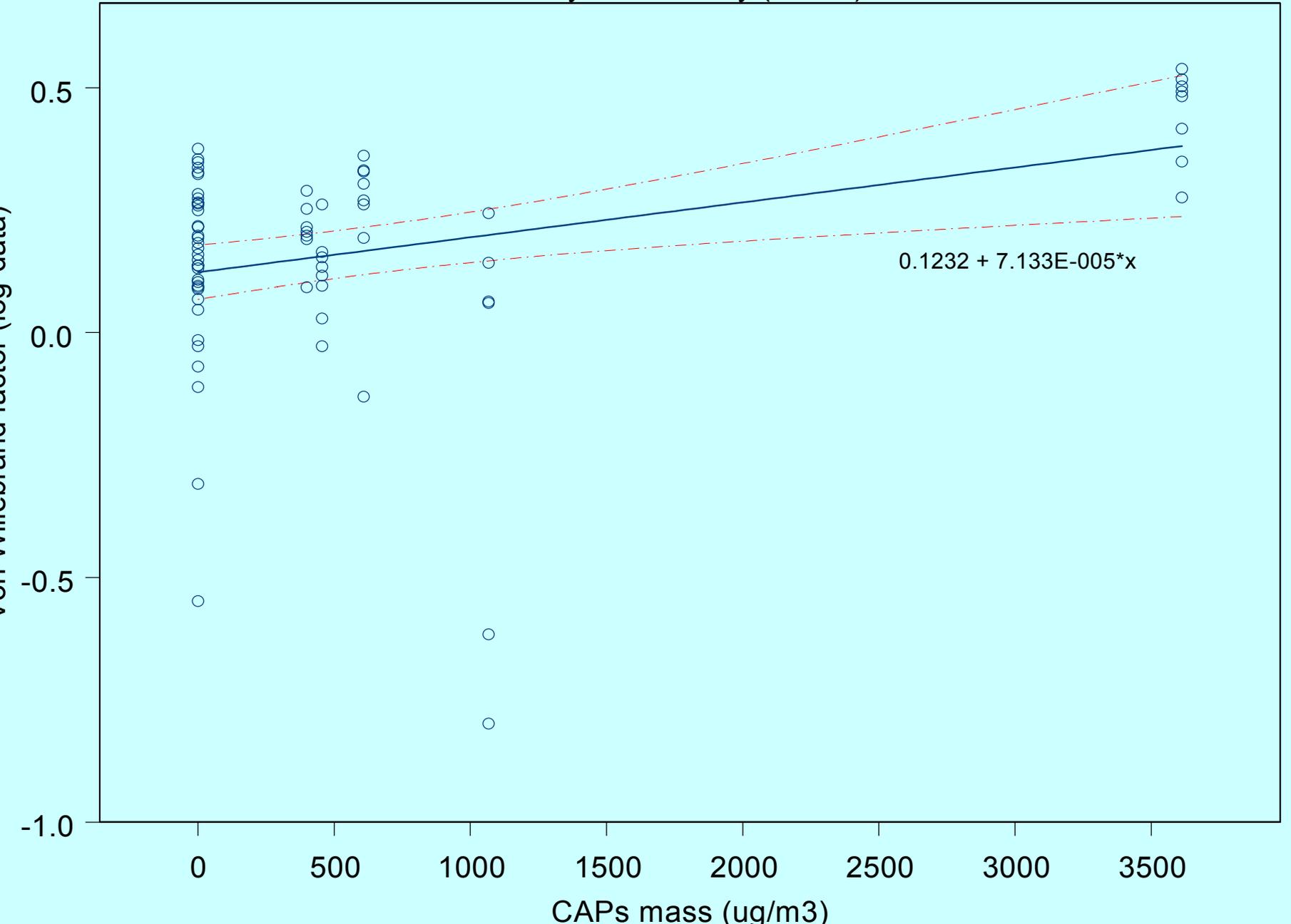
# Relation between mass of CAPs and number of macrophages with inclusions in BALF

( Mass = 2 day's mean value).

