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 **Empa**
Materials Science and Technology

Secondary emissions from emission control devices and their impact on occupational health and safety



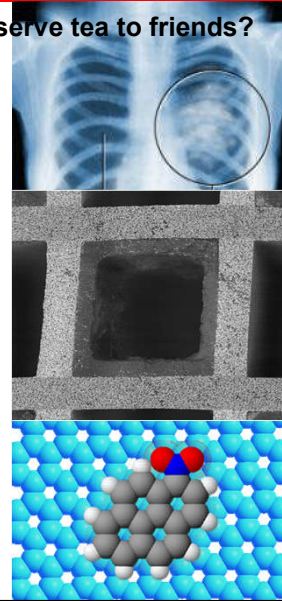
27th Mining Diesel Emissions Council Conference, MDEC
On line, November 30th- December 1st, 2021

Secondary emissions from emission control devices and their impact on occupational health and safety

Would you run a diesel engine in your living room and serve tea to friends?

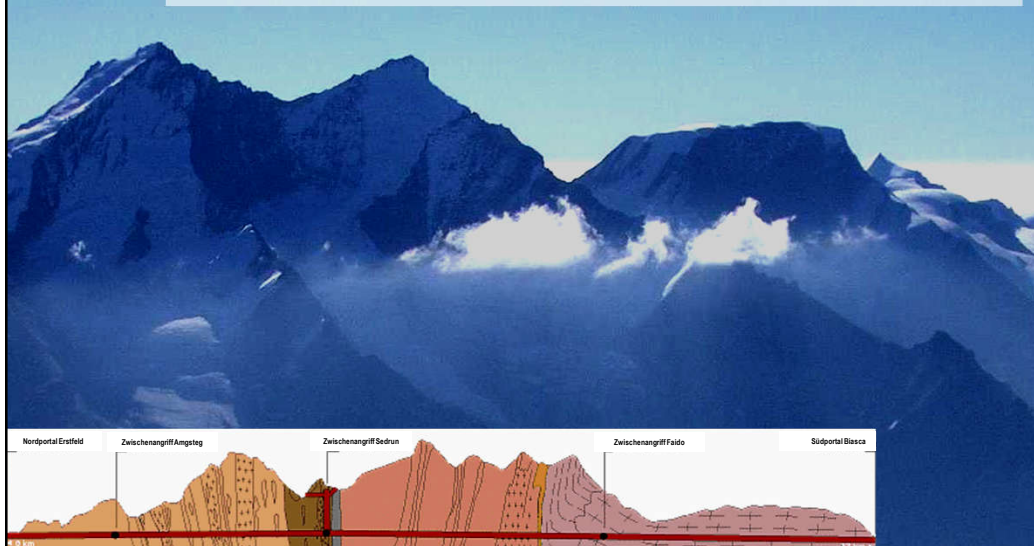
Outline

- **A success story to begin with**
 - VERT-approved DPFs in the NEAT tunnels
- **Risks and health impact of diesel exhausts**
 - What should you know about it?
- **Catalytic diesel particle filters**
 - Do DPFs detoxify diesel exhausts?
- **Secondary emissions of emission control devices**
 - How to avoid or manage them?



NEAT – the longest railway tunnel in the world

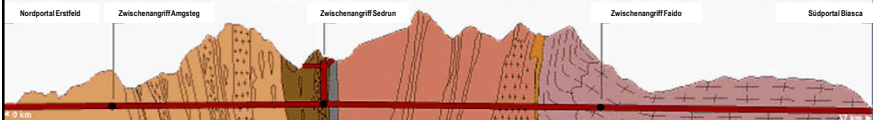
Visionary decisions of the Swiss people 30 years ago!



NEAT – the longest railway tunnel in the world

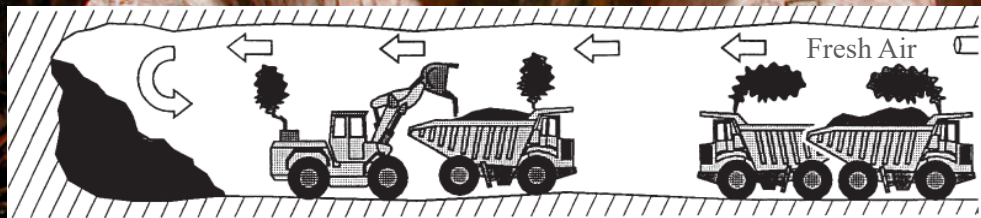
Visionary decisions of the Swiss people 30 years ago!

- 2 National votes:
 - Construction of a railway link through the Swiss Alps, 21. 9. 1992
 - Initiative to protect the Alps, 20. 2. 1994
- 2 Tunnels of 57 km, overall 153 km shafts & tunnels
 - At costs of more than 20 billion CHF
 - Constructed from 1993-2017
 - Hundreds of construction workers and diesel engines involved



NEAT – the longest railway tunnel in the world

- Enough fresh air at the construction site?



NEAT – the longest railway tunnel in the world

- Enough fresh air at the construction site?
- Concentration of pollutants within occupational health limits?
(MWC of diesel exhausts: 0.1 mg EC / m³ (> 1000x dilution needed))
- Start of the VERT project (since 1995 with Empa)
VERT: Verminderung der Emissionen von Realmaschinen im Tunnelbau
(reduction of emissions of real-world engines in tunnels)

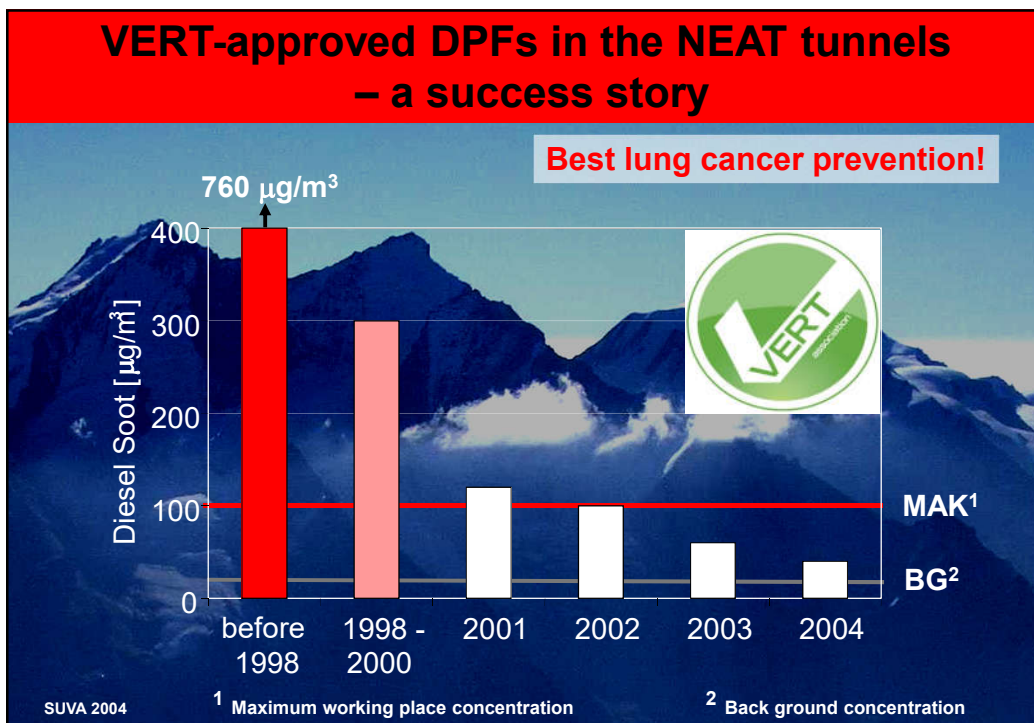
[mg/Nm ³]	Gases				Aerosols	
	CO	NO	NO ₂	SO ₂	PM / DME	H ₂ SO ₄
Emissions of construction heavy duty engines	1000	2700	300	100	250	25
Exposition limits						
Switzerland MAK (max. working place conc.)	35	30	6	5	0.2 (EC + OC)	1
Germany TRGS (limits for working places)	35	30	6	5	0.1 (EC)	1
Required dilution	>26	>90	>50	>20	>1000	>25

**NEAT – the longest railway tunnel of the world
built with VERT-approved DPFs**



**NEAT – the longest railway tunnel of the world
built with VERT-approved DPFs**





20 years VERT filter tests

How to test DPFs, the VERT approach



**Something to celebrate!
in 2018**



20 years VERT filter tests

How to test DPFs, the VERT approach

Approved filters have to:

- Reduce PM- & PN-emissions (>98%)
Both in new and aged conditions (VFT1, VFT3)
- Reduce toxic compounds a.m.a.p.
- Low risks for secondary emissions (VSET)

Are all diesel particle filters safe?

Suitability test

VFT 1
VERT Filter Test 1
Stationary ISO 8178
+ transient
+ regeneration

VSET
VERT Secondary
Emissions Test
ISO 8178
Approx. 200 substances


VFT2
Controlled Field Test
> 2,000 op.hr.
With data-logging

VFT3
Verification test
similar to VFT1,
after field test

Filter List
Approved trap
systems

Field Inspection

100% periodic
field inspection
Smoke puff during free
acceleration
Opacity < 10%



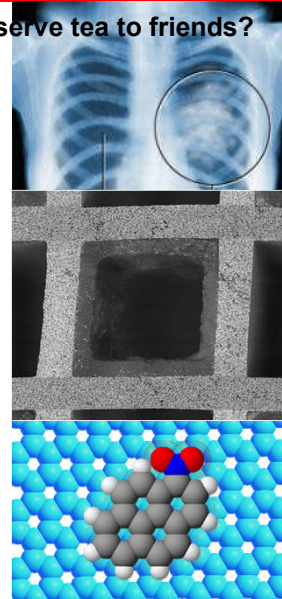
Retained in the Filter List,
if failure rate < 3%
Rejected if >5%

Secondary emissions from emission control devices and their impact on occupational health and safety

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Risks and health impact of diesel exhausts

What do we know about diesel exhaust after 130 years of application?





World Health Organization, IARC
Diesel engine exhaust: A group 1 carcinogen

Diesel engine exhausts cause cancer in humans

International Agency for Research on Cancer
 World Health Organization

PRESS RELEASE
 N° 213

12 June 2012

IARC: DIESEL ENGINE EXHAUST CARCINOGENIC

Lyon, France, June 12, 2012 -- After a week-long meeting of international experts, the International Agency for Research on Cancer (IARC), which is part of the World Health Organization (WHO), today classified diesel engine exhaust as **carcinogenic to humans (Group 1)**, based on sufficient evidence that exposure is associated with an increased risk for lung cancer.

Background
 In 1988, IARC classified diesel exhaust as *probably carcinogenic to humans (Group 2A)*. An Advisory Group which reviews and recommends future priorities for the IARC Monographs Program had recommended diesel exhaust as a high priority for re-evaluation since 1998.

There has been mounting concern about the cancer-causing potential of diesel exhaust, particularly based on findings in epidemiological studies of workers exposed in various settings. This was re-emphasized by the publication in March 2012 of the results of a large US National Cancer Institute/National Institute for Occupational Safety and Health study of occupational exposure to such emissions in underground miners, which showed an increased risk of death from lung cancer in exposed workers (1).

Group 1

Lung cancer in exposed workers

June 12, 2012

World Health Organization, IARC

Diesel engine exhaust: a group 1 carcinogen

Diesel engine exhaust cause cancer in humans

The Diesel Exhaust in Miners Study: A Nested Case-Control Study of Lung Cancer and Diesel Exhaust

Debra T. Silverman, Claudine M. Samanic, Jay H. Lubin, Aaron E. Blair, Patricia A. Stewart, Roel Vermeulen, Joseph B. Coble, Nathaniel Rothman, Patricia L. Schleiff, William D. Travis, Regina G. Ziegler, Sholom Wacholder, Michael D. Attfield

Manuscript received February 16, 2011; revised June 3, 2011; accepted October 21, 2011.

Correspondence to: Debra T. Silverman, ScD, Occupational and Environmental Epidemiology Branch, Division of Cancer Epidemiology and Genetics, National Cancer Institute, Rm 8108, 6120 Executive Blvd, Bethesda, MD 20816 (e-mail: silvermd@mail.nih.gov).

Background Most studies of the association between diesel exhaust exposure and lung cancer suggest a modest, but consistent, increased risk. However, to our knowledge, no study to date has had quantitative data on historical diesel exhaust exposure coupled with adequate sample size to evaluate the exposure-response relationship between diesel exhaust and lung cancer. Our purpose was to evaluate the relationship between quantitative estimates of exposure to diesel exhaust and lung cancer mortality after adjustment for smoking and other potential confounders.

Methods We conducted a nested case-control study in a cohort of 12315 workers in eight non-metal mining facilities, which included 198 lung cancer deaths and 562 incidence density-sampled control subjects. For each case subject, we selected up to 10 control subjects, individually matched on mining facility, sex, race/ethnicity, and birth year (within 5 years), from among workers who were alive before the day the case subject died. We estimated diesel exhaust exposure, represented by respirable elemental carbon (REC), by job and year, for each subject, based on an extensive retrospective exposure assessment at each mining facility. We conducted both categorical and continuous regression analyses adjusted for cigarette smoking and other potential confounding variables (eg, history of employment in high-risk occupations for lung cancer and a history of respiratory disease) to estimate odds ratios (ORs) and 95% confidence intervals (CIs). Analyses were both univariate and multivariate, excluding recent exposure such as that occurring in the 15 years directly before the date of death (or date of reference date for control subjects). All statistical tests were two-sided.

Results We observed statistically significant increasing trends in lung cancer risk with increasing cumulative REC and average REC intensity. Cumulative REC, lagged 15 years, yielded a statistically significant positive gradient in lung cancer risk overall ($P_{trend} = .001$); among heavily exposed workers (ie, above the median of the top quartile [$REC \geq 1006 \mu g/m^3 \cdot y$]), risk was approximately three times greater (OR = 3.20, 95% CI = 1.33 to 7.69) than that among workers in the lowest quartile of exposure. Among never smokers, odds ratios were 1.0, 1.47 (95% CI = 0.28 to 7.50), and 7.30 (95% CI = 1.46 to 36.57) for workers with 15-year lagged cumulative REC exposure of less than 8, 8 to less than 304, and 304 $\mu g/m^3 \cdot y$ or more, respectively. We also observed an interaction between smoking and 15-year lagged cumulative REC ($P_{interaction} = .08$) such that the effect of each of these exposures was attenuated in the presence of high levels of the other.

Conclusion Our findings provide further evidence that diesel exhaust exposure may cause lung cancer in humans and represent a potential public health burden.

J Natl Cancer Inst 2012;104:1-14

Lung cancer in exposed workers

12'315 workers, 8 mines

198 lung cancer death

(16'000 in 1'000'000)

(1 in 1'000'000, target value Swiss LRV)

diesel exhaust exposure: a potential public health burden

Occupational health regulation in Switzerland

Grenzwerte am Arbeitsplatz 2009

suvapro
Sicher arbeiten

Stoff [CAS-Nummer]	MAK-Wert		Kurzzeitgrenzwerte		HSB	C	M	R ₁	R ₂	SS	Messmethoden/ besondere Bemerkungen
	ml/m ³	mg/m ³	ml/m ³ (ppm)	mg/m ³							
1,3-Dichlorpropen (cis und trans) [542-75-6]	0,11	0,5			HS	2	3				
2,2-Dichlorpropionsäure [75-99-0] und ihr Natriumsalz [127-20-8]	1	6	1	6	15 min						
1,2-Dichlor-1,1,2,2-tetrafluorethan (R 114) [76-14-2]	1000	7000									DFG, NIOSH
Dicyclopentadienyleisen [102-54-9]		10 e									NIOSH
Dieldrin (HEOD) [60-57-1]		0,25 e			H	3					
Dieselmotor-Emissionen (gemessen als elementarer Kohlenstoff)		0,1 a				2					BG

„Diesel engine emissions, measured as elemental carbon, should not exceed 100 $\mu g/m^3$ “

→ The **precautionary principle**: Because diesel engine emissions are **carcinogenic (WHO)** inducing lung cancer in humans, general measures have to be applied to lower exposure to diesel exhausts with best available technology (**BAT**).

