Experience with trapping diesel nanoparticles in Switzerland
Experiences with Trapping Diesel Nanoparticles in Switzerland

A. Mayer, TTM
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F. Legerer, AKPF

According to the Swiss off-road emission inventory 1993, contributions of the off-road and of the on-road sector to particulate matter were, and probably still are, about equal; however, off-road tends to take the lead because forthcoming legislation with regard to emissions to be set by the EU will apply exclusively to on-road.

In 1994 SUVA, the Swiss occupational health authority, edited a new threshold-list for toxic air contaminants at the working place; for first time a limit for Diesel particulate DME was established, being as low as 0.2 mg/m\(^3\), -- reduced to 0.1 mg/m\(^3\) EC since and defined as EC+OC; whereas particle size range was defined according to the Johannesburg convention. The same list, which has legally binding character, declared Diesel particulate emissions as “carcinogenic” which in turn requires minimisation according to best available technology, BAT.

However, what was BAT at that time? And were there any technical solutions available to reduce particulate emissions of existing off-road engines by a factor of > 50 down to the required level?

These questions prompted the VERT-project, a co-operation of the occupational health and clean air authorities of Switzerland, Germany and Austria in 1994. After 2 years of basic research reviewing engine developments, fuel developments and after-treatment methods it became quite clear that only hot exhaust gas filtration could be sufficiently efficient to trap the Diesel-typical nanoparticles within the mobility size range of 10-500 nm.

During a 2 years follow-up period of field testing it could be demonstrated that reliable technology was available and filtration quality of some available products was exceeding 98 %.

Meanwhile a number of important improving steps with regard to implement of retrofit technology had been accomplished:

- **Consensus on a definition of Diesel particulate emissions**: “Solid particles and everything adsorbed to them within the size range of 10-500 nm”; whereas “solid” means solid at filter flow conditions, reference-temperature of measurement at 300 °C was accepted.
- **Trap-system specifications**, a very important guideline for manufacturers; this has influenced technology to a great deal since.
- **Verification protocol for trap systems VFT=VERT tests for filters and for catalytic regeneration tools**, like coatings or fuel catalysts and an additional VSET= VERT secondary emissions test. Meanwhile, these protocols have been accepted as well and approved by the Canadian DEEP and US-MSHA, while harmonisation with CARB is on the way. VFT and VSET have since permitted to exclude inefficient and unreliable systems from the market and to ban fuel additives which increase gaseous emissions like NO\(_2\) and generate secondary emission like dioxins, furanes, PAH, nitro-PAH or other toxic substances.

Measurement of nanosize particles grew to a very important issue during this period. Observation of artefacts due to inappropriate sampling conditions lead to the development and application of thermodenuders and hot/high dilution systems. Comparison of all available nanoparticle measurement systems like DMA, CPC, SMPS, ELPI and PAS and systematic comparison with PM- and coulometric EC/OC-analysis made inherent problems apparent. The necessity for development of improved measurement technology was clearly recognised. NanoMet was started as a project and resulted in solutions like the rotating disc diluter, the combination of DCS- and PAS- sensor and finally the diffusion battery DYNAC for the combination of mobility sizing and detection of surface and number in one single step only and online.
BAT had been defined by then and all necessary tools for implementation have been developed. Legislation started with LRV 98 and the SUVA Filter Obligatorium in 2000. By now, about 4'000 vehicles have been retrofitted with particulate traps ranging from 10-1’100 kW and from construction machines to city buses. Application of traps is mandatory at construction sites in cities and underground and this extremely efficient solution to clean Diesel exhaust is picked up by more and more authorities, institutions and private companies on a voluntary basis. Originally, at the beginnings, failure rate was > 5 %; it could be reduced to less than 3 % by now, observing carefully VFT+VSET and further reduction is expected to only 1 % or even less by introduction of electronic on-board controls and application of active systems to a greater extent. Nanoparticles are invisible, their diameters being substantially less than the wave length of visible light, but they effect human health and consequently the necessity of measurement methods for nanoparticle number counts is widely accepted. Opinion crystallises that ambient and roadside measurements should be performed and criteria accordingly established. New filter systems are appearing and their efficiencies of nanoparticle-trapping are as high as 99,8 %. There exists a clear potential for traps, for more as well as for less bulky ones, less costly ones and fully automatic systems envisioned. Therefore, Swiss authorities are studying retrofits of all HD-vehicles, on-road and off-road.
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A. Mayer, TTM
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The Swiss Chronicle to reduce Diesel Particulate emissions by > 99 %