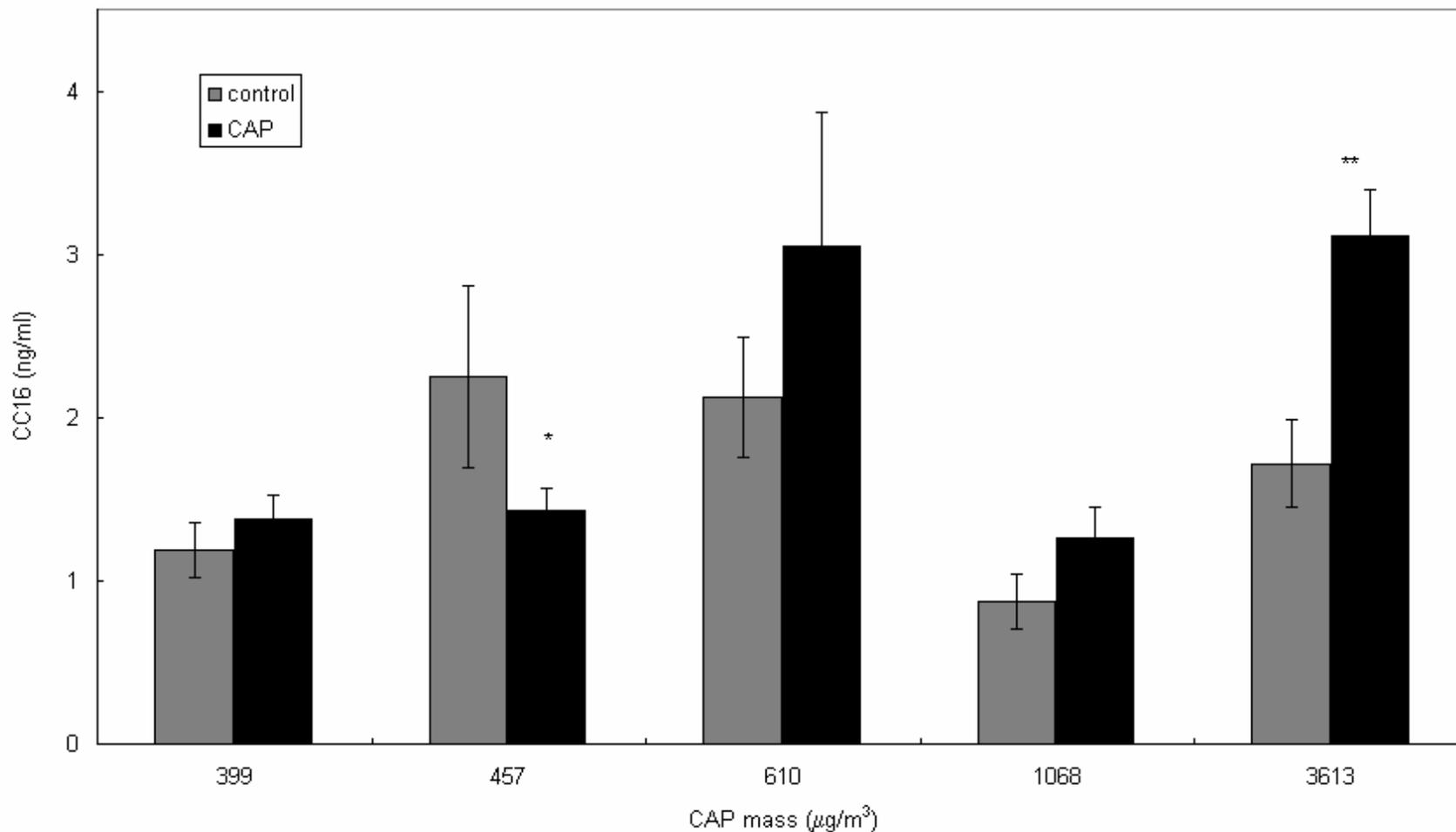


# Relation between Von Willebrand factor and CAPs mass

2 day CAPs study (AFPC)



# Effect on Clara cell protein: no relation with mass



# Conclusions 2-day CAPs

- There were no clear signs of adverse effects in the lung upon exposure, although rats exposed to 'fine' show a trend of increment for 11 out of 15 BALF parameters
- The significant increased number of macrophages with inclusion bodies clearly demonstrated that a substantial part of PM was deposited in the alveolar region
- However....blood analysis showed a significant decrease in total white blood cells and the amount of neutrophils

# Summary

Studies in rodents have shown that

- Both fine and coarse PM have toxic potencies but differ in the mode of action
- PM collected at different locations has differences in toxic potency due to differences in composition
- Effect of PM differ throughout the year
- Mass concentrations of ambient PM are not a good indicator for health effects after inhalation
- Animal models may help us to understand the mechanism and to determine what makes people more susceptible
- Studies in animals has indicated that even in the absence of clear responses in the lung, systemic effects can occur

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Thank you



*rivm*