

# Messung von Russpartikeln mit neuen technischen Verfahren als Grundlage für einen Zertifizierungsansatz

Measurement of soot particles with state-of-the-art methods  
as a basis for a new certification approach

- SOOTCERT -

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Institut für Physik der Atmosphäre  
Oberpfaffenhofen, 82234 Wessling

This study was prepared by

ANDREAS PETZOLD

[andreas.petzold@dlr.de](mailto:andreas.petzold@dlr.de)

Deutsches Zentrum für Luft- und Raumfahrt  
Institut für Physik der Atmosphäre  
Oberpfaffenhofen  
82234 Wessling  
Germany

ANDREAS HOTES, ANDRES RADIG

AVISTRA GmbH  
Reinhardtstr. 58  
10117 Berlin  
Germany

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<b>16) Abstract</b> <p>The emission of particulate matter from aircraft engines, their potential impacts on Earth's climate and human health, and state-of-the-art methods for measuring particulate matter in aircraft engine exhaust are high up on the agenda of research activities as well as of international bodies like ICAO, FAA, and EASA. There are parallel ongoing activities as part of the CAEP process, at the FAA framed into the PM roadmap, and recently within EASA and within the German Federal Ministry of Transport, Building and Urban Affairs (BMVBS), Aviation and Space Division. On the request of ICAO, the SAE E-31 Committee on Aircraft Exhaust Emissions Measurement has worked in the past years towards new measurement approaches for particulate matter from aircraft engines. A consensus has been achieved on the methods which are in principle available for this task. However, there is ongoing discussion on the appropriate measures for particle emissions with respect to aerosol-cloud interaction (climate impact) and adverse human health effects. An even more pressing issue are appropriate measurement approaches for volatile, thermally instable particles which so far are postponed from the approach chosen by the SAE E-31 committee because of unsolved technical questions, although these particles are considered relevant for human health aspects e.g., by US EPA.</p> <p>This report summarises the BMVBS funded activities for the development of a new particle matter emissions regulation for new aircraft engines. It incorporates recent activities of the authors in the framework of ICAO / CAEP and SAE E-31 committee work in order to align the selected approach with international ongoing activities.</p> <p>The conclusions concerning potential particle metrics and related limiting values are:</p> <ol style="list-style-type: none"> <li>1. The non-volatile combustion particle fraction is correlated to engine technology. This fraction is stable because it does not depend on sample handling as long as particle coagulation and losses to the sample line walls are avoided.</li> <li>2. The consideration of volatile particulate matter emissions suffers from the lack of knowledge of the contributing compounds. However, volatile particulate mass is predominantly linked to fuel sulphur and atmospheric conditions during plume dilution rather than to engine technology.</li> <li>3. Potential precisely accessible particle properties are particle number and particle mass. Particle number, because the scientific debate on health effects of particulate matter focuses on insoluble nanometer-sized particles by number. Particle mass, because current air quality regulations focused on particle mass.</li> <li>4. A limiting value will have to be related to the rated thrust, irrespective of the measured variable, i.e. mass and/or number, so that it is defined analogous to other already existing limiting values.</li> </ol>		
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<p>Die Emission von Partikeln aus Flugzeugtriebwerken, deren potentielle Auswirkungen auf das Klima und auf die menschliche Gesundheit sowie Messverfahren für Partikel nach dem Stand der Technik stehen derzeit sehr weit oben auf der Tagesordnung sowohl von Forschungsaktivitäten als auch von internationalen Einrichtungen wie ICAO, FAA und EASA. Aktuell laufen parallele Aktivitäten im Rahmen des CAEP - Prozesses, innerhalb der FAA als Teil der Particulate Matter Roadmap und in jüngster Zeit auch innerhalb der EASA und des Bundesministeriums für Verkehr, Bau und Stadtentwicklung (BMVBS), Abteilung Luft- und Raumfahrt. Auf Anfrage der ICAO befasst sich das SAE E-31 Committee on Aircraft Exhaust Emissions Measurement seit mehreren Jahren mit neuen Messansätzen für partikelförmige Emissionen aus Flugzeugtriebwerken. Hinsichtlich der für diese Frage prinzipiell zur Verfügung stehenden Messverfahren wurde Übereinstimmung erzielt. Die aktuelle Debatte konzentriert sich auf die Frage, welche Partikelparameter für die Charakterisierung der Emissionen im Bezug auf ihre Klima- und Gesundheitswirkung sinnvoll sind. Insbesondere die Bedeutung von flüchtigen, thermisch instabilen Partikeln, die bisher vom Ansatz des SAE E-31 ausgeschlossen sind, wird diskutiert, da diese Partikelklasse von Institutionen wie der US EPA als sehr wichtig für eine potentielle Gesundheitsgefährdung eingestuft werden.</p> <p>Der vorliegende Bericht fasst die durch das BMVBS geförderten Aktivitäten zur Entwicklung eines neuen Emissionsgrenzwertes für Partikelemissionen aus neuen Flugzeugtriebwerken zusammen. Er berücksichtigt auch aktuelle Aktivitäten im Rahmen von ICAO / CAEP und SAE E-31.</p> <p>Die Schlussfolgerungen im Bezug auf Partikelmaßzahlen und damit im Zusammenhang stehende Grenzwerte sind:</p> <ol style="list-style-type: none"> <li>1. Der Anteil der nichtflüchtigen Verbrennungspartikel hängt eng mit der Triebwerkstechnologie zusammen. Diese Partikelfraktion ist stabil, da sie nicht von der Probenahme abhängt, solange Partikelkoagulation und Verluste an der Probenahmeleitung vermieden werden.</li> <li>2. Die Berücksichtigung von flüchtigen Partikelbestandteilen wird erschwert durch fehlendes Wissen über die chemische Zusammensetzung. Die Hauptkomponenten hängen jedoch eher vom Schwefelgehalt des Treibstoffs und von den atmosphärischen Bedingungen während der Verdünnung des Abgases ab als von der Triebwerkstechnologie.</li> <li>3. Potentielle, präzise messbare Partikeleigenschaften sind Partikelanzahl und Partikelmasse. Partikelanzahl, weil sich die wissenschaftliche Diskussion über Gesundheitswirkungen auf die Anzahlkonzentration nichtlöslicher Nanopartikel konzentriert. Partikelmasse, weil die derzeitigen Luftqualitätskriterien auf der Partikelmassenkonzentration basieren.</li> <li>4. Ein Grenzwert muss bezogen werden auf den Schub, unabhängig vom Parameter wie Masse und/oder Anzahl, so dass der Grenzwert analog zu weiteren, bereits bestehenden Grenzwerten definiert ist.</li> </ol>		
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# Chapter 1      Background and Objectives