Swiss clean air act (LRV): List of carcinogenic compounds

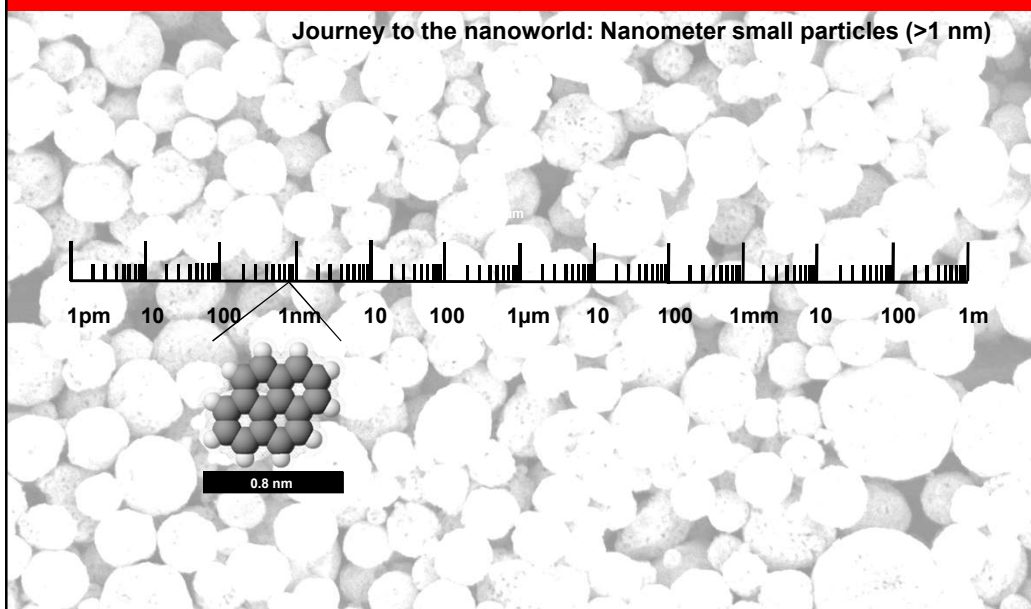
**Luftreinhalte-Verordnung
(LRV)**

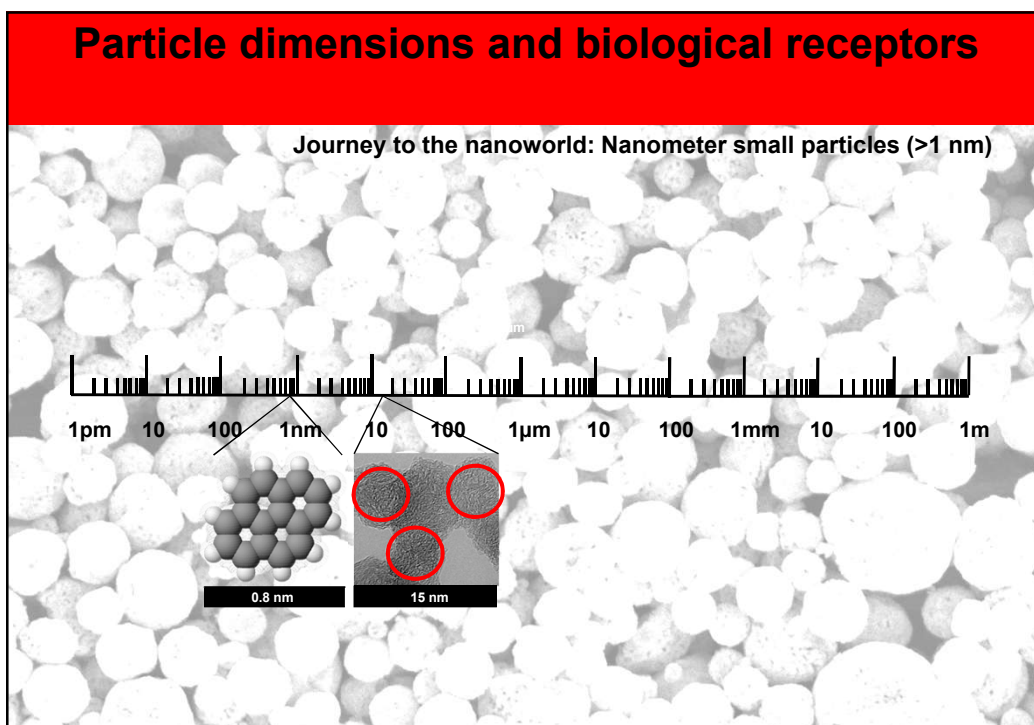
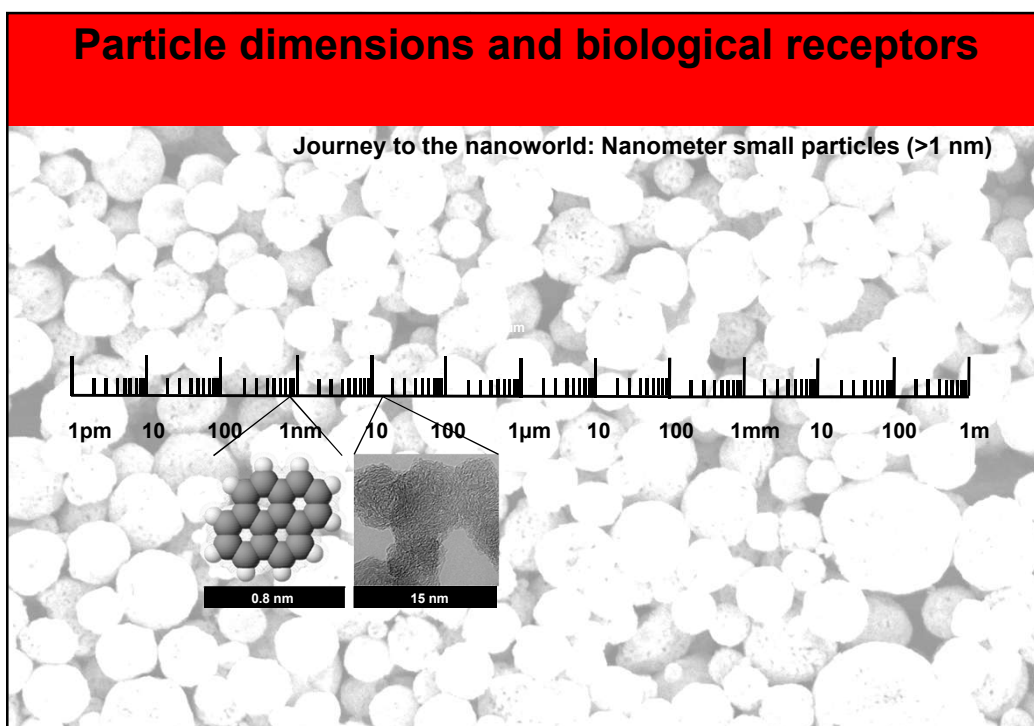
814.318.142.1

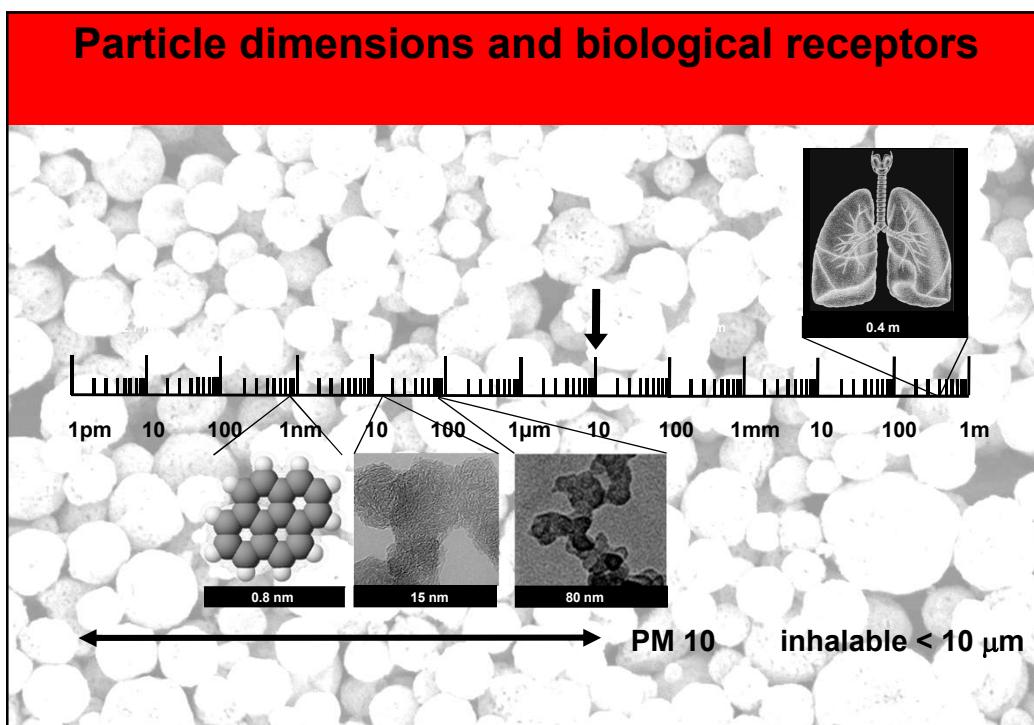
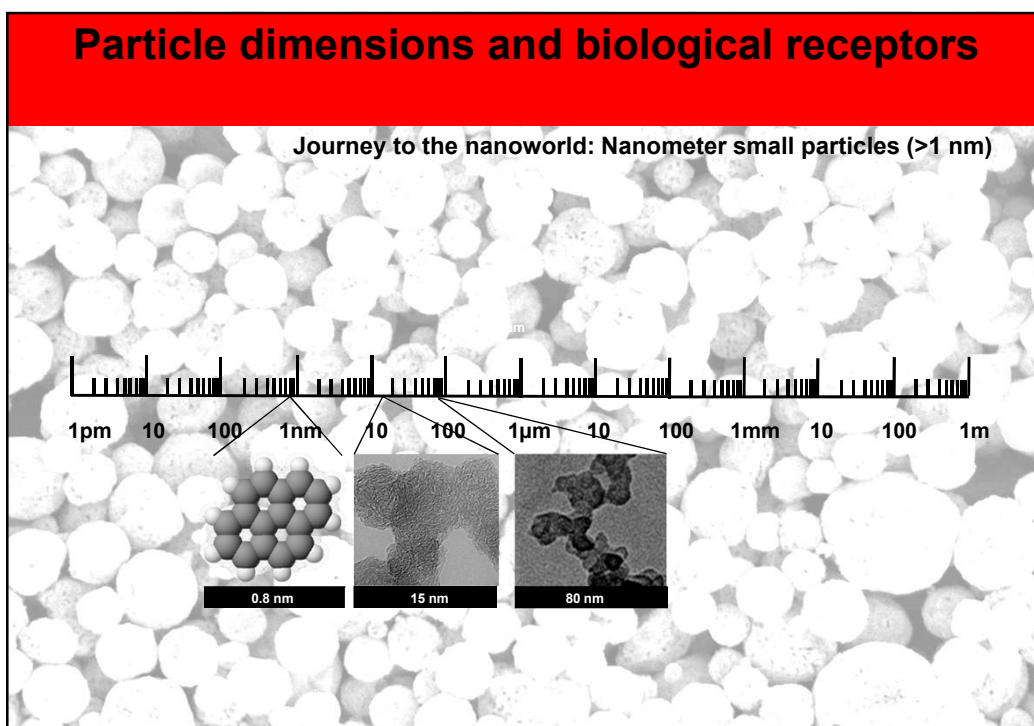
83 Tabelle von krebserzeugenden Stoffen

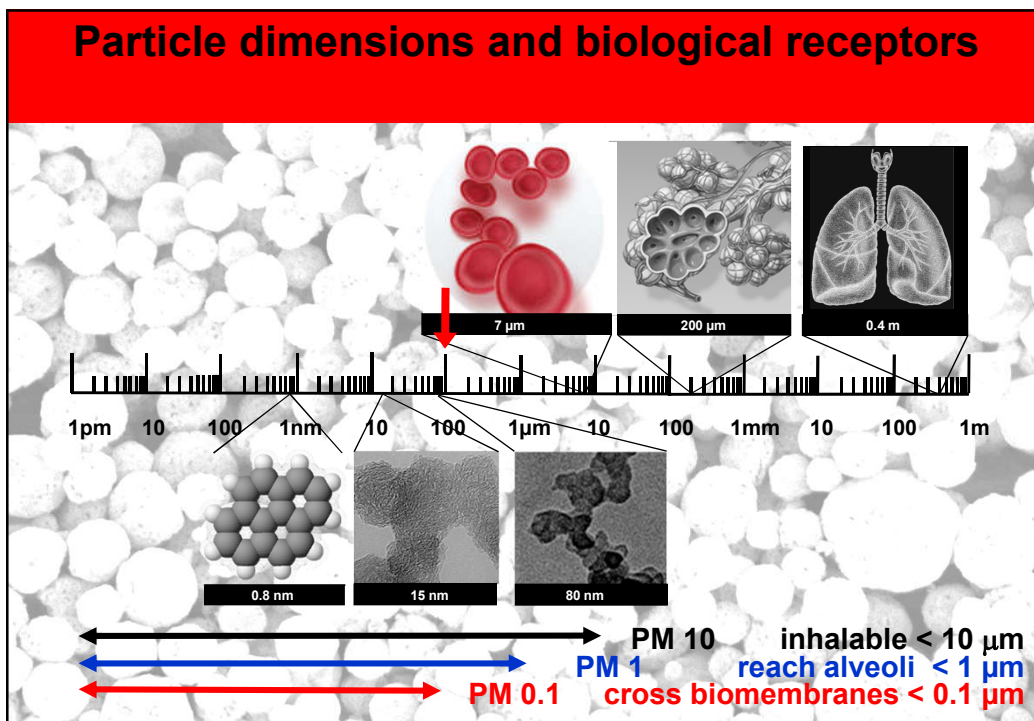
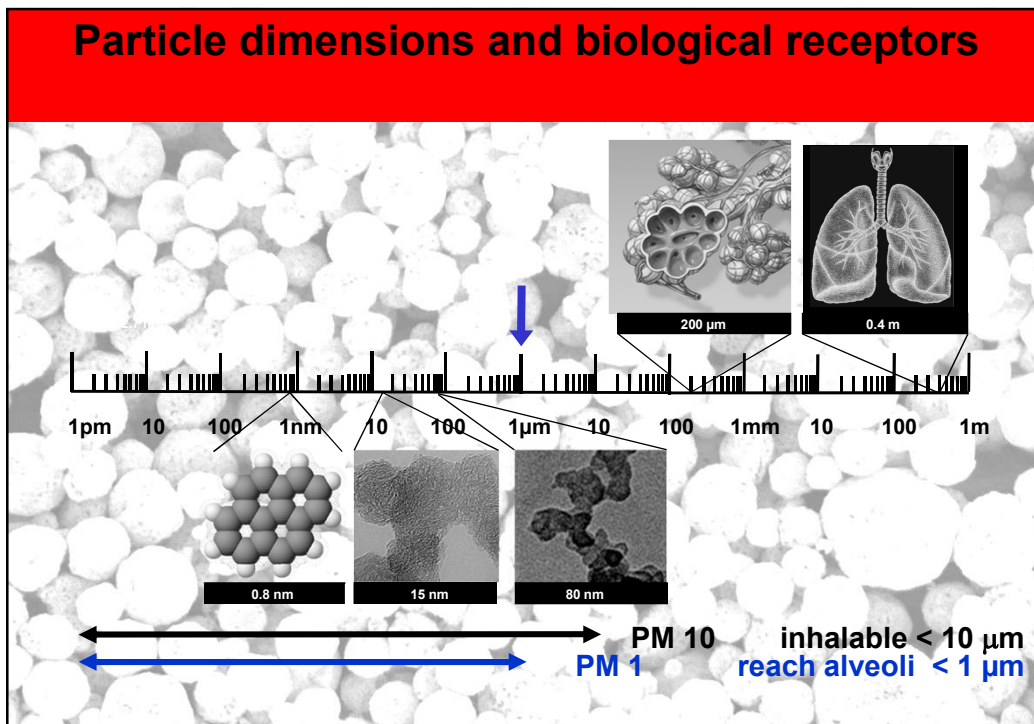
Stoff	Summenformel	Klasse
Benzo(a)pyren	$C_{20}H_{12}$	1
Benzol	C_6H_6	3
Dibenz(a, h)anthracen	$C_{22}H_{14}$	1
1,2-Dibromethan	$C_2H_4Br_2$	3
1,4 Dichlorbenzol	$C_6H_4Cl_2$	3
1,2-Dichlorethan	$C_2H_4Cl_2$	3
Diesels Russ ←		3
Diethylsulfat	$C_4H_{10}O_4S$	2