- Offroad Emissions Inventory 1993
  (computer-model for predictions now available on CD)
- VERT-Project 1994-1998 to define and prove BAT
- BUWAL (Swiss EPA):
  Clean Air Act Amendments 1998
  - Diesel Particulate classified as carcinogenic
    (to be minimized acc. to BAT irrespective of cost)
  - PM 10 – Limit 20 µg/m3 (exceeded in all cities)
  - Traps for construction sites in/nearby cities
- SUVA (Swiss Occupational Health Authority):
  Traps mandatory for the working place 3/2000
- Implementation Tools:
  - Trap-system specification 1996
  - Trap-system-certification procedure 1997
  - Filter-list of all certified traps on the Net 1998
  - Ultrafine particle metrology “NanoMet” 1999
  - EC-oriented calibration method by EAM 1999
  - Trap OBD-systems “LogLink” 1999
  - Trap manufacturer association AKPF 1998
  - Nanoparticle measurement conference since 1997
- > 4000 traps in operation – controlled by field testing
  (should be 100 % yearly control – now only 10-15 %)

STUMP-Postulat: “Retrofit of all swiss HDV-onroad (66'000)
with efficient filters vehicles as soon as possible”
Government decision expected end of 2001
# History of particulate trap retrofits in Switzerland

<table>
<thead>
<tr>
<th></th>
<th>Truck</th>
<th>Bus</th>
<th>Construction machine</th>
<th>Forklift</th>
<th>Ship</th>
<th>Stationary</th>
<th>Total</th>
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<td>Vor 1995</td>
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<td>238</td>
<td>65</td>
<td>81</td>
<td>32</td>
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<td>95 - 98</td>
<td>4</td>
<td>17</td>
<td>40</td>
<td>275</td>
<td>35</td>
<td>11</td>
<td>382</td>
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<tr>
<td>1999</td>
<td>4</td>
<td>130</td>
<td>269</td>
<td>122</td>
<td>19</td>
<td>33</td>
<td>577</td>
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<tr>
<td>2000</td>
<td>35</td>
<td>304</td>
<td>416</td>
<td>111</td>
<td>26</td>
<td>35</td>
<td>927</td>
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<tr>
<td>Total</td>
<td>78</td>
<td>689</td>
<td>790</td>
<td>589</td>
<td>112</td>
<td>81</td>
<td>2339</td>
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</tbody>
</table>
History of particulate trap retrofits in Switzerland
Which engines should be retrofitted and why?

- Old engines are high emitters, must be retrofitted
- New engines are cleaner but live longer, must be retrofitted
- Small engines emit specifically (g/kWh) more particles (power [kW] x emission factor [g/kWh] is nearly constant) must be retrofitted

⇒ all Diesel engines should be retrofitted, except …

Which engines should not be retrofitted and why?

- 2-stroke engines since they emit too much lubeoil and are very backpressure-sensitive
- Any engine consuming more than 2% (1% ?) lubeoil compared to fuel consumption
- Excessive smokers: Bosch > 5 / Opacity > 80%

What to do with such engines: rebuild or scrap !