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The emission of particulate matter from aircraft engines, their potential impacts on Earth's climate and human health, and state-of-the-art methods for measuring particulate matter in aircraft engine exhaust are high up on the agenda of research activities as well as of international bodies like ICAO, FAA, and EASA. There are parallel ongoing activities as part of the CAEP process, at the FAA framed into the PM roadmap, and recently within EASA and within the German Department for Transport BMVBS. This report summarises the BMVBS activities which work towards the development of a new particle matter emissions regulation for new aircraft engines.

On the request of ICAO, the SAE E-31 Committee on Aircraft Exhaust Emissions Measurement has worked in the past years towards new measurement approaches for particulate matter from aircraft engines. At present, the respective measure is the SAE smoke number, which is related to the darkening of particle-loaded filters by the deposited particulate matter. Historically, the justification came from a visibility criterion for aircraft plumes. Along with cleaner engines in terms of smoke and with increasing interest in an environmentally compatible air transportation system, this measure is no longer regarded as appropriate for meeting current needs which focus on climate effects via contrail and cirrus-contrail interactions, and on particle-related health effects. The replacement of this out-dated method is one of the key issues of ongoing committee work.

A consensus has been achieved on the methods which are in principle available for this task. The agreement is laid down in a position paper on the measurement of particulate matter from gas turbines (SAE, 2002). This position paper highlights the committee's views and thoughts about concentrating on non-volatile particles and shifting the measurement of volatile particulate matter to a later stage until consensus is achieved on how to treat the volatile particulate matter issue. Based on this position paper, the committee has prepared the Aerospace Information Report AIR 5892 (SAE, 2004) which reviews the methods available for the measurement of non-volatile particulate matter in engine emissions.

However, there is ongoing discussion on the appropriate measures for particle emissions with respect to aerosol-cloud interaction (climate impact) and adverse human health effects. An even more pressing issue are appropriate measurement approaches for volatile, thermally instable particles which so far are postponed from the approach chosen by the SAE E-31 committee because of unsolved technical questions, although these particles are considered relevant for human health aspects. In a second position paper the committee has expressed its opinion on open questions about measuring volatile particles (SAE, 2005). Recent CAEP information papers, e.g., CAEP/7-IP/6 "Particulate Matter Characterisation" (CAEP, 2007) reviewed the efforts undertaken during the 1990's and in the early 21<sup>st</sup> century in Europe and in the USA. The main achievements of research were summarised and the First Order Approximation (V3.0) was introduced which works towards an estimate of particulate matter emissions from standard ICAO data information to be used for local air quality assessments.

This report incorporates recent activities of the authors in the framework of ICAO / CAEP and SAE E-31 committee work in order to align the selected approach with international ongoing activities. In a first attempt potential consequences of particle emission regulatory standards on aviation industry are investigated and discussed. The overall objective of this study is to review properties of particles emitted from aircraft engines, discuss consequences for potential measurement methods and investigate possible particles metrics for future particulate matter emission regulatory standards.