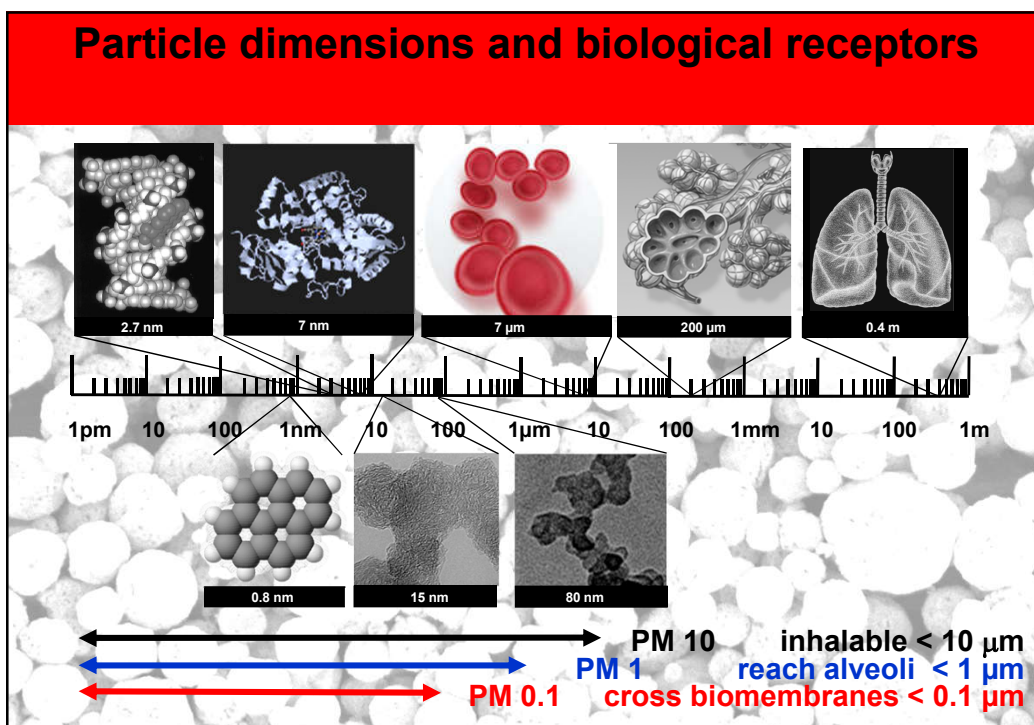
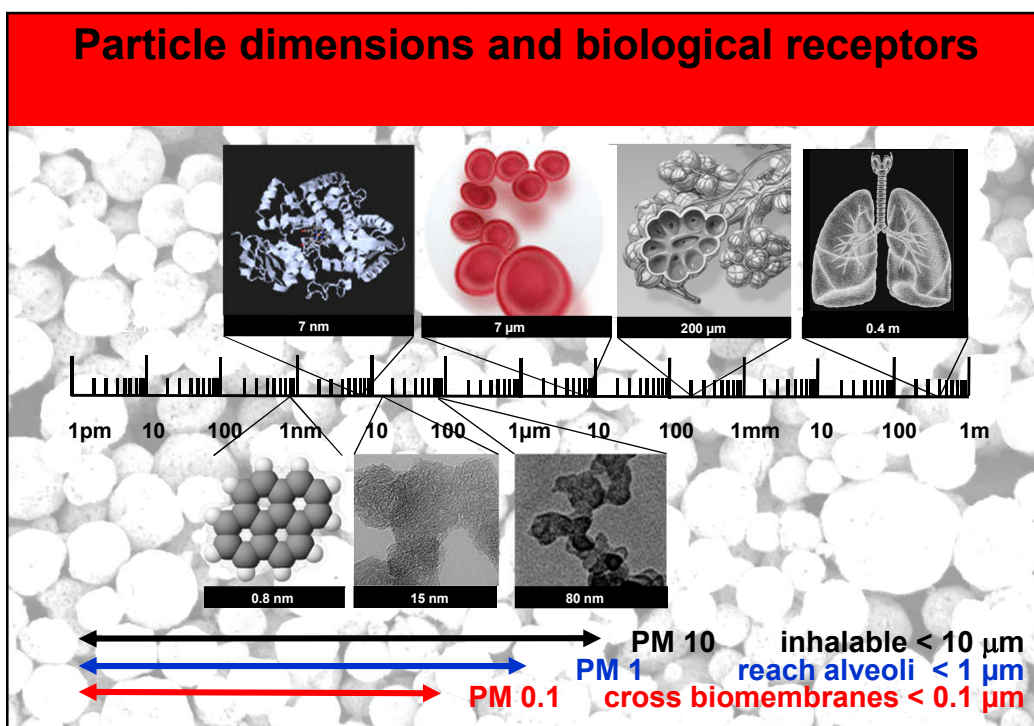
Particle dimensions and biological receptors

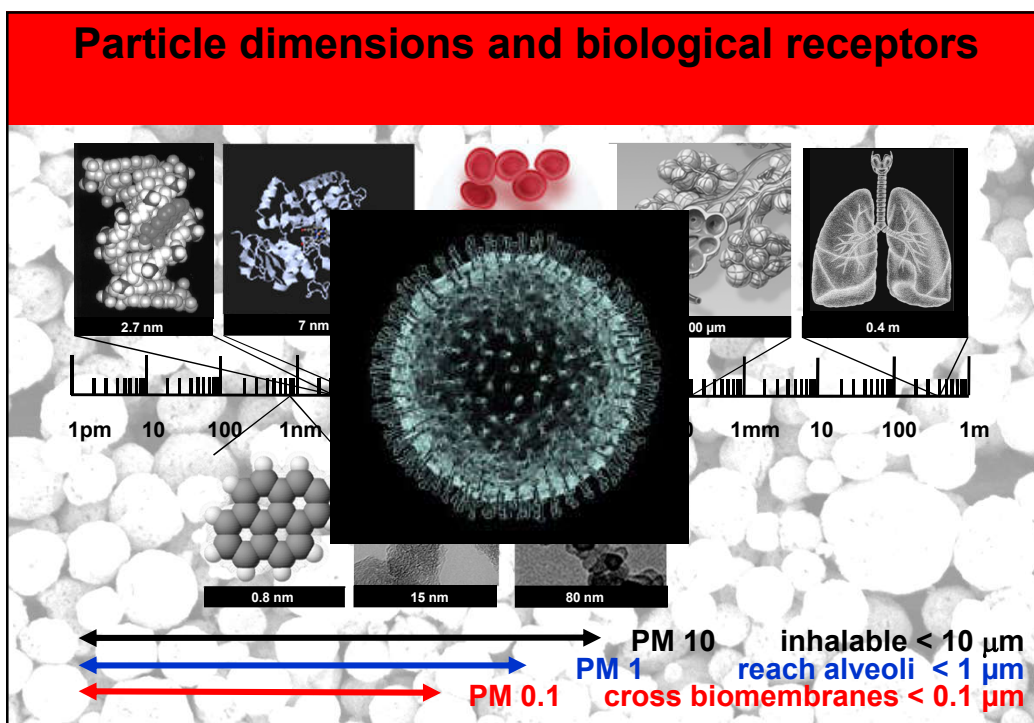
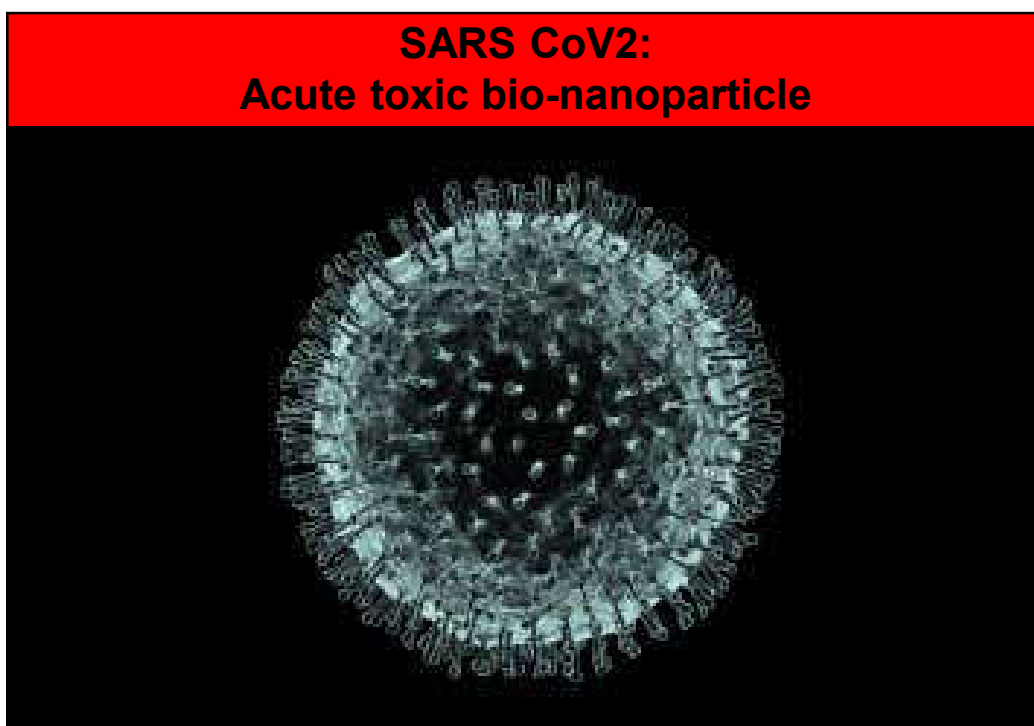


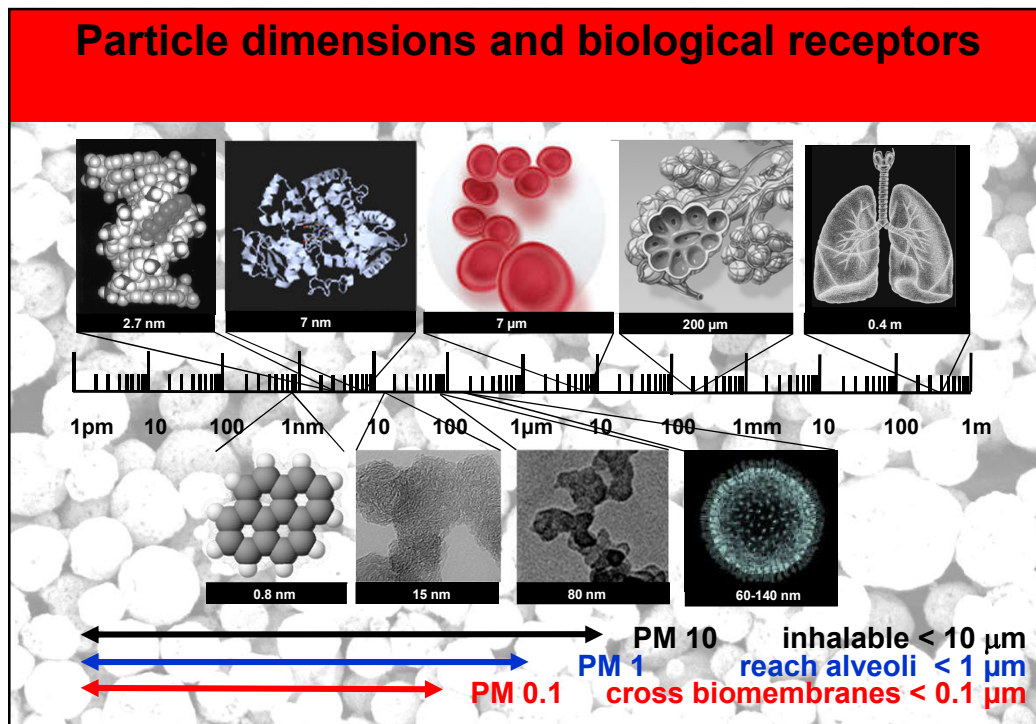


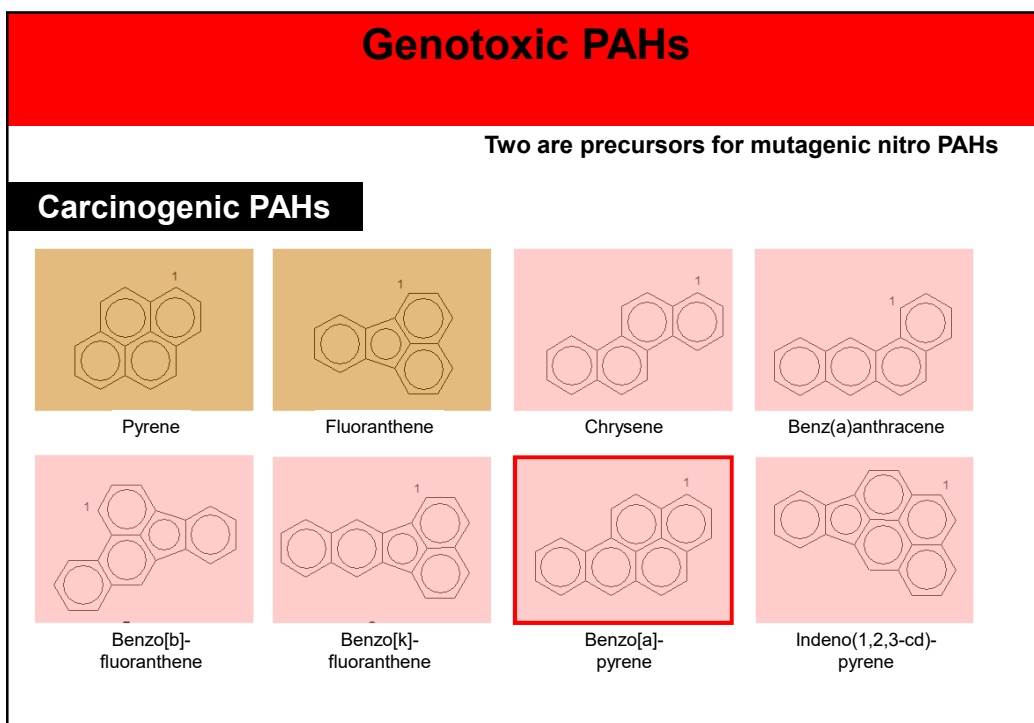
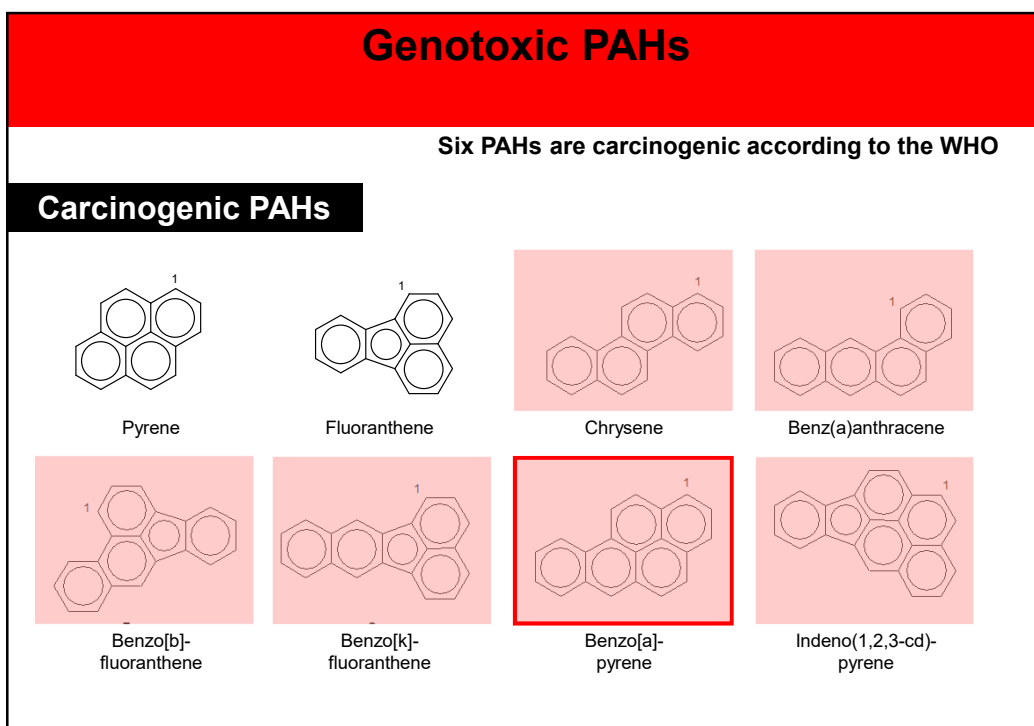