Swiss BauRLL: Diesel engines which can not be retrofitted with particulate traps for some technical or other reasons are no longer allowed to be operated
Verification Tests and Control Schemes for Diesel Particulate Filters in Switzerland

### Type Approval

**VFT 1**
- **VERT Filter Test**
  - Steady state + transient during regeneration
  - EC/PM/SMPS/NM

**VSET**
- **VERT secondary emission test**
  - Steady state
  - PAH/PCDD/Nitro-PAH
  - Size spec. analysis
  - GC-MS../ELPI chem. analysis

**VFT 2**
- **VERT field Test**
  - Controlled field test > 2000 hrs datalogging
  - P/T/RPM

**VFT 3**
- Repetition of VFT 1
  - Steady state + transient during regeneration

### Field Control

- **Steady state** mandatory
- **transient** periodic field test 1 x p/year
- **Opacity / BC**
- Steady state
- PAH/PCDD/Nitro-PAH low/high idle
- Gas emissions CO/CO₂/O₂/NO/NO₂/HC
- **Optional:**
  - Statistical:
    - Steady state free accell.
    - fine particle filtration measurement
  - EC-Cells
  - NDIR

- **Nanomet**

### Filter List
- Recommended systems

Filter listing confirmed, if failure rate < 5%
- rejected if > 5%
**Test-Protocol for VSET**

Sampling during 2-4 hrs of repeated ISO 8178 test cycles

- **Engine**
- **Exhaust**
- **Part flow dilution tunnel**
- **NanoMet**
- **SMPS**
- **ELPI** (13 stages)
- **Flow proportional undiluted sample gas + particulate**
- **GC (VOC)**
- **HPLC (VOCOX)**
- **HPLC (PAH)**
- **SOF/INSOF**
- **GC-MS (PCDP/F/Nitro-PAH)**
- **X-Ray Diffraction**
- **SEM/EDX**
- **ICP-MS** (inductively coupled plasma mass spectrometry: detection limit = 0.001 µg/stage)
Metrology Concepts of the swiss Group

- **Sampling**
  direct sampling upstream/downstream
  short, el. conductive sampling lines heated > 150°C
  avoiding artefacts by condensation

- **Conditioning**
  high/hot dilution with ultraclean air > 1:100
  avoiding artefacts by agglomeration and pollution

- **Classifying**
  acc. to mobility
  size range 10-500 nm
  up to 5 size classes

- **Measurement Targets**
  mobility diameter
  number and surface
  chemical fingerprint

- **Instrumentation**
  SMPS / ELPI / PAS / DC
  compared to EC-mass and PM

- **Calibration**
  CAST
  compared to EC-mass
Trap Type Approval, Test Concept

**VFT = VERT filter test**
- Stage 1: bench test with new filter
- Stage 2: > 2000 field operation hours
- Stage 3: control bench test

All system components e.g., additiv dosing system and electronic control must be included

**VSET = VERT secondary emission test**
- must be performed with all systems with build-in catalytic activity
- PAH; Nitro-PAH; PCDD/F; NO₂; N₂O, Aldehydes ….

**VFTₖ = shortened VFT**
- for systems which have successfully passed VFT after minor technical modifications

Conditions to use regenerations additives
- Additive must be registered (new substance, toxicity)
- VFT successfully passed
- VSET successfully passed
- Dosing system automatic on-board
- Electronic control with dosing-stop if filter leaks
- Environmentally friendly system to clean filter from additive ashes
Field Control for Trap-Systems

**Mandatory:** Black smoke during free acceleration
mobile Opacimeter
not very sensitive
but good enough to identify trap damage
widely available in Switzerland
because of onroad legislation

However Opacimetry does not monitor ultrafine particulate emission and will not be sufficiently sensitive for future engines

**Optional:** Gas measurement during study state
Low idle, high idle,
torque converter stall
CO, CO₂, O₂, NO, NO₂, HC
NDIR and EC cells

**Optional:** Ultrafine Particle Filtration Efficiency
NanoMet identifies trap efficiency
by simultaneous transient measurement upstream and downstream during snap-on acceleration and can monitor ambient effects (threshold control) with the same instrument
### Filtration Efficiency of Particulate Filters according to VERT Filter Test VFT