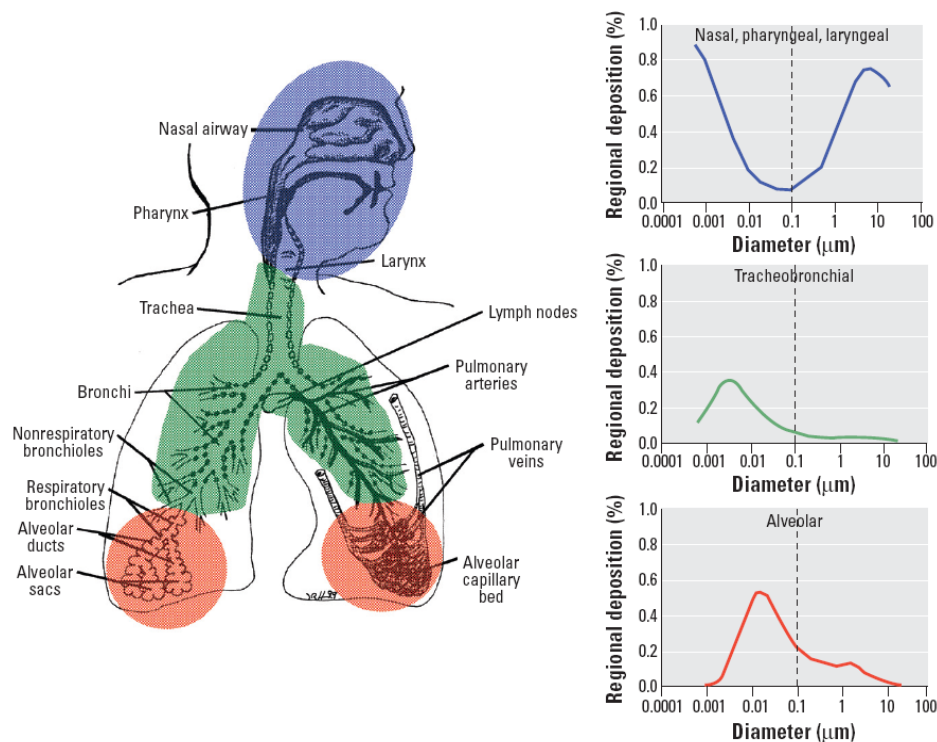
## Chapter 2 Health Effects

### 2.1 Impact of Particulate Matter on Human Health

Adverse health effects of inhaled particulate matter have been widely studied in humans and animals and include asthma, lung cancer, cardiovascular issues, and premature death. An extensive review of this rapidly developing research area is given by Oberdörster et al., (2005). Impacts of inhaled particulate matter on human health are related to cardiovascular effects (e.g., heart disease, vascular inflammation, atherosclerosis), respiratory effects (e.g., pulmonary disease, lung inflammation), and translocation and toxicity to other organs like liver or even the brain.

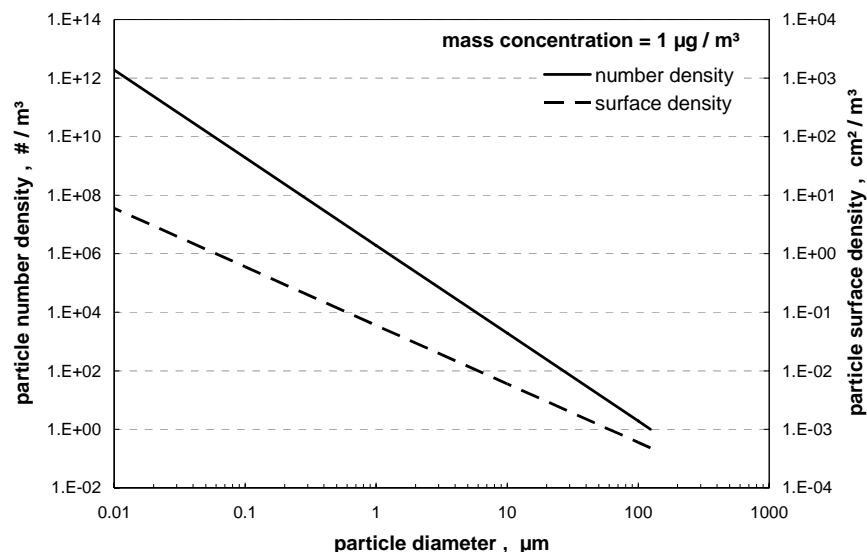
Particle size is a main determinant of where in the respiratory tract the particle will come to rest when inhaled. Particles larger than 10  $\mu\text{m}$  in diameter are generally filtered in the nose and throat and do not cause problems, particles smaller than about 10  $\mu\text{m}$  ( $\text{PM}_{10}$ ) can settle in the bronchi and lungs, particles smaller than 2.5  $\mu\text{m}$  ( $\text{PM}_{2.5}$ ) tend to penetrate into the gas-exchange regions of the lung, and very small particles < 100 nanometer (nanoparticles or nanometer-sized particles) may even pass through the lungs and cell membranes into the blood circulation system and affect other organs. Figure 2-1 shows the deposition regions in the pulmonary system for particles of different sizes (Oberdörster et al., 2005).