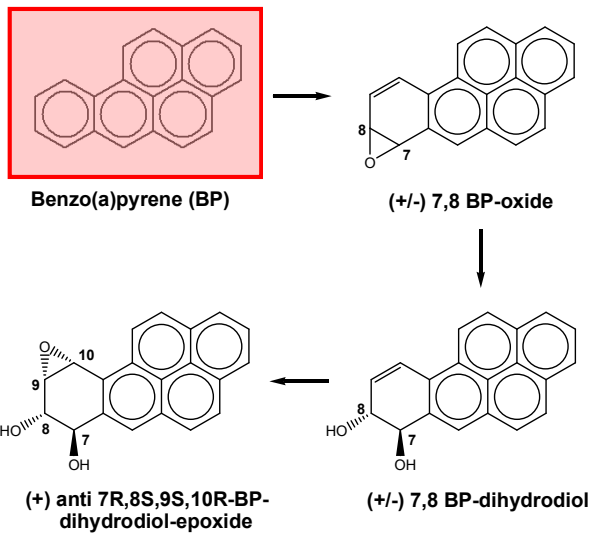






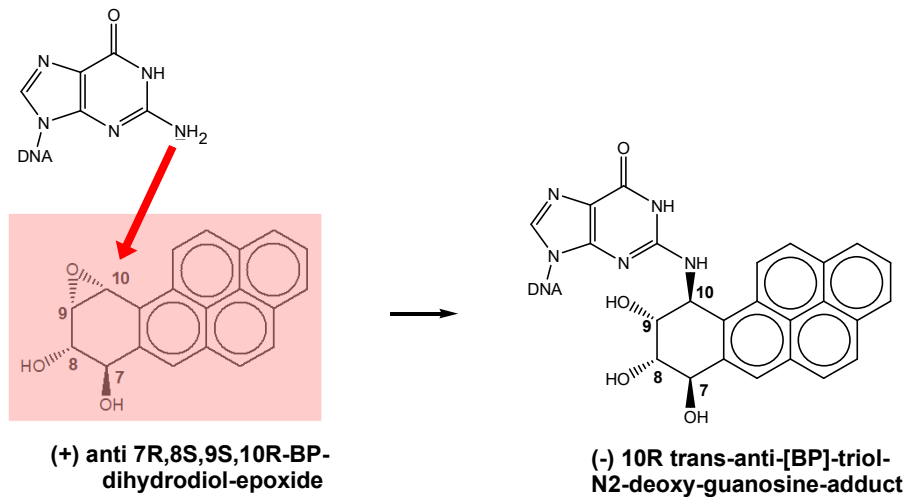
Carcinogenesis of benzo(a)pyrene

Oxidative metabolic activation of benzo(a)pyrene

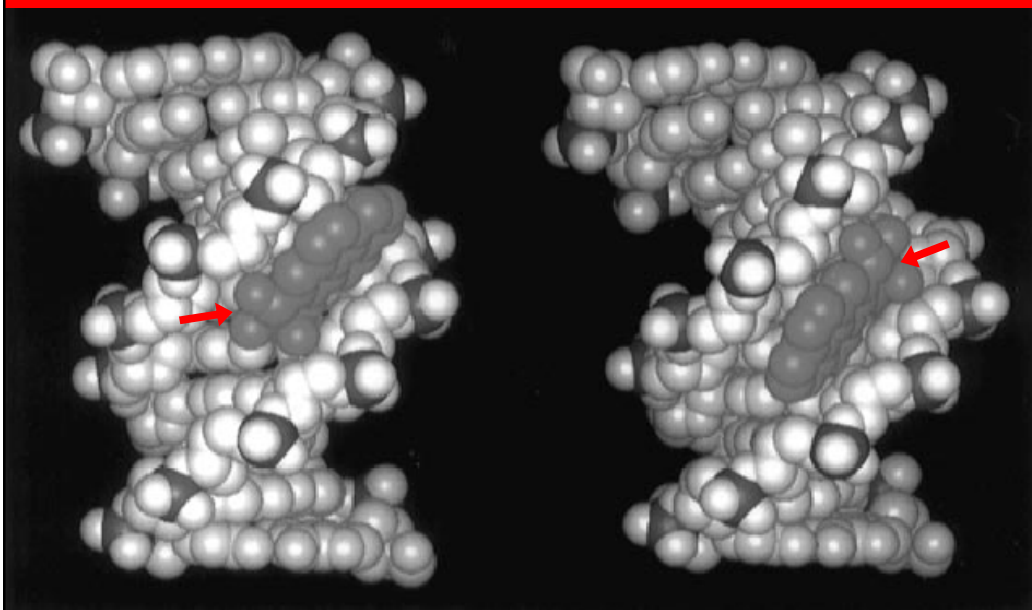


Carcinogenesis of benzo(a)pyrene

Stereoselective formation of benzo(a)pyrene-DNA adducts



Carcinogenesis of benzo(a)pyrene



Diesel soot particles – Trojan horses for genotoxic compounds

Non-treated diesel exhaust is a toxic cocktail



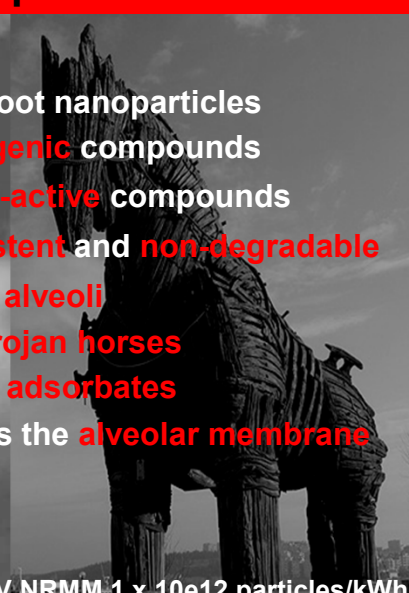
Diesel soot particles – Trojan horses for genotoxic compounds

Non-treated diesel exhaust is a toxic cocktail

Problems

- consists of **billions** of soot nanoparticles
- **carcinogenic** and **mutagenic** compounds
- **hormone-like** and **redox-active** compounds
- soot particles are **persistent** and **non-degradable**
- nanoparticles reach the **alveoli**
- soot particles act like **Trojan horses**
- nanoparticles transport **adsorbates**
- nanoparticles may cross the **alveolar membrane**

EU LDV PN limit 600 billion particles/km, EURO-V NRMM 1 x 10¹² particles/kWh



Diesel soot particles – Trojan horses for genotoxic compounds

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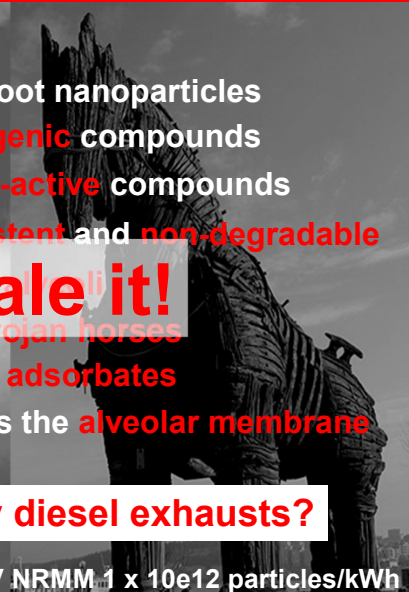
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- nanoparticles may cross the **alveolar membrane**

Do not inhale it!

Do catalytic DPFs detoxify diesel exhausts?

EU LDV PN limit 600 billion particles/km, Euro-V NRMM 1 x 10¹² particles/kWh

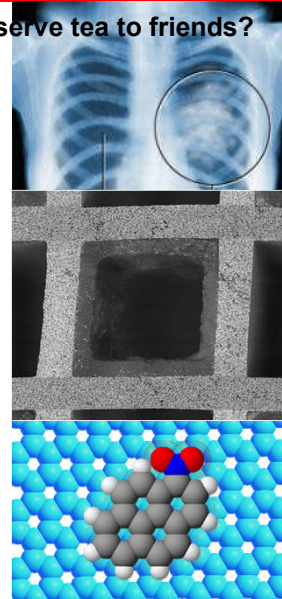


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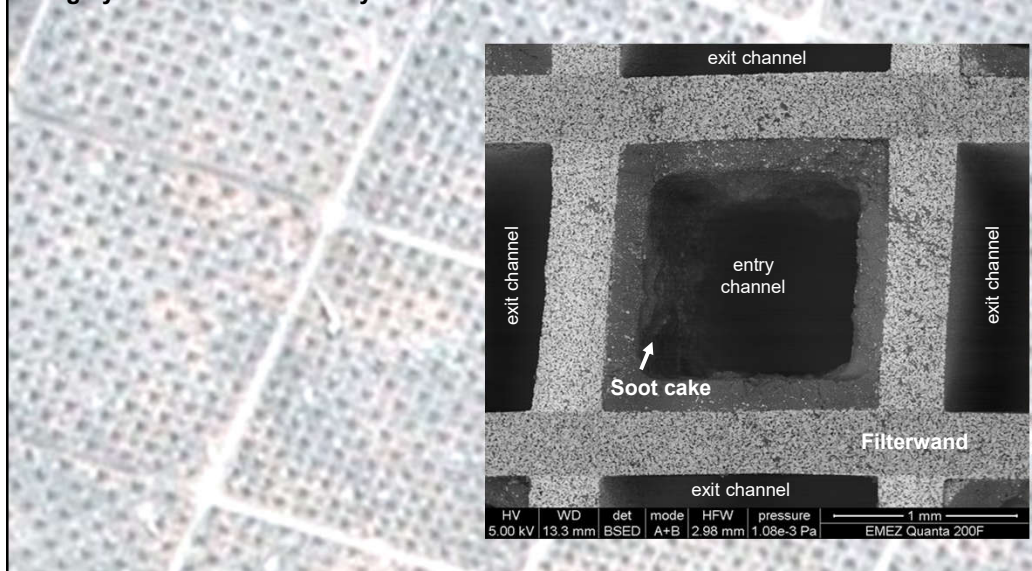
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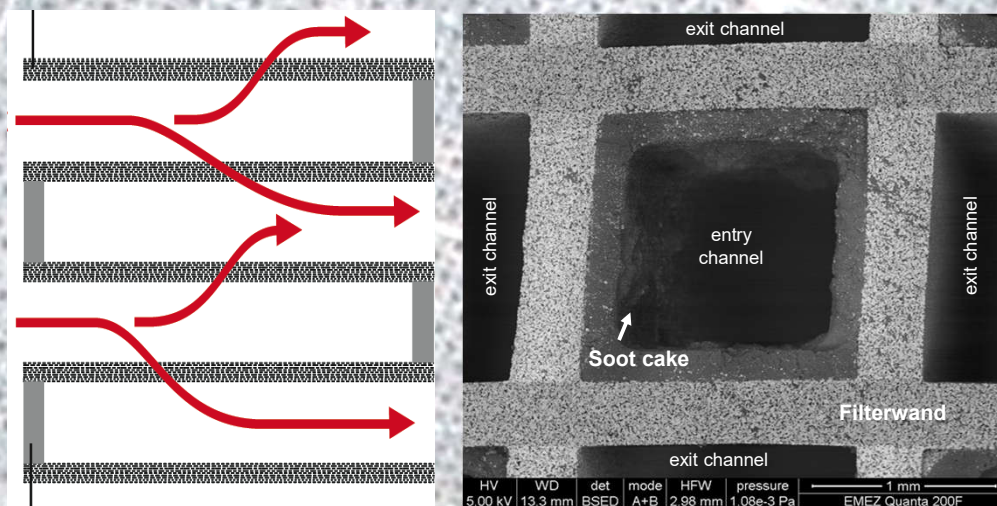
Scheme of wall-flow filters

Highly efficient filtration only in closed filters where exhausts have to cross the wall



Scheme of wall-flow filters

Highly efficient filtration only in closed filters where exhausts have to cross the wall



The visible effect of particle filters

7 m³ exhaust collected (2 minutes operation of an Euro-3 engine (6.1 L, 105 kW))

450x10¹² particles
150 ng benzo(a)pyrene

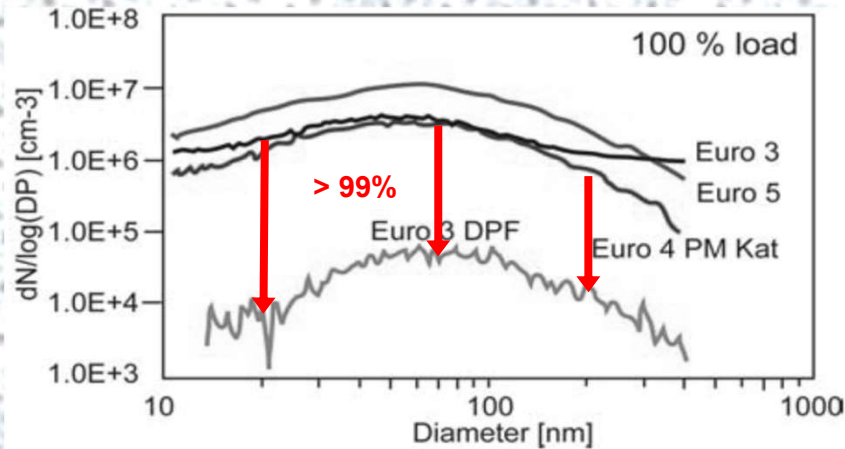


0.1x10¹² particles
7 ng benzo(a)pyrene



The invisible effect of particle filters

We have to count particles not weighing them!
The world needs a number-based particle legislation for nanoparticles!



A. Mayer et al., SAE 2007-01-1112

Nanoparticle emission limits of LDV vehicles and non-road mobile machinery in Europe

You know one million. You know one billion. What's next?

One million (US) = 1'000'000 = 1×10^6

One billion (US) = 1'000'000'000 = 1×10^9

Nanoparticle emission limits of LDV vehicles and non-road mobile machinery in Europe

You know one million. You know one billion. What's next?

One million (US) = 1'000'000 = 1×10^6

One billion (US) = 1'000'000'000 = 1×10^9

One trillion (US) = 1'000'000'000'000 = 1×10^{12}

600 billion (US) = 6×10^{11} particles/km is the legal PN limit for:

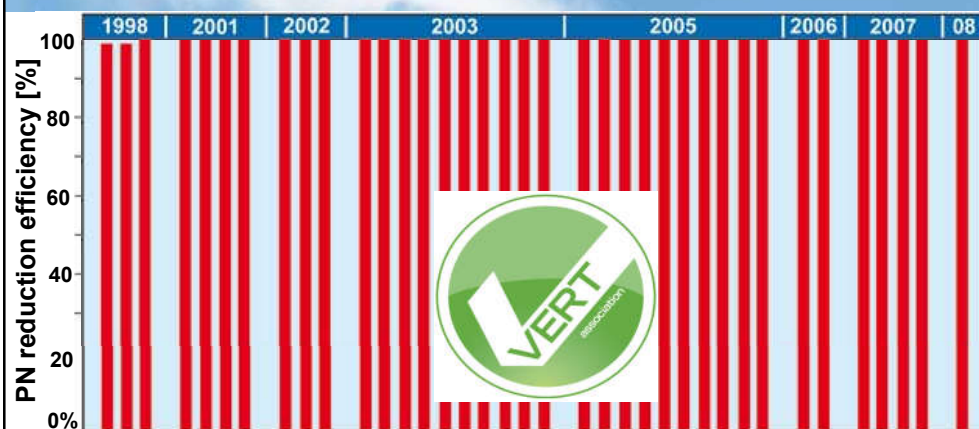
- Euro-5/6 diesel passenger cars (since 2011)
- Euro-6 gasoline direct injection passenger cars (since 2018)

One trillion (US) = 1×10^{12} particles/kWh is the legal PN limit for:

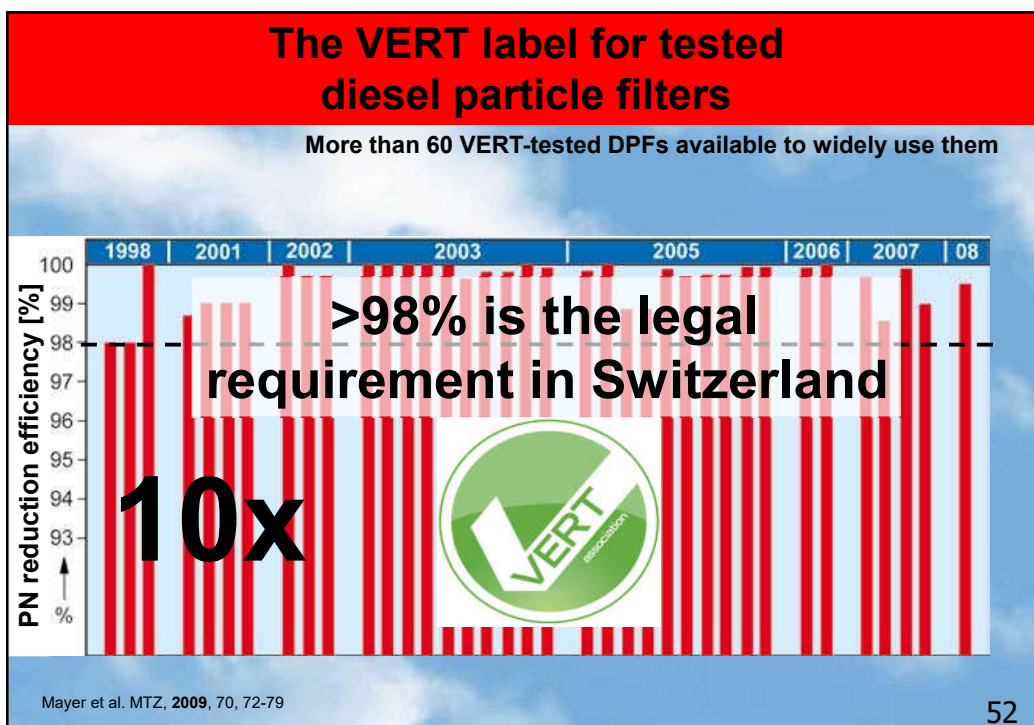
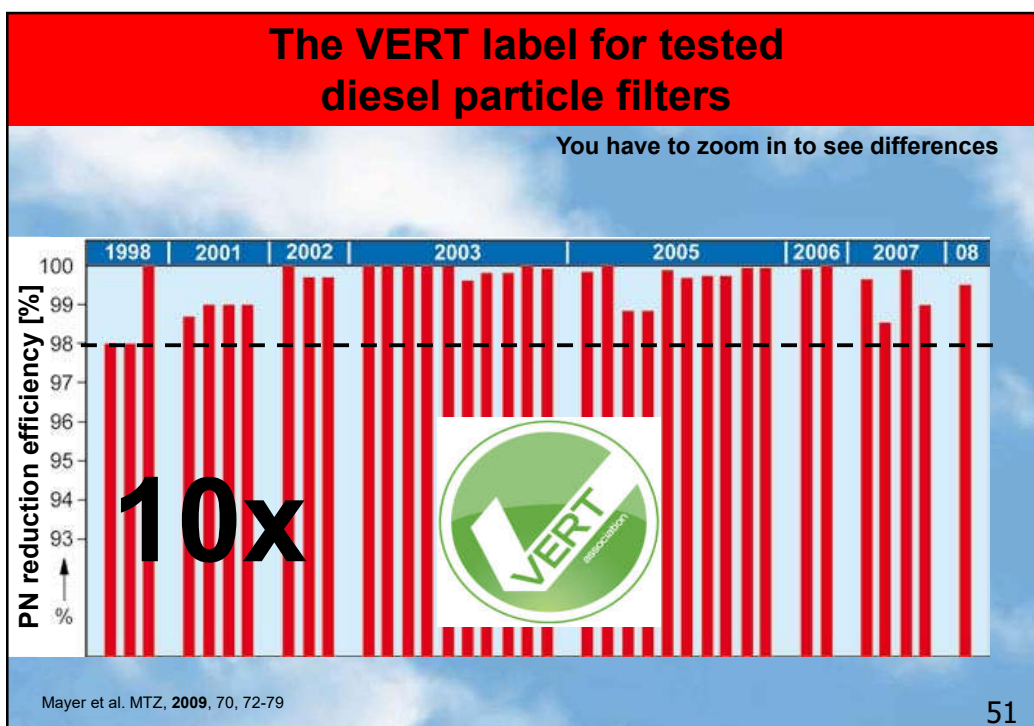
- Euro-V non-road mobile machinery (since 2018)

The VERT label for tested diesel particle filters

More than 60 VERT-tested DPFs available to widely use them



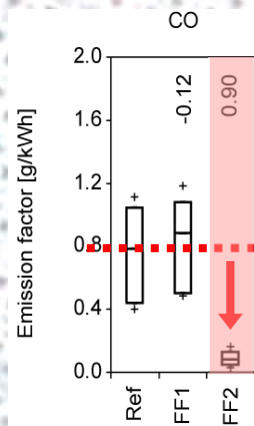
Mayer et al. MTZ, 2009, 70, 72-79



Low- & high-oxidation potential DPFs

We have identified 2 filter families, one converts CO the other doesn't!

Carbon monoxide



- Ref: Engine-out
- FF1: Low oxidation potential (n=6)
- FF2: High oxidation potential (n=8)

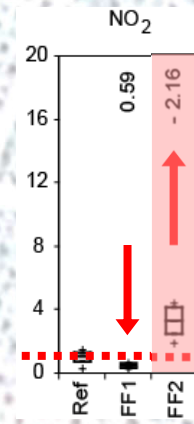
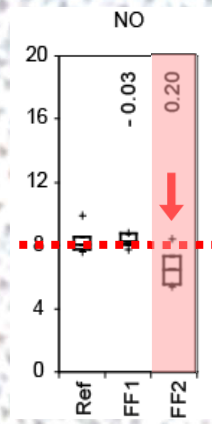
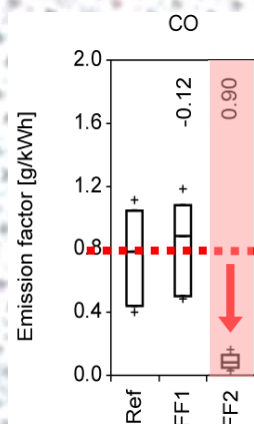
Heeb et al. ES&T, 2008, 42, 3773-3779

Heeb et al. ES&T, 2010, 442, 1078-1084

Low- & high-oxidation potential DPFs

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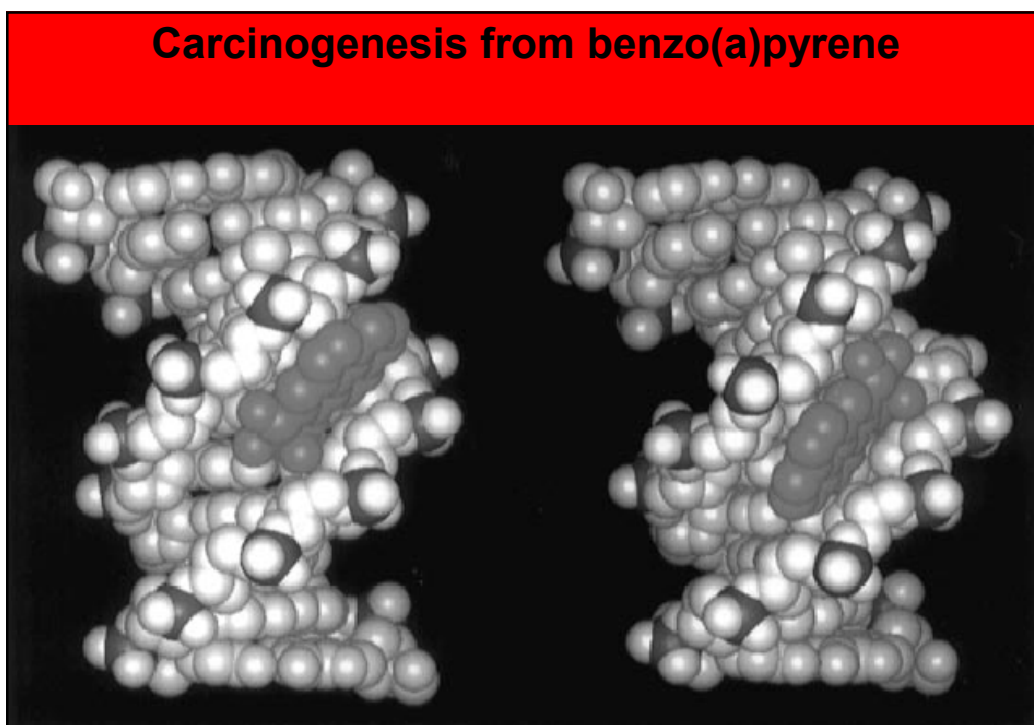
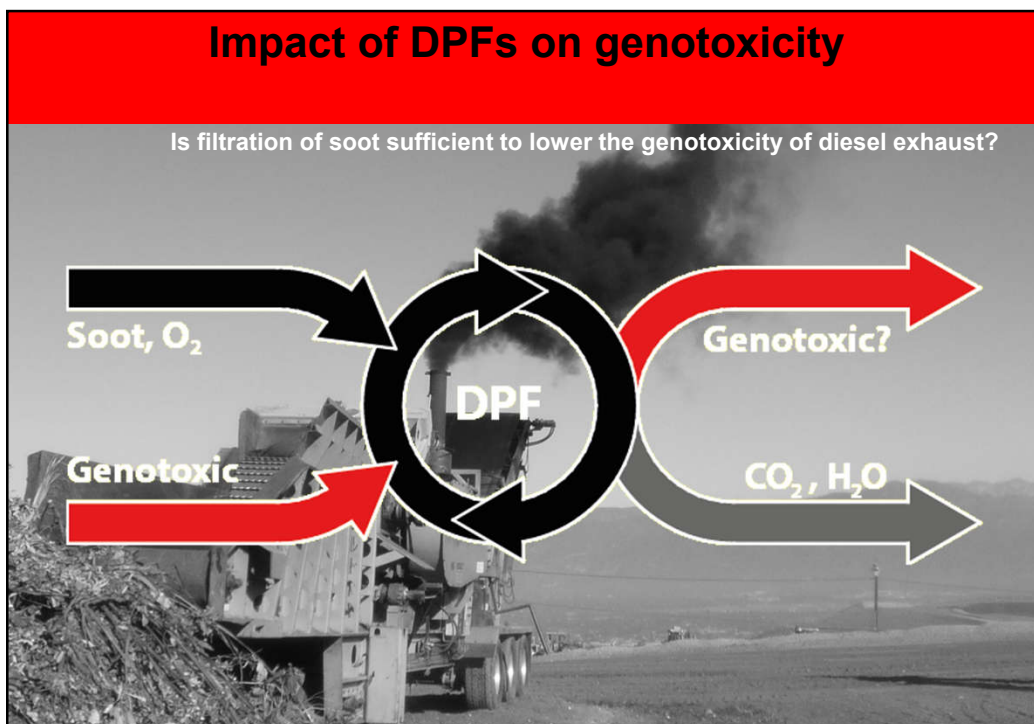
CO, NO, NO₂



Secondary NO₂ emission

Heeb et al. ES&T, 2008, 42, 3773-3779

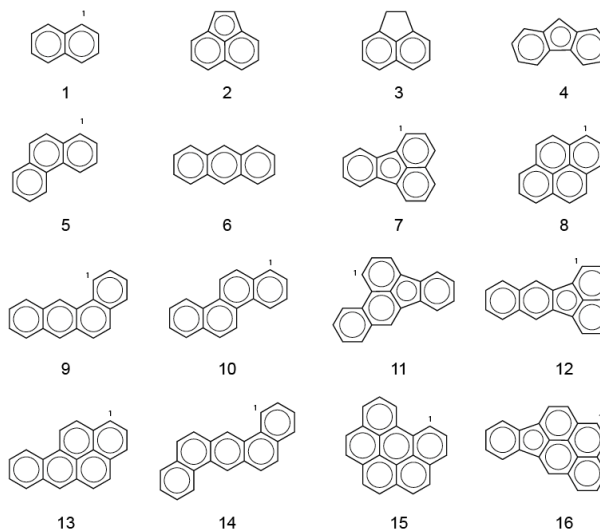
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Polycyclic aromatic hydrocarbons

PAHs - a diverse class of compounds with variable physicochemical properties

2- to 6-ring PAHs

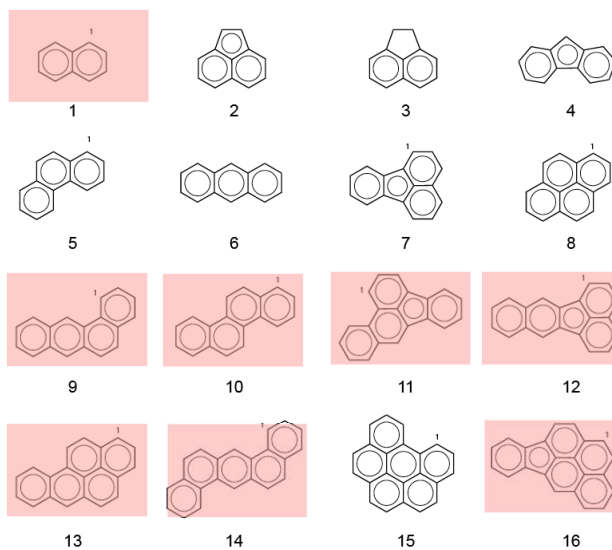


Polycyclic aromatic hydrocarbons

PAHs - a diverse class of compounds with variable physicochemical properties

2- to 6-ring PAHs

some PAHs are
genotoxic



Polycyclic aromatic hydrocarbons

PAHs - a diverse class of compounds with variable physicochemical properties

2- to 6-ring PAHs

some PAHs are genotoxic

all PAHs are potential nitro-PAH precursors

several nitro-PAHs are strong mutagens

PAH penetration of a non-catalyzed DPF

Non-catalyzed filters are as efficient for soot. How about genotoxic compounds?

Non-catalyzed DPFs:
Accumulate soot (>98%)

Can PAHs penetrate soot loaded DPFs?

Do DPFs remove genotoxic compounds a.m.a.p?

Do DPFs support a formation of toxic secondary pollutants like nitro-PAHs?

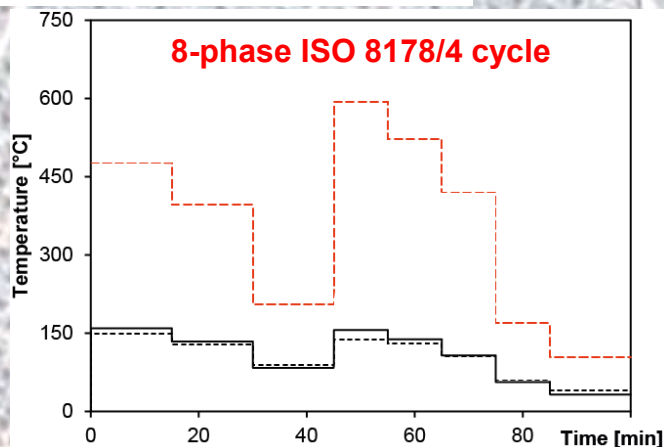
PAH penetration of a non-catalyzed DPF

Can PAHs penetrate non-catalyzed filters if operated $<200^{\circ}\text{C}$?