Average of 4 points of operation, ISO 8178

<table>
<thead>
<tr>
<th>Products</th>
<th>PMAG 1)</th>
<th>PZAG 2)</th>
<th>ECAG 3)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>without additive</td>
<td>with additive</td>
<td>without additive</td>
</tr>
<tr>
<td><strong>New filters</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3M</td>
<td>80.7</td>
<td>-</td>
<td>98.6</td>
</tr>
<tr>
<td>OBERLAND Typ A</td>
<td>90.5</td>
<td>-</td>
<td>98.4</td>
</tr>
<tr>
<td>OBERLAND Typ B</td>
<td>86.1</td>
<td>80.2</td>
<td>98.5</td>
</tr>
<tr>
<td>JMC</td>
<td>84.5</td>
<td>-</td>
<td>99.3</td>
</tr>
<tr>
<td>IBIDEN</td>
<td>87.2</td>
<td>-</td>
<td>99.9</td>
</tr>
<tr>
<td>CORNING</td>
<td>83.8</td>
<td>86.2</td>
<td>99.7</td>
</tr>
<tr>
<td>HJS/CRT</td>
<td>83.8</td>
<td>-</td>
<td>99.4</td>
</tr>
<tr>
<td>AMMANN Typ B</td>
<td>-</td>
<td>76.1</td>
<td>96.3</td>
</tr>
<tr>
<td>AMMANN Typ K-2</td>
<td>-</td>
<td>85.2</td>
<td>96.3</td>
</tr>
<tr>
<td>DCL</td>
<td>-</td>
<td>77.7</td>
<td>99.4</td>
</tr>
<tr>
<td>EHC L20</td>
<td>-</td>
<td>-</td>
<td>96.7</td>
</tr>
<tr>
<td>GREENTOP/Buck</td>
<td>-</td>
<td>72.2</td>
<td>-</td>
</tr>
<tr>
<td>INTECO</td>
<td>88.9</td>
<td>-</td>
<td>98.1</td>
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<tr>
<td><strong>After field test 2000 hrs:</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SHW (LIB1)</td>
<td>3.2</td>
<td>22.2</td>
<td>96.3</td>
</tr>
<tr>
<td>SHW (CAT1)</td>
<td>77.5</td>
<td>87.6</td>
<td>97.8</td>
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<td>BUCK (LIB2)</td>
<td>76.5</td>
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<td>BUCK (CAT3)</td>
<td>64.2</td>
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<td>ECS (LIB3)</td>
<td>12.4</td>
<td>43.0</td>
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<td>UNIKAT (CAT4)</td>
<td>54.7</td>
<td>76.2</td>
<td>99.0</td>
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<tr>
<td><strong>Average</strong></td>
<td>70.5</td>
<td>98.3</td>
<td>96.8</td>
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</tbody>
</table>

1) PMAG: Particulate mass based efficiency  
2) PZAG: Particulate number based efficiency  
3) ECAG: Elementary carbon based efficiency
Particle size spectrum of a modern trap: IBIDEN SiC 200 cpi with RHODIA fuel additive DPX9

Liebherr D914 T, 1400 min⁻¹/ full load, after reg. with thermodenuder
Formation of NO/NO$_2$
in function of exhaust gas temperature and rpm

Bus 2:
Volvo/EMINOX  10 ppm S

Bus 3:
MB/HJS  10 ppm S

Bus 4:
NAW/EMINOX  30 ppm S
Combined toxicity according to CML-method
(CML: Centrum voor Milieukunde Leiden)

Basis: Average HDV, Year 2000, without aftertreatment
Particulate trap 95% Efficiency
Particulate trap + Oxidation catalyst
Assumption: NO2 toxicity = 5 x NO toxicity