In particular, there are indications that  $\text{PM}_{2.5}$  leads to high plaque deposits in arteries, causing vascular inflammation and atherosclerosis – a hardening of the arteries that reduces elasticity, which can lead to heart attacks and other cardiovascular problems (Pope, et al., 2002). Recent results suggest that even short-term exposure at elevated concentrations can significantly contribute to heart disease.



**Figure 2-1:** Deposition regions in the pulmonary system for particles of different sizes (Oberdörster et al., 2005).





**Figure 2-2:** Particle number density and particle surface density as a function of particle diameter for a constant mass concentration of  $1 \mu\text{g} / \text{m}^3$ .

There is also evidence that particles smaller than 100 nanometres in diameter can pass through cell membranes into the blood circulation system of humans (Nemmar et al., 2002). For example, particles may migrate into the brain. It has been suggested that particulate matter can cause similar brain damage as that found in Alzheimer patients. Particles emitted from modern diesel engines are typically in the size range of 100 nanometres (nm.). In addition, these soot particles also carry carcinogenic components like benzopyrenes adsorbed on their surface.

Clearance pathways of the respiratory system can be divided into physical translocation of particles by different mechanisms, and to chemical clearance processes such as leaching, or chemical dissolution of soluble components. The most relevant clearance process for solid particles in the alveolar region is mediated through macrophage-particle interaction. Studies showed, that approx. 80% of  $0.5 - 10 \mu\text{m}$  particles could be retrieved with the macrophages, whereas only approx. 20% of the nanometer-sized particles  $< 80 \text{ nm}$  could be lavaged by macrophages (Oberdörster et al., 2005).

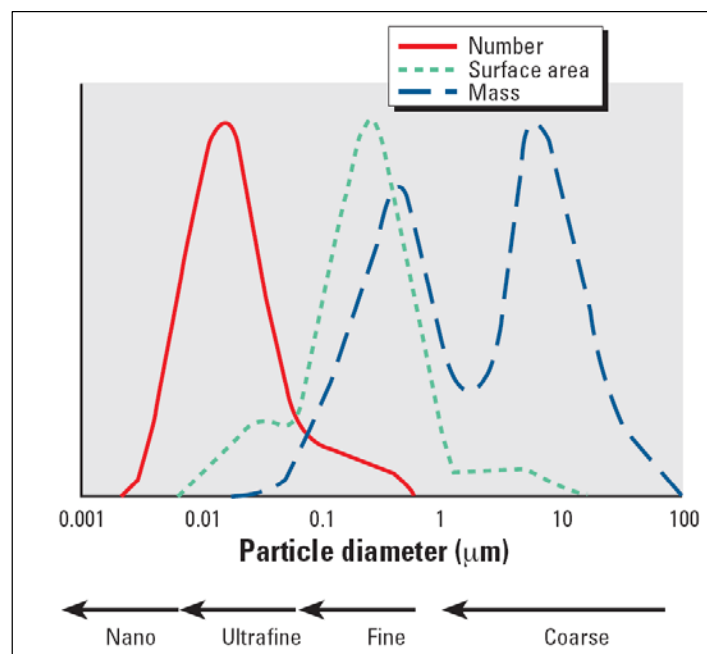
Particle microphysical properties being most relevant for adverse health effects are not yet identified. There is growing evidence that the particle surface area might act as an appropriate dose metric for particle-related health effects (Oberdörster et al., 2005; Stöger et al., 2006). Particle number is also considered an important parameter. On the other hand, particle mass is an inadequate measure since particle surface area associated with an aerosol of constant mass concentration can vary over several orders of magnitude, depending on the size of the particles.