We studied 2 cellulose-based filters, a new and a soot-loaded filter (>2000 h road application)

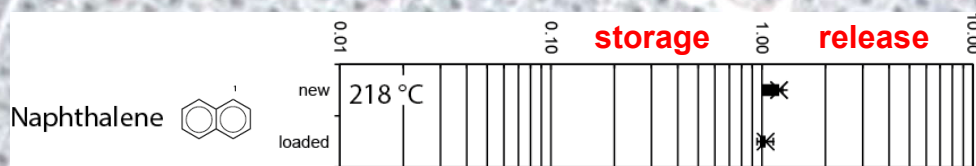
engine-out \rightarrow

before DPF \rightarrow
after DPF \rightarrow



PAH penetration of a non-catalyzed DPF

Non-catalyzed filter operated $<200^{\circ}\text{C}$ do accumulate soot

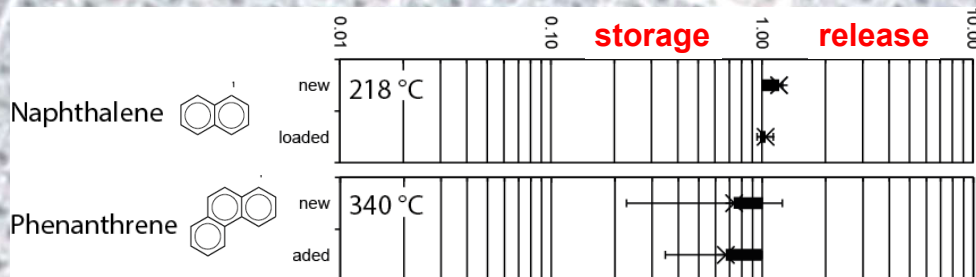


- no retention of naphthalene in a new and a soot-loaded DPF

- naphthalene is too volatile, it escapes even from a cold filter ($<200^{\circ}\text{C}$)

PAH penetration of a non-catalyzed DPF

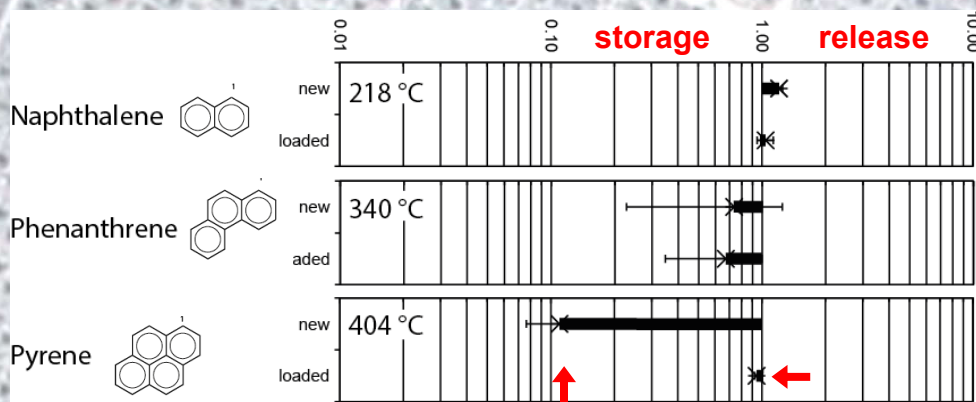
Non-catalyzed filter operated <200 °C do accumulate soot and some hydrocarbons



- about **30% retention**, both in a new and a soot-loaded DPF
- phenanthrene is partly stored in both filters (<200 °C).

PAH penetration of a non-catalyzed DPF

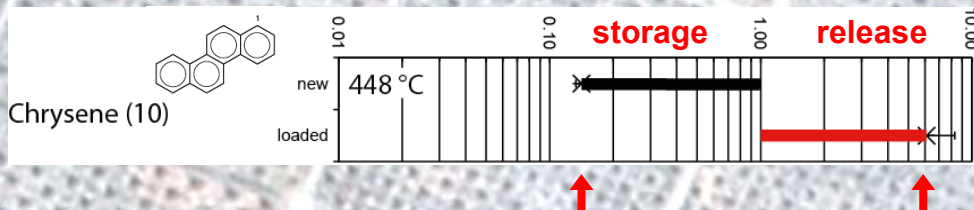
Non-catalyzed filter operated <200 °C do accumulate soot and some hydrocarbons



- **90% pyrene is retained** in a new, **only 5%** in a soot-loaded DPF

PAH store-and-release in a non-catalyzed DPF

Non-catalyzed filter operated <200 °C do accumulate soot and some hydrocarbons

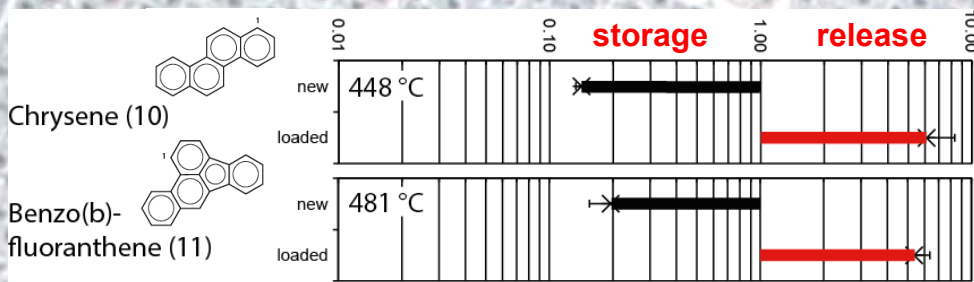


- **85% retention** in the new DPF

- **6x higher emissions** from the soot-loaded DPF

PAH store-and-release in a non-catalyzed DPF

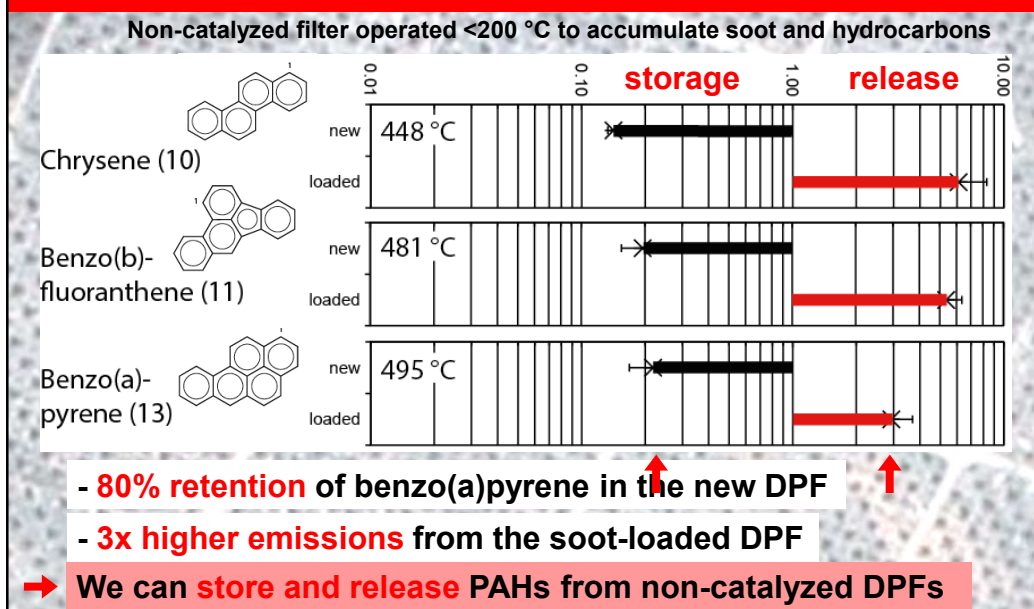
Non-catalyzed filter operated <200 °C do accumulate soot and hydrocarbons



- **80% benzo(b)fluoranthene is retained** in the new DPF

- **5x higher emissions** from the soot-loaded DPF

PAH store-and-release in a non-catalyzed DPF



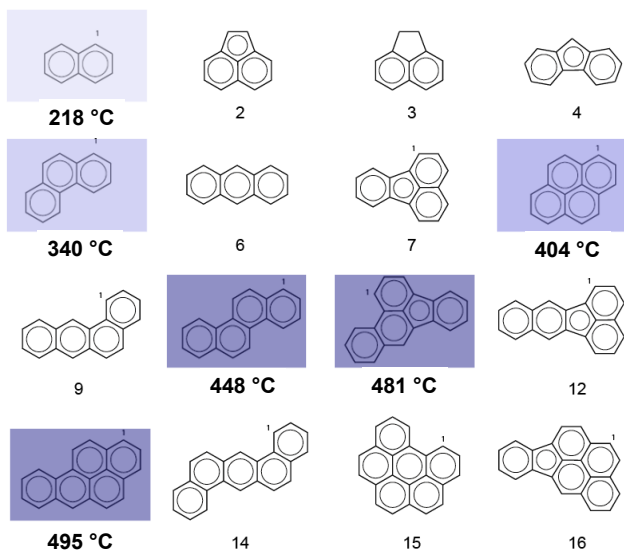
Polycyclic aromatic hydrocarbons

PAHs - a diverse class of compounds with variable physicochemical properties

2- to 6-ring PAHs

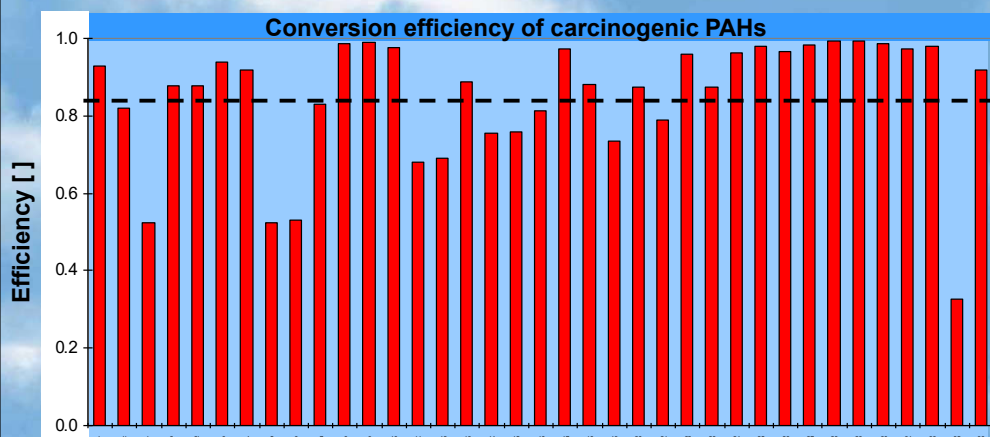
- **Volatile PAHs** can penetrate DPFs both, new and soot-loaded ones

- **Semi-volatile PAHs** are stored in new, but can be released again from soot-loaded DPFs



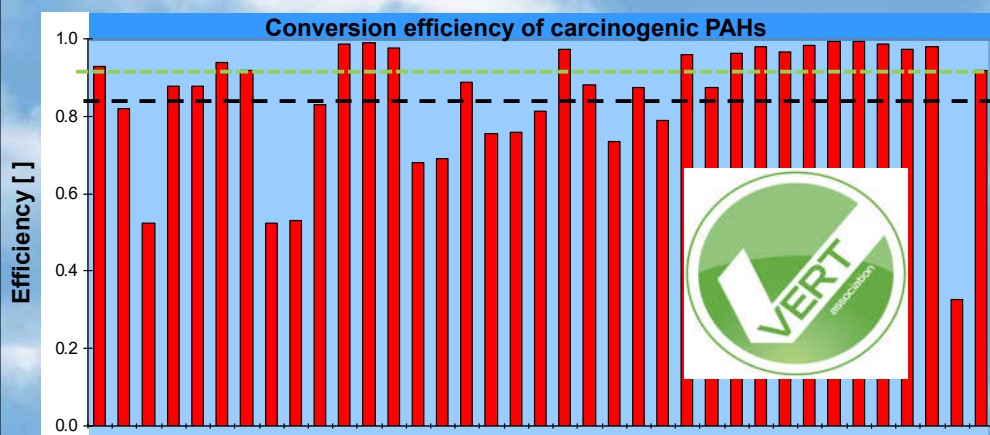
Adverse health effects of diesel exhaust

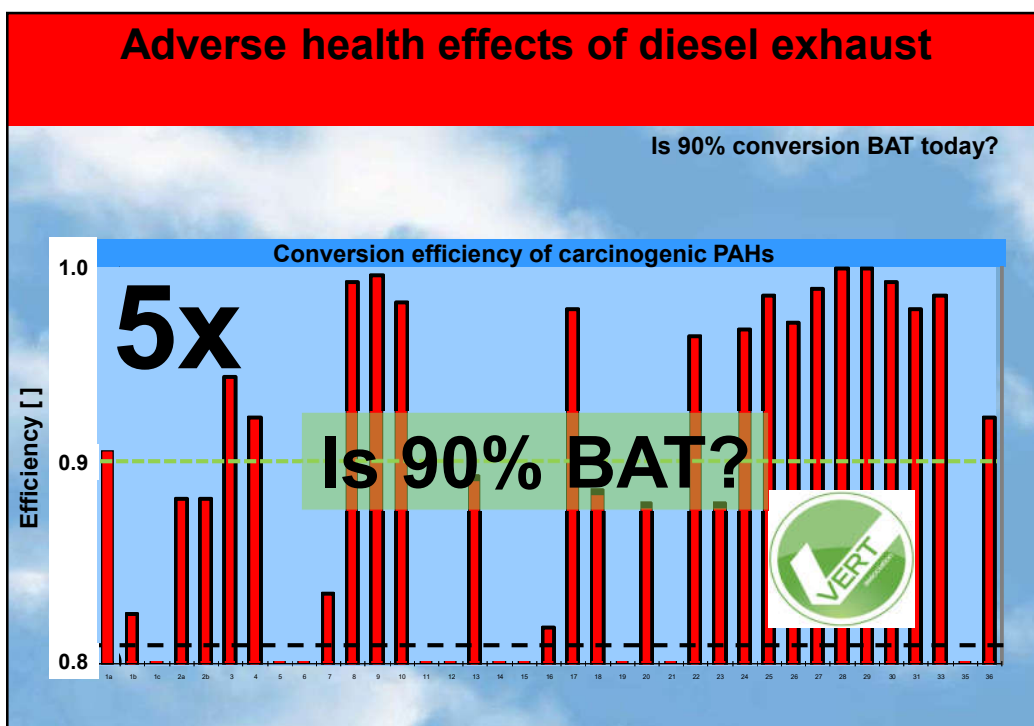
VERT-tested catalytic DPFs convert carcinogenic PAHs (on average 85%)



Adverse health effects of diesel exhaust

What is BAT today?





Adverse health effects of diesel exhaust

Problem: Genotoxicity

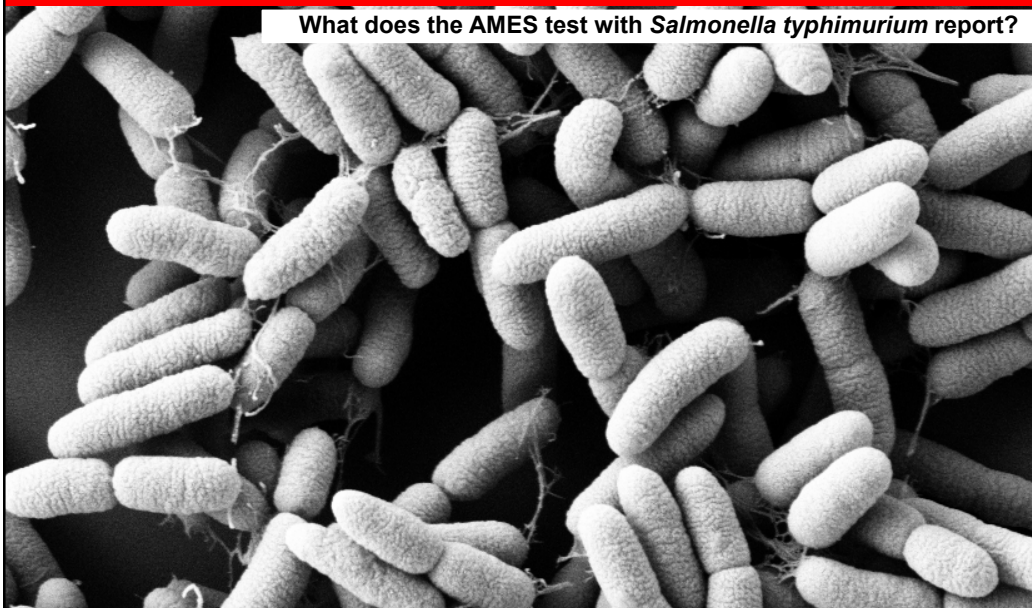
- Non-filtered diesel exhaust is genotoxic
- Filtration as such is not sufficient to remove genotoxic compounds
- Efficient catalysts are needed to convert genotoxic compounds

Is
90%
BAT?