Source: BUWAL 2000
Regeneration of SHW particle filter, 15 ppm Ce and 1 ppm Pt
### Emission factors for dioxines and furanes

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Value</th>
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<tr>
<td>O</td>
<td>No filter</td>
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</tr>
<tr>
<td>C</td>
<td>Cerium additive</td>
<td></td>
</tr>
<tr>
<td>H</td>
<td>11 ppm Chlorine</td>
<td></td>
</tr>
<tr>
<td>X</td>
<td>100 ppm Chlorine</td>
<td></td>
</tr>
<tr>
<td>M</td>
<td>Sintered metal filter</td>
<td></td>
</tr>
<tr>
<td>E</td>
<td>Iron additive</td>
<td></td>
</tr>
<tr>
<td>K</td>
<td>Copper additive</td>
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</table>

The diagram illustrates the emission factors in pg/l for different conditions. The symbols and values correspond to the described conditions.
Transient Measurement of fine particle filtration efficiency by NanoMet

![Graph showing transient measurement of fine particle filtration efficiency](image-url)
## Application oriented Trap Selection (2)

<table>
<thead>
<tr>
<th>Type of Trap Regeneration</th>
<th>Exh. Temp. low or uncertain</th>
<th>Engine Emissions</th>
<th>Dynamic or Stationary</th>
<th>Fuel</th>
<th>Lube Oil</th>
<th>Dependability</th>
<th>Infra-structure</th>
<th>Commerc. Availibilty</th>
<th>Application unlimited?</th>
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<tr>
<td><strong>Active Regeneration</strong></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Diesel Burner</td>
<td>full flow online</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td></td>
<td>standstill online</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>-</td>
<td>-</td>
<td>+</td>
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<tr>
<td>Electric Burner</td>
<td>full flow online</td>
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<td>+</td>
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<td>+</td>
<td>+</td>
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<td></td>
<td>Standstill onboard</td>
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<td>+</td>
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<td>-</td>
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<td>Catalytic Coating</td>
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<td>+</td>
<td>(-)</td>
<td>(+)</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>-</td>
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<tr>
<td>NO₂-Process CRT</td>
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<td>+</td>
<td>-</td>
<td>+</td>
<td>+</td>
<td>+</td>
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<td><strong>Combinations</strong></td>
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<td></td>
<td></td>
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<tr>
<td>- Diesel B + Add.</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>-</td>
<td>+</td>
</tr>
<tr>
<td>- El.B. + Add. + Cat.</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>-</td>
<td>+</td>
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<tr>
<td><strong>Active Assist</strong></td>
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<tr>
<td>Engine Heat Release</td>
<td>- Fuel Intection</td>
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<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>-</td>
<td>+</td>
</tr>
<tr>
<td></td>
<td>- Intake Throtteling</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>+</td>
<td>-</td>
<td>+</td>
</tr>
</tbody>
</table>
Distribution of dwell time at given temperature levels for vehicle category 2. “Frequency” here means the number of observations per temperature class (25 °C steps) and dwell time class (1 sec.-steps) with duration 1 sec per observation.
Trap Retrofits in Switzerland (10/2000)

• **Statistics**

<table>
<thead>
<tr>
<th>Application</th>
<th>Reported Failures</th>
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<tbody>
<tr>
<td>Passenger Car</td>
<td>-</td>
</tr>
<tr>
<td>Truck</td>
<td>78</td>
</tr>
<tr>
<td>City Bus</td>
<td>689</td>
</tr>
<tr>
<td>Coaches</td>
<td>-</td>
</tr>
<tr>
<td>Construction</td>
<td>800</td>
</tr>
<tr>
<td>Fork Lift</td>
<td>589</td>
</tr>
<tr>
<td>Ship / Rail</td>
<td>118</td>
</tr>
<tr>
<td>Stationary</td>
<td>81</td>
</tr>
<tr>
<td>Snap on</td>
<td>28</td>
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• **Efficiencies** (8 systems qualified)