Discussion is focusing on the question whether volatile and thus soluble particles may have a higher potential for hazardous effects than non-volatile and thus predominantly insoluble particles. Scientific literature does not provide a clear statement on this. Toxic compounds like polycyclic aromatic hydrocarbons are part of the volatile organic fraction, but they are mainly associated to insoluble carbonaceous combustion particles. Volatile nanometer-sized particles being composed predominantly of sulphuric acid-water and of condensed organic compounds are soluble and are expected to be dissolved in the humid environment of the human respiratory

tract so that they will have a much shorter residence time inside the lung than insoluble nanometer-sized particles which are removed very inefficiently from the lung. Because of these contradicting processes, the picture of potential hazardous particle compounds is still very unclear. Nevertheless, the large number of deaths and other health problems associated with particulate pollution was first demonstrated in the early 1970s (Lave and Seskin, 1973) and has been reproduced many times since. PM pollution is estimated to cause 22,000-52,000 deaths per year in the United States and 200,000 deaths per year in Europe (Mokdad et al., 2004).

It is becoming increasingly clear that the legislative limits for engines (Smoke Number or  $PM_{10}$ ) in terms of emitted mass are not a proper measure with regard to the health hazard. One spherical particle of 10  $\mu m$  in diameter has the same mass as 1 million particles of 100 nm diameter, but it is clearly much less hazardous, as it probably never enters the human body - and if it does, it is removed quickly. On the other hand, the surface of 1 million spherical particles of 100 nm diameter is 10.000 times larger than the surface of one particle of 10  $\mu m$  diameter. Figure 2-2 shows the number of particles per  $m^3$  of air (solid line) and the total surface of this aerosol per  $m^3$  of air (dashed line), when the mass concentration of this aerosol is kept constant at a representative value of  $1 \mu g / m^3$ . A particle mass density of  $1 g / cm^3$  is assumed. Proposals for new regulations exist in some countries, with suggestions to limit the particle surface area or the particle number.

Exhaust particles from combustion processes, such as emissions from automotive engines or gas turbines have a number concentration peak at about 20 – 60 nm in diameter, which also is the size of maximum lung deposition rate. Figure 2-3 demonstrates for a typical atmospheric aerosol that particles larger than 100 nm contain almost all particle mass, while for the same aerosol more than 90% of particles by number fall into the range of nanoparticles. Furthermore, toxic materials can be adsorbed on the particle surface of small particles with a high surface area. These nanometer-sized particles in high number concentrations can potentially carry toxic materials into the lungs (SMHI, 2006). There is recent evidence that the particle surface area might act as a threshold dose for acute lung inflammation (Stöger et al., 2006).



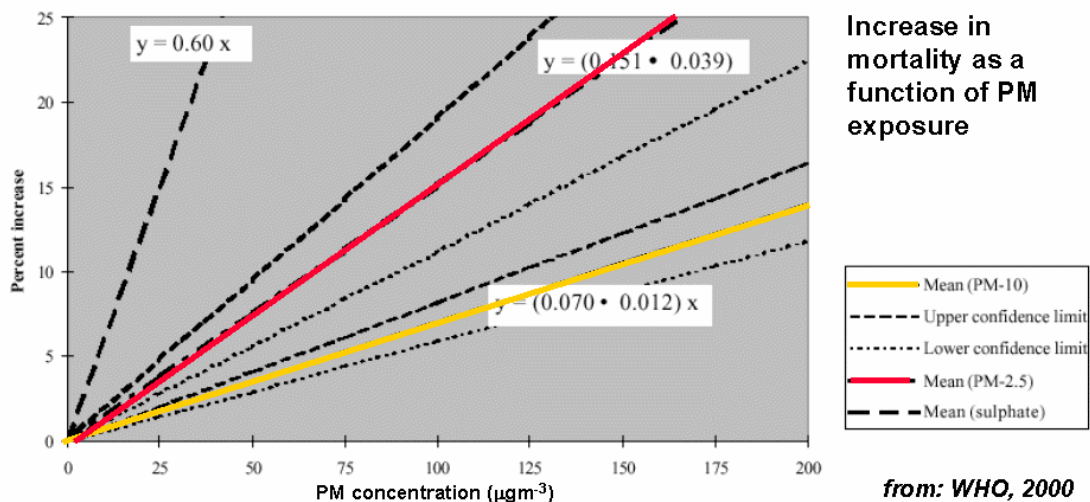
**Figure 2-3:** Number, surface, and mass size distribution for a typical atmospheric aerosol.

## 2.2 Current Limiting Values for Ambient Air

Current EU legislation regulates the mass of inhalable particles ( $PM_{10}$ ). In directives 1999/30/EC and 96/62/EC, the European Commission has set limits for  $PM_{10}$  in the air which are compiled in Table 2-1 together with respective values for other countries. However, recent findings indicate a stronger relation between health effects and fine particles  $PM_{2.5}$ , as shown in Figure 2-4. WHO has therefore recommended that guidelines for regulation of the mass of the fine fraction, i.e., of particles with diameter below  $2.5\ \mu m$  ( $PM_{2.5}$ ) shall be developed.

**Table 2-1:** Limit for  $PM_{10}$  in ambient air (R. Gardner, private communication)

State/Organisation	Particulate Size	Time-weighted average concentration in air	Averaging time	Comments
European Union	$PM_{10}$	$50\mu g/m^3$	24 hours	EU does not have standards for $PM_{2.5}$
		$40\mu g/m^3$	Annual	$20\mu g/m^3$ by 2010
United States	$PM_{10}$	$150\mu g/m^3$	24 hours	Primary & Secondary <sup>1</sup>
		$50\mu g/m^3$	Annual	Primary & Secondary
	$PM_{2.5}$	$65\mu g/m^3$	24 hours	Primary & Secondary
		$15\mu g/m^3$	Annual	Primary & Secondary
WHO				No guidelines values recommended for particulates#
Canada	$PM_{2.5}$	$30\mu g/m^3$	24 hours	By 2010 National Objective
British Columbia (as example)	$PM_{10}$	$50\mu g/m^3$	24 hours	Provinces have their own standards
Australia	$PM_{10}$	$50\mu g/m^3$	24 hours	
	$PM_{2.5}$	$25\mu g/m^3$	24 hours	
		$8\mu g/m^3$	Annual	
Brazil	Free Particulates <sup>‡</sup>	$150\mu g/m^3$	24 hours	Brazil also has primary & secondary standards*
		$50\mu g/m^3$	Annual	
	TSP <sup>2</sup>	$80\mu g/m^3$	Annual	Primary Standard
		$240\mu g/m^3$	24 hours	Primary Standard
		$60\mu g/m^3$	Annual	Secondary Standard
		$150\mu g/m^3$	24 hours	Secondary Standard
Japan	TSP <sup>2</sup>	$200\mu g/m^3$	1 hour	Japan only has standards for TSP
		$100\mu g/m^3$	24 hours	



**Figure 2-4:** Increase in mortality as a function of PM exposure.

However, the typical urban aerosol has no natural division at 2.5  $\mu\text{m}$ , rather it shows three peaks named as the coarse mode ( $\mu\text{m}$  particles), accumulation size mode (typical peak at 100-200 nm) and the nuclei size mode (peak at 10-20 nm), see for example Figure 2-3. The smaller the particles, the deeper they will penetrate into the respiratory system. Ultrafine particles (diameter <100 nm) enter the alveolar region.

### **2.3 Conclusion**

The question in defining a limiting value is whether there is a threshold below which no effects of the pollutant on health can be expected to occur in all people. Therefore, it has to be asked whether the evidence supports the concept of thresholds, i.e., concentrations below which effects are not observed, either in the general population or in selected susceptible populations of specific concern for particular pollutants.

The existence of a threshold implies that a specific guidelines value could be set at a level below which safety could be assured and a margin of safety incorporated into setting the level of the standard. In the absence of a threshold, evidence of exposure-risk or concentration-risk relationships is needed to identify levels for standards that provide an acceptable level of risk.

In responding to the question on thresholds, the WHO-WG noted the following (WHO, 2003):

- 1 Increasingly sensitive epidemiological study designs have identified adverse effects from air pollution at increasingly lower levels.
- 2 Thresholds differ depending on the outcome selected. Any threshold is a function of the endpoint chosen (death, diminished pulmonary function, or molecular changes), the nature of the responding population (from the most healthy to the most ill), as well as the time at which the response is measured (immediate vs. delayed or accumulated).
- 3 For some pollutants and adverse health effects, the population distribution of susceptibility may be such that effects are expected at low levels, even where current air quality standards are being met.
- 4 Observational (epidemiological) studies have limited statistical power for characterizing thresholds. Toxicological studies are similarly limited.
- 5 A lack of evidence for a health effect should not be interpreted as implying a lack of effect. ("Absence of evidence is not the same as evidence of absence".)
- 6 It is worth considering replacing the threshold concept with a more complete exposure risk function.

**It is concluded from recent scientific studies that the surface area of ultrafine particles has to be considered as key particle property related to adverse health effects. Next, particle number is as well correlated to health effects, while the total mass of particles is linked to health effects only for extremely high particle loads. Therefore, current legislation for limiting  $\text{PM}_{10}$  is not sufficient**

**A limiting value for aircraft engine emissions could be defined independent from potential adverse health effects, as the rationale should be that aircraft engines emit as few particles (in mass and number) as possible.**

**WHO recommends that complete exposure/concentration-response relationships for different health endpoints provide better information for designing effective strategies to reduce adverse effects on human health.**

## Chapter 3      Impact of Particles on the Atmosphere

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Pathways for the impact of particle emissions from aviation on the global climate are related to the fact that

- under certain conditions particles emitted from aircraft generate contrails;
- contrails may transform into persistent contrails and further into cirrus clouds, causing additional cirrus cloud cover compared to the unperturbed atmosphere;
- particles emitted from aircraft can generate additional cirrus clouds in otherwise cloud free but ice supersaturated regions of the atmosphere by initiating ice particle formation on the released particles;
- at sufficiently low temperatures,  $\text{HNO}_3$  is taken up by the surface of ice crystals resulting in a removal of  $\text{NO}_x$  from the influenced region of the atmosphere which impacts ozone chemistry;
- at sufficiently low temperatures, halogen compounds ( $\text{ClO}_x$ ,  $\text{BrO}_x$ ) may become activated on the surface of ice crystals and cause ozone destruction.

Anthropogenically caused cirrus clouds and contrails will influence the radiation balance of the global atmosphere and will thus contribute to global warming. Impacts on the ozone chemistry of the upper free troposphere and lowermost stratosphere (9 – 13 km altitude above sea level) in the main air traffic flight levels will also have an influence on global warming, since ozone is a very efficient greenhouse gas. An updated review of the impact of aviation on the global climate is currently prepared within the European Assessment of Transport Impacts on Climate Change and Ozone Depletion Project (<http://www.ssa-attica.eu>). The respective report is under preparation and will be published as a special issue of Atmospheric Environment.

As stated in the recent FAA document on the Current State of Research Coordinated under the PM Roadmap (FAA, 2007), today's state of science and technological knowledge on PM emissions is insufficiently mature to guide aircraft engine development for emissions reduction or to set specific policy objectives. In other words, we do not know at a sufficiently firm level whether or not particulate emissions from aircraft impact cirrus clouds and climate significantly.

Contrail formation processes and their link to particle emissions are well understood. The knowledge is extensively summarised in the IPCC Special Report on Aviation and the Global Atmosphere (IPCC, 1999). Effects of aircraft propulsion efficiency and fuel composition on contrail properties is summarised by Schumann et al. (2002). Major uncertainties arise from serious gaps of understanding in the parameters which govern contrail-cirrus evolution and in the role of particle emissions from aviation in cirrus formation.

In a recent publication on the potential role of black carbon (BC) particles and related particulate matter in cirrus formation (Kärcher et al., 2007), the authors state in the conclusions from their extensive modelling and laboratory studies, that

*...a critical discussion of laboratory experiments reveals that the ice nucleation efficiency of soot particles depends strongly on their source, and, by inference, on atmospheric aging processes. Mass and chemistry of soluble surface coatings appear to be crucial factors. Immersed soot particles tend to be poor ice nuclei, some bare ones nucleate ice at low supersaturations. However, a fundamental understanding of these studies is lacking, rendering extrapolations to atmospheric conditions speculative. In particular, we cannot yet decide which indirect aircraft effect scenario is more plausible, and options suggested to mitigate the problem remain uncertain.*

They attribute highest uncertainties associated with the impact of BC particles from aviation on cirrus cloud formation to

- 1 the lack of in-situ measurements of cirrus formation,
- 2 the lack of information on properties of ice supersaturated regions,
- 3 the impact of organic matter and sulphuric acid on the ice nucleation ability of combustion particles from aircraft engines,
- 4 the size distribution of particles in particular after pre-activation in contrails,
- 5 the separation of aerosol effects on cirrus formation from effects of atmospheric dynamics.

Related open key scientific questions are

1. Which particles form ice and why?
2. How large are ice supersaturated regions, can we forecast them?
3. Which particle properties influence ice nucleation ability?
4. Size is a crucial aerosol property for ice formation, how does it change after contrail processing?
5. Cirrus formation occurs during lifting of cold and humid air masses. Is vertical wind speed and atmospheric turbulence more important than the properties of potential ice nuclei?

At present, we know from observations that the ice-active particle population is mainly found in the size range above 100 nm in diameter (DeMott et al., 2003; Richardson et al., 2007). Although direct particle emissions from aircraft engines are much smaller in size, as is discussed later, particles may be transformed by atmospheric ageing processes to fewer but larger particles which fall into the ice-active size domain (Petzold et al., 1998). Nevertheless, the aerosol-cirrus interaction is predominantly based on particle number instead of particle mass in any case (see Kärcher et al., 2007, and references given there).

Model studies combined with in-situ observations also suggest that cloud modifications induced by aircraft BC particles could change the ice crystal number concentration at northern midlatitudes significantly (10–40% changes of annual mean zonal averages at main flight altitudes), provided that such BC particles serve as efficient ice nuclei (Hendricks et al., 2005). The presented results demonstrated that, based on the current knowledge on the ice-forming ability of particles, significant cirrus modifications by BC from aircraft cannot be excluded.

**In the very near future we cannot expect a robust and simple YES or NO answer to the question whether particles emitted from aircraft have a significant impact on cirrus clouds, heterogeneous chemistry, and thus on climate. However, research indicates that the climate effect of particle emissions is related closer to particle number than to particle mass.**

## Chapter 4      Particle Metrics and Methods

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### 4.1      *Particle Metrics*

Although there are still serious gaps of knowledge on the effects of particulate matter from aviation on climate and human health, aviation industry, rulemaking agencies and associated working groups and committees are encouraged to consider particulate matter emissions as an upcoming target for regulations in the near future. The joint agreement on accepted measurement methodologies for key particle properties should therefore be up high on the agenda of the involved parties.

Besides gaps in scientific understanding, part of the immaturity of this field arises from the missing common understanding of particle characteristics and their relation to available measurement techniques. The two key points of debate are

1. What is the appropriate measure for potential particle effects: is it particle mass, particle number, particle surface area, chemical composition or any combination of those?
2. What is volatile particulate matter and how is it related to different measurement approaches?

The present knowledge on the properties of particles emitted from aircraft engines can be summarised as follows:

1. Particulate matter emitted from aircraft engines at cruise conditions is composed of > 95% carbonaceous matter (organic and elemental carbon, partially referred to as soot) and < 5% sulphate and sulphate-associated water. The black carbon fraction amounts up to 70-80% of total carbon.
2. At low power conditions (idle, taxi), the organic carbon fraction contributes up to 90% to the total carbon, leaving only 10% by mass for black carbon. Total carbon contributes > 90% of mass at low thrust conditions. Fractions of sulphates and sulphate-associated water are < 10% for idle conditions and < 4% for thrust settings > 70%. Using low FSC fuel, the sulphate fraction is below the detection limit of the applied methods.
3. Volatile particulate matter consists of sulphate and sulphate-associated water, and of organic compounds which volatilize at a certain threshold temperature which requires prior definition. If the volatilisation temperature during sampling is set to approx. 250°C; organic compounds emitted at cruise conditions are mostly thermodynamically stable while sulphate and sulphate-bound water volatilizes. For low power conditions there exists an organic fraction which volatilizes below this temperature. Its mass fraction and chemical composition, however, are almost unknown.
4. The number-weighted average size of combustion mode particles emitted from a gas turbine ranges from 25 nm in diameter for modern-type engines to approx. 100 nm for 1960s technology. Respective mass-based average sizes are of the order of 50 nm to 200 nm.
5. Number and size of the non-volatile combustion mode particles forming during combustion are influenced only at a minor level by the fuel sulphur content.
6. Nucleation mode particles form when supersaturation of the condensable species occurs, i.e., when the hot plume mixes with ambient air, or when the hot exhaust gas is

strongly diluted during sampling, i.e., particle formation depends on sample treatment and fuel sulphur content. In general, nucleation mode particles form downstream the engine exit nozzle plane.

7. Nucleation mode particles forming in the cooling gas are for almost all conditions (cruise, advected plume, exhaust gas sample) smaller than 20 nm in diameter if plume ages < 5 s are considered.
8. A prominent nucleation mode is found in plumes advected from engines across the airfield, indicating that secondary aerosol particles composed of sulphuric acid and low-volatility organic species form rapidly within the plume as it expands and cools.
9. The number fraction of volatile nucleation mode particles is highly variable and depends on fuel properties, engine operation conditions and sample treatment.

The understanding of particle characteristics emitted at cruise conditions is well developed while the transformation of combustion particles once emitted from a cruising aircraft into the size range favourable for ice-nucleation is still open. Particle characteristics in emissions from low-power conditions are known in terms of size and mass. The pictures for cruise emissions and emissions during ground testing are coherent. The non-volatile particle fraction shows reasonable correlation with engine technology.

The key unknowns at present are

1. the mass fraction of volatile components at low power conditions;
2. the key organic compounds involved in volatile particle growth after nucleation via sulphate-water cluster formation, including their potential toxicity;
3. the connection between volatile particle nucleation and growth at one hand and engine technology on the other hand.

An essential prerequisite for the agreement on a particle measurement method is the identification of relevant particle properties to be measured. Key parameters are mass concentration, number concentration, and size distribution. Any other properties like, e.g., surface density and surface distribution can be calculated from these basic properties using reasonable assumptions on particle mass density and particle shape. An agreement is required to determine which fraction of the exhaust aerosol becomes subject to regulatory rule. From recent studies it is known that refractory carbonaceous particles make up the most stable fraction of the exhaust aerosol while the volatile nanoparticle mode is highly variable and depends strongly on the sampling conditions. When setting up a new regulatory standard for the measurement of particulate emissions from aircraft turbine engines, the conditions for sampling and considered particle size ranges have to be described very carefully to define the object of measurement unambiguously.

#### **4.1.1 Mass**

##### *Emissions at Cruise Conditions*

The chemical composition of the particulate matter emitted from aircraft engines under cruise conditions can be summarised as carbonaceous material