DPFs are BAT to lower the genotoxicity of diesel exhaust, but some are considerably better than others!

Impact of DPFs on genotoxicity

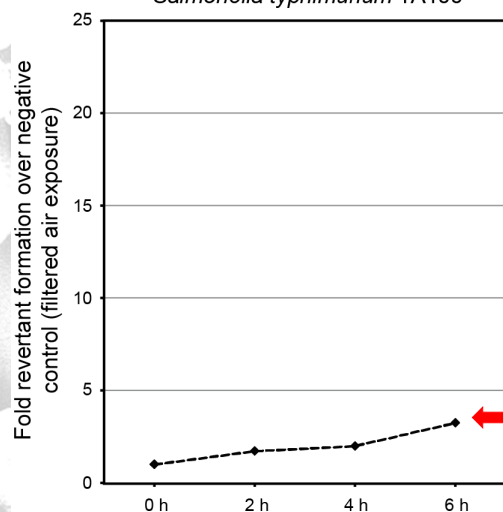
What does the AMES test with *Salmonella typhimurium* report?



Impact of DPFs on genotoxicity

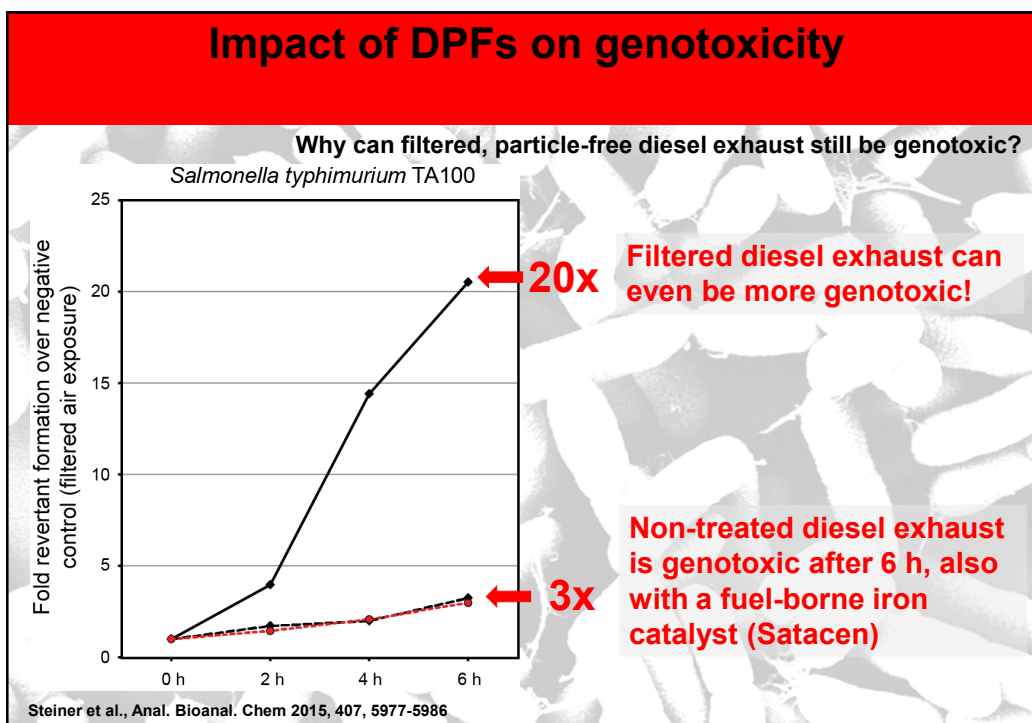
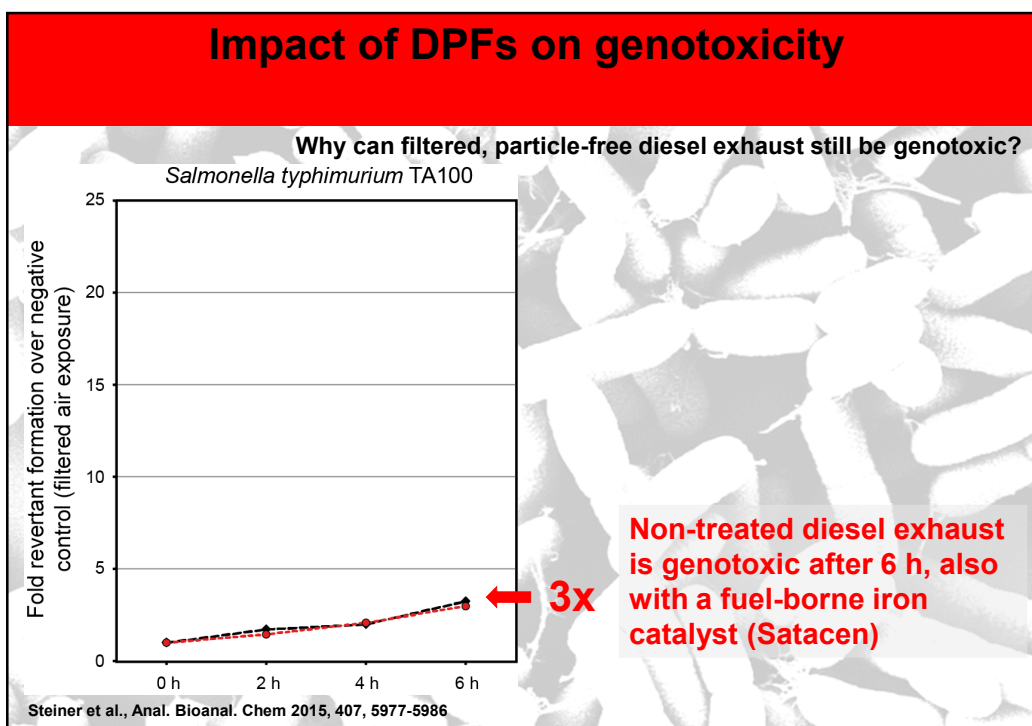
Why can filtered, particle-free diesel exhaust still be genotoxic?

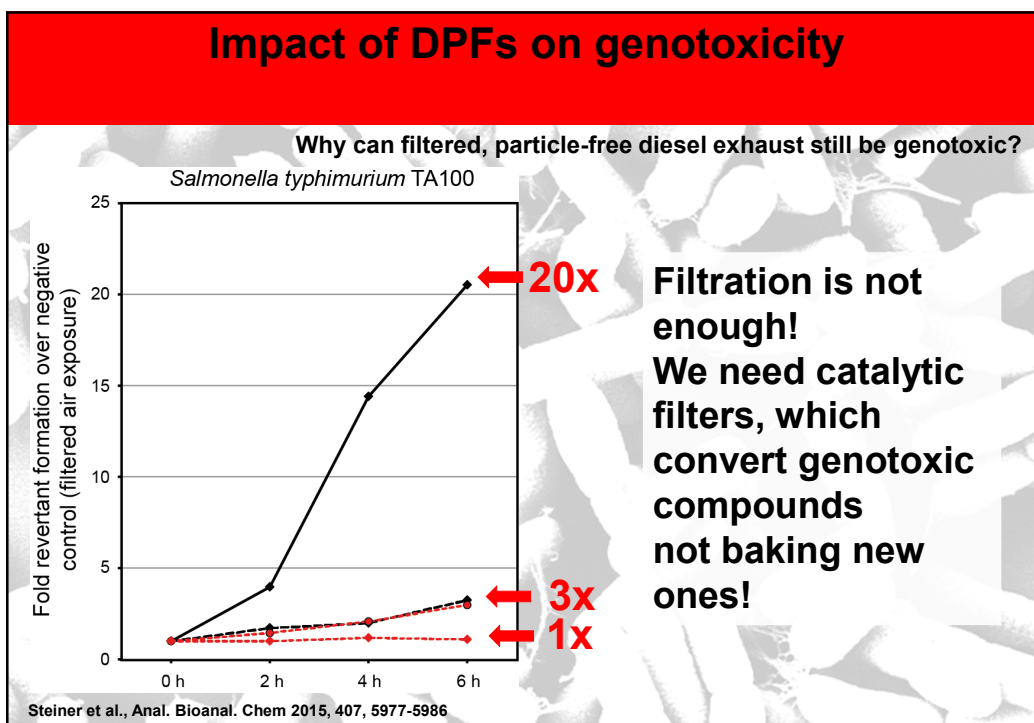
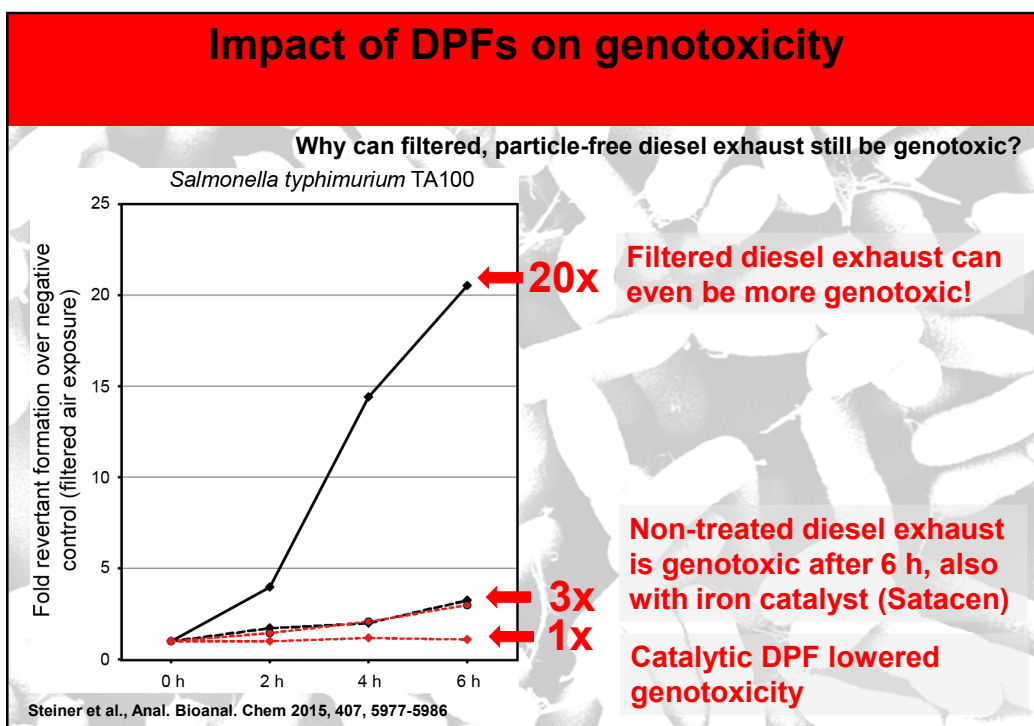
Salmonella typhimurium TA100



Non-treated diesel exhaust
is genotoxic after 6 h

Steiner et al., Anal. Bioanal. Chem 2015, 407, 5977-5986





Secondary emissions from emission control devices and their impact on occupational health and safety

Would you run a diesel engine in your living room and serve tea to friends?

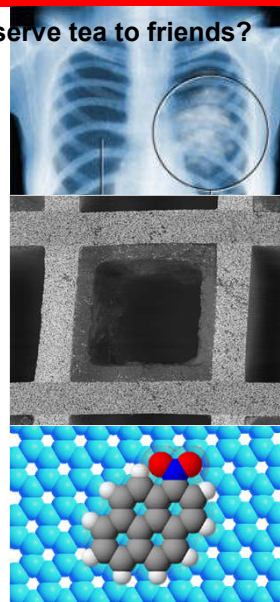
Outline

- **A success story to begin with**
 - VERT-approved DPFs in the NEAT tunnels
- **Risks and health impact of diesel exhausts**
 - What should you know about it?
- **Catalytic diesel particle filters**
 - Do DPFs detoxify diesel exhausts?
- **Secondary emissions of emission control devices**
 - How to avoid or manage them?



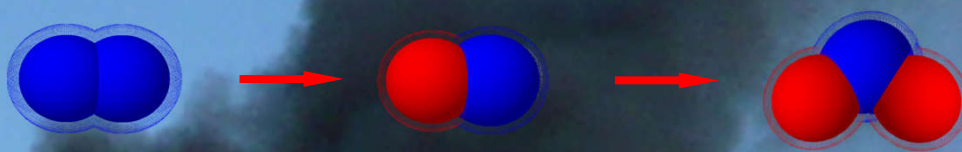
Empa

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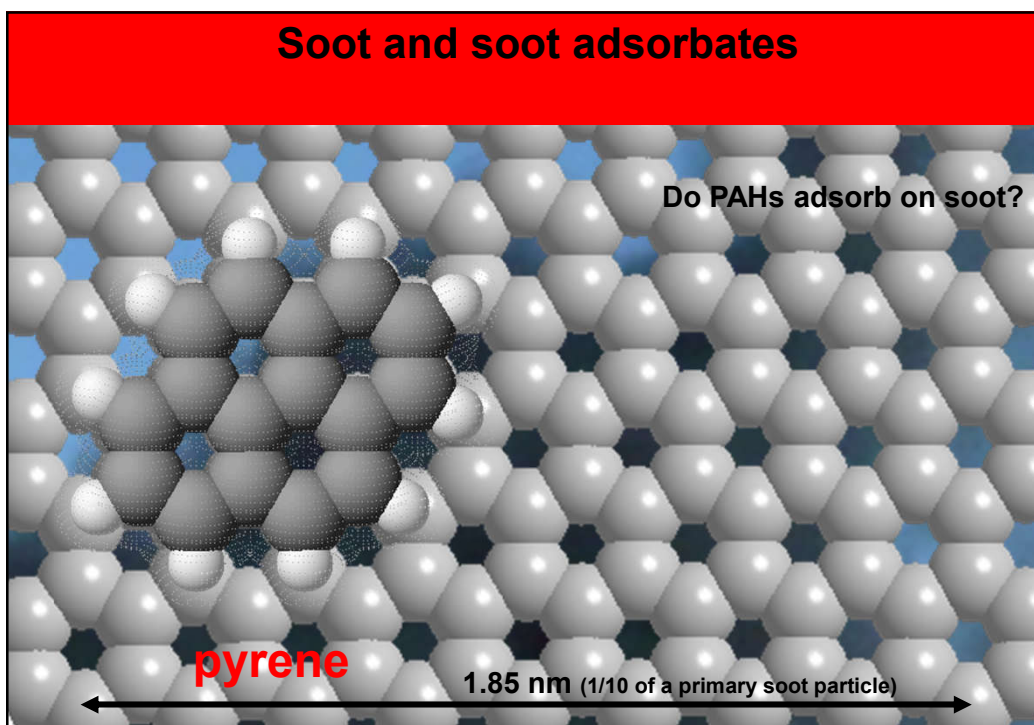
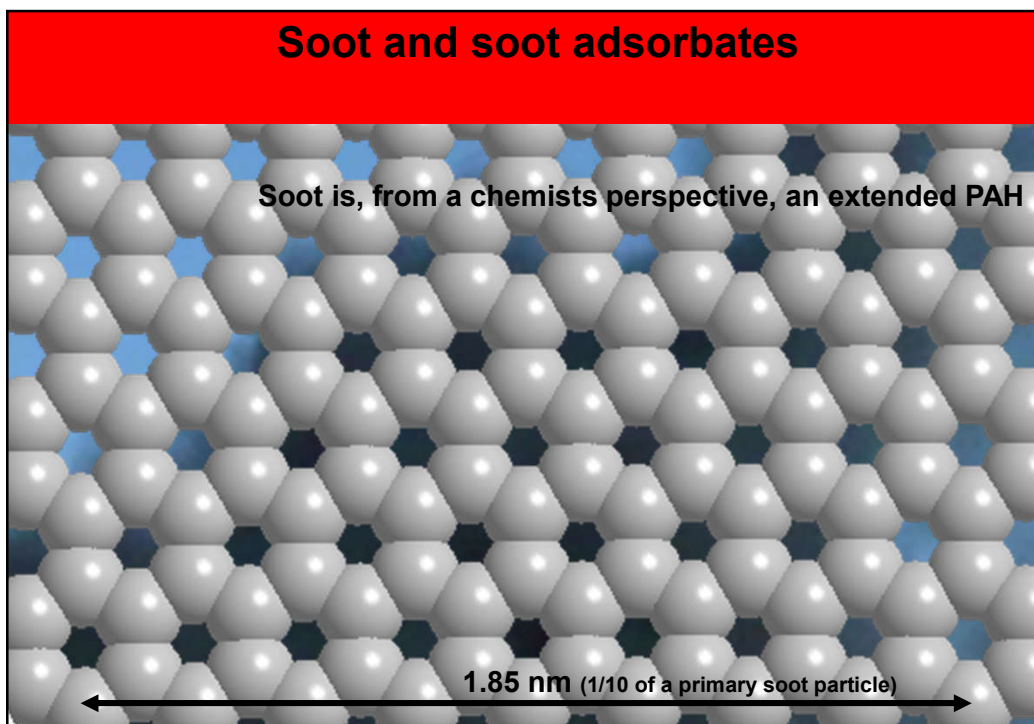
Nitration chemistry in non-catalyzed DPFs

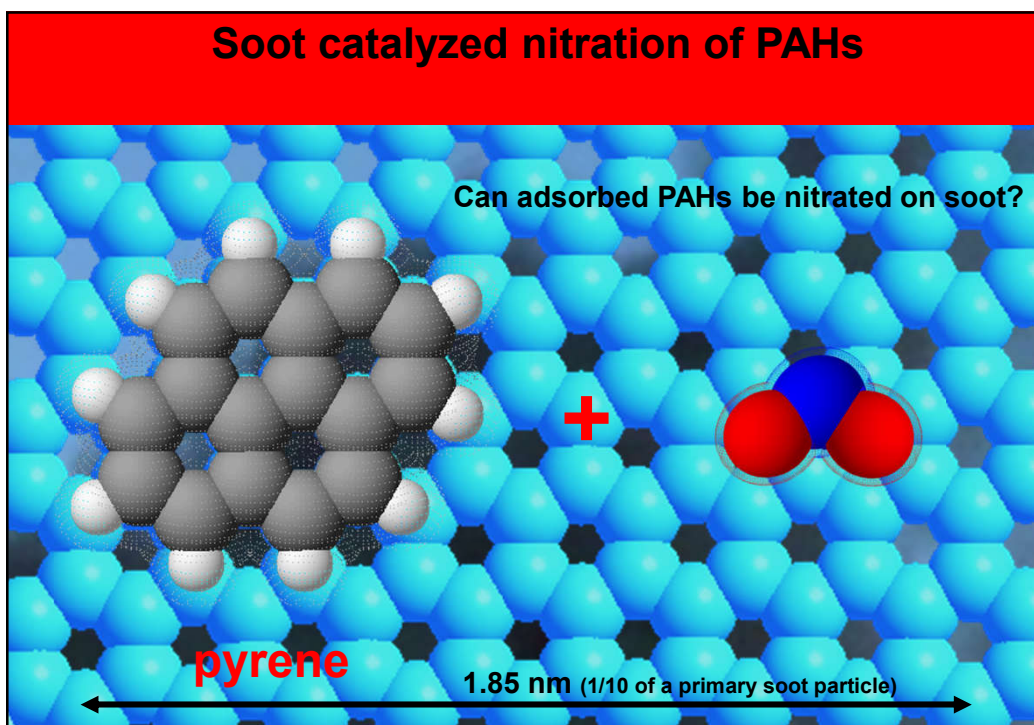
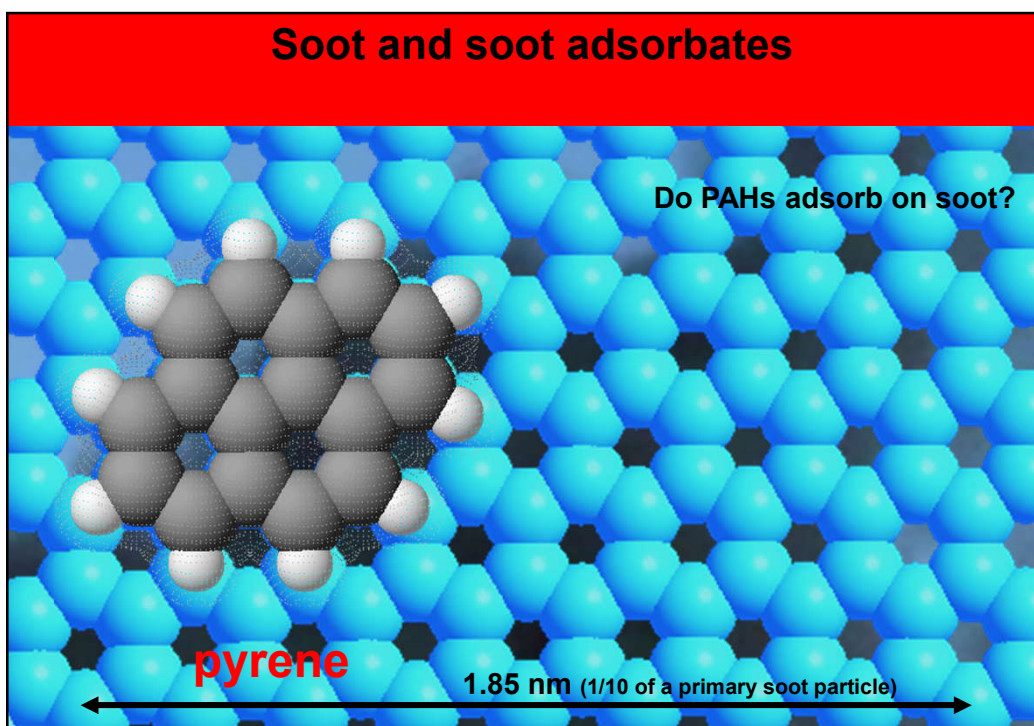
Does exposure of soot and adsorbates to NO_x alter exhaust toxicity?

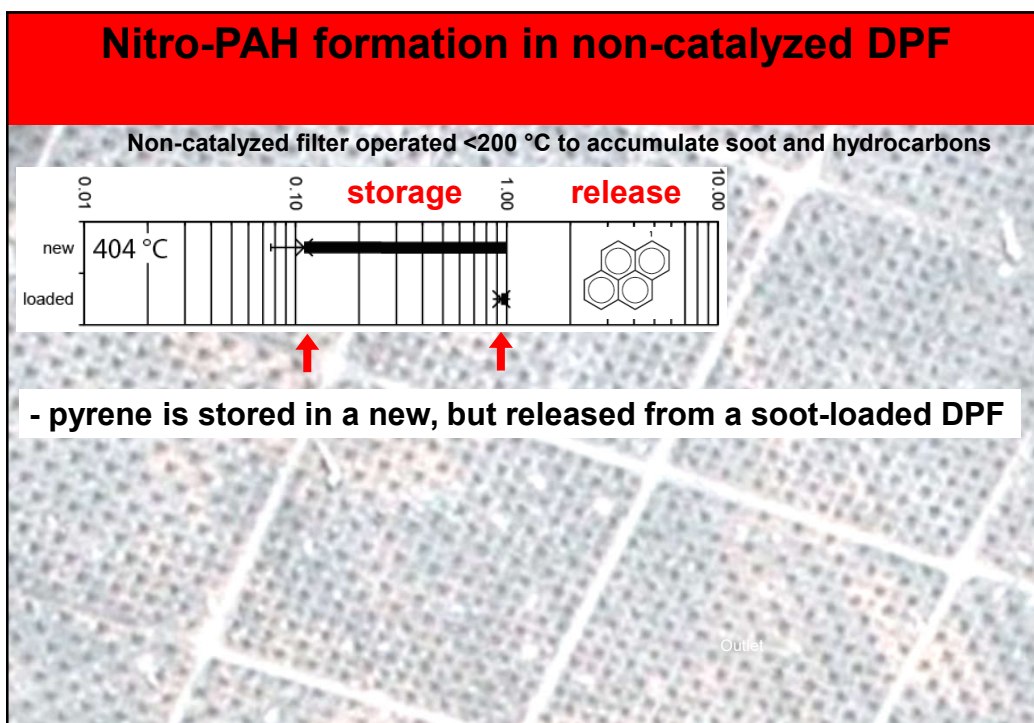
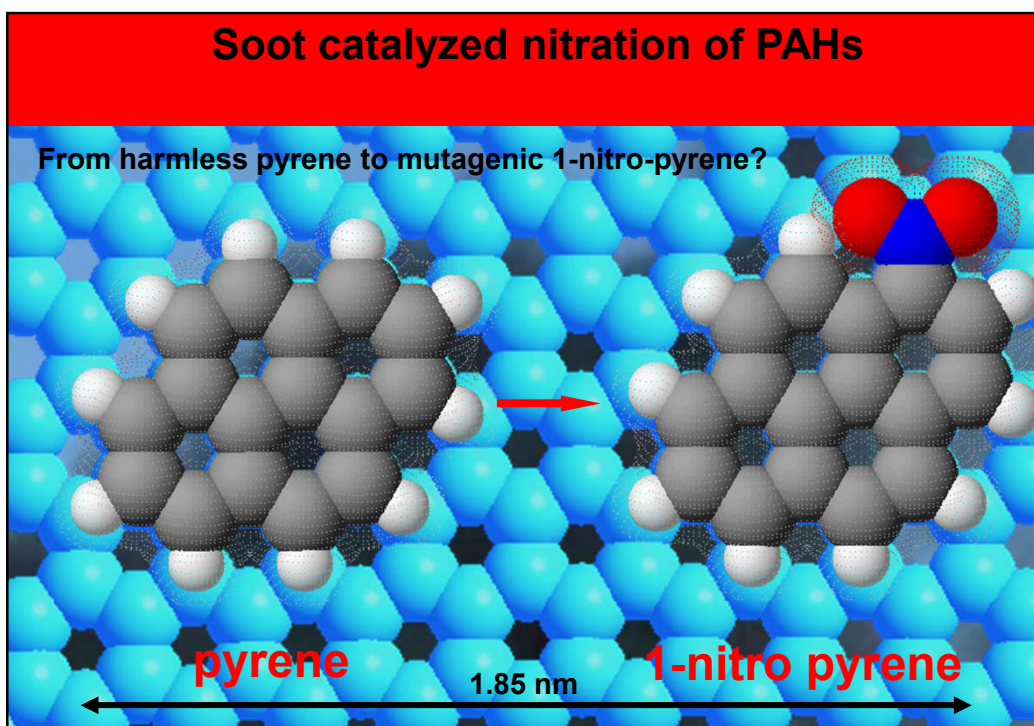


Empa

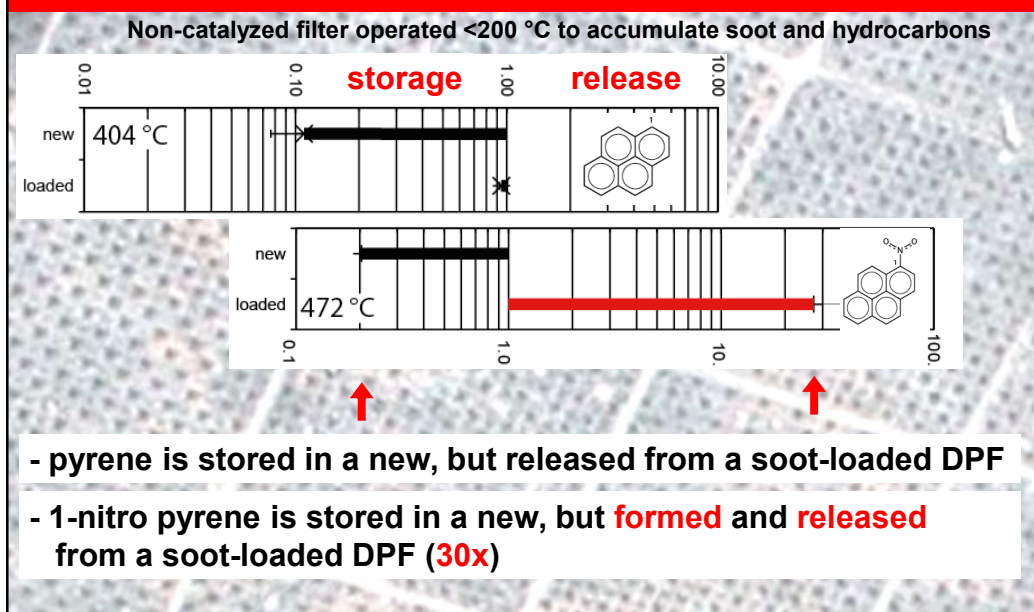
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Nitro-PAH formation in non-catalyzed DPF

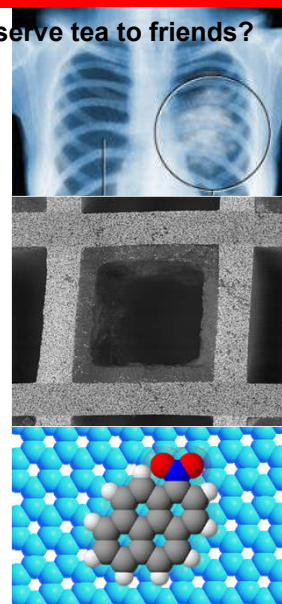


Secondary emissions from emission control devices and their impact on occupational health and safety

Would you run a diesel engine in your living room and serve tea to friends?

Outline

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Secondary emissions from emission control devices and their impact on occupational health and safety

It's a long way from diesel soot to CO₂ and H₂O!

Results

- DPFs lower PN emissions >98% (n>60)
- DPFs lower emissions of genotoxic PAHs
- Some DPFs can form mutagenic nitro PAHs

Conclusions



Best available filter technologies:

- remove persistent soot nanoparticles
- lower the genotoxic potential of the exhaust
- have low risks for secondary pollutants

Consider VERT-approved filters for retrofitting

Secondary emissions from emission control devices and their impact on occupational health and safety

A combined effort with many important contributions

Thanks:

- **VERT team:** Andreas Mayer, TTM, Niederrohrdorf
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Hans Jaeckle, Urs Debrunner, Oliver Schumm, Intertek Caleb Brett, Schlieren.
- **Empa colleagues:** Brigitte Buchmann, Thomas Bühner, Lukas Emmenegger, Anna-Maria Forss, Urs Gfeller, Maria Guecheva, Peter Graf, Roland Graf, Erika Guyer, Regula Haag, Peter Honnegger, Judith Kobler, Martin Kohler, Peter Lienemann, Alfred Mack, Peter Mattrel, Martin Mohr, Joachim Mohn, Christof Moor, Maria Munoz, Andreas Paul, Peter Schmid, Cornelia Seiler, Andrea Ulrich, Heinz Vonmont, Thomas Walter, Max Wolfensberger, Daniela Wenger, Adrian Wichser, Simon Wyss, Markus Zennegg, Kerstin Zeyer.
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Thomas Gasser, Heinz Berger, Gerhard Stucki, Swiss Federal Road Office
- **Filter- & catalyst manufacturers:** >60 different diesel particulate filter systems




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SOCIÉTÉ SUISSE DE CHIMIE SSC
SWISS CHEMICAL SOCIETY SCS

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