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<th>System</th>
<th>Efficiency</th>
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<tbody>
<tr>
<td>1</td>
<td>99,8%</td>
</tr>
<tr>
<td>2</td>
<td>99,6%</td>
</tr>
<tr>
<td>3</td>
<td>99,4%</td>
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<tr>
<td>4</td>
<td>99,4%</td>
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<tr>
<td>5</td>
<td>99,3%</td>
</tr>
<tr>
<td>6</td>
<td>98,1%</td>
</tr>
<tr>
<td>7</td>
<td>96,9%</td>
</tr>
<tr>
<td>8</td>
<td>96,7%</td>
</tr>
</tbody>
</table>

• **Operation**

<table>
<thead>
<tr>
<th>Fahrzeugtyp</th>
<th>Filter Type</th>
<th>Operation</th>
<th>Location</th>
</tr>
</thead>
<tbody>
<tr>
<td>City Bus</td>
<td>DB/ M&amp;H</td>
<td>&gt; 750'000 km</td>
<td>VBZ Zürich</td>
</tr>
<tr>
<td>Truck</td>
<td>UNIKAT</td>
<td>&gt; 500'000 km</td>
<td>KIBAG Zürich</td>
</tr>
<tr>
<td>Construction</td>
<td>UNIKAT</td>
<td>&gt; 10'000 op.hrs</td>
<td>Dübendorfer</td>
</tr>
<tr>
<td>Fork Lift</td>
<td>HUSS</td>
<td>&gt; 20'000 op.hrs</td>
<td>Landor AG</td>
</tr>
<tr>
<td>Ship</td>
<td>HUG</td>
<td>&gt; 20'000 op.hrs</td>
<td>Lake of Konstanz</td>
</tr>
<tr>
<td>Stationary</td>
<td>BUCK</td>
<td>&gt; 10'000 op.hrs</td>
<td>Reichstag Berlin</td>
</tr>
</tbody>
</table>
Trap-Retrofit Statistics and Field Results/Overview

- **2383 Traps in use in Switzerland (October 2000)**
  - 78 on trucks
  - 689 on buses
  - 800 on construction machines
  - 589 on fork lifts
  - 81 on stationary engines
  - 118 on rail and ships
  - 28 for periodic use (garages)

It is expected that this numbers will double in 2001/2

- **Failures:**
  - 154 reported failures over 10 years: 6.5%
  - 84 reported failures after 1995: 3.5%
  - 56 excluding failures of experimental systems
  → failure rate net: 2.6%
  may be somewhat higher (5% ?)
  Target: < 1 %

- **Emission Measurements in the Field in 2000**
  - 207 measurements analysed
  - complains (exceeding 0,24 1/m Opacity): 37 total
  - excluding unsuccessful experimental systems: 7
  → complains net: 3.3%
Risks for Filters
- Filter overheating during regeneration
- Filter fracture due to vibrations
- Filter material damage due to oil ash compounds
- Filter clogging due to silencer fibers
- Filter overloading with following fracture or blow off

Regeneration Malfunctions
- Operation temperature too low or peaks too short
- Additive dosing inadequate
- Engine deterioration (injection nozzle failure)

Risk for the Engine
- Excessive backpressure
  Increases fuel consumption and material temperature
- No engine damage known due to particulate trap application
- Additive suppliers warranty in case of engine damage

Risks for the environment and people
- High exhaust emission temperature during regenerations

Measures to minimize such risks
- Electronic on-board control with 2 alarm levels
- Automatic on-board additive dosing
- Active systems whenever operation temperature fluctuates too much and low temperature level operation is not excluded
- Permit only systems which have passed 2000 hrs-test
- Assure proper trap selection with preceeding data-logging
- Periodic emission control once per year
- Exclude high-emitters
- Use cleaner fuels and luboils
With these measures we expect to reduce the failure rate to

\(< 1\% \text{ per year within 1 year}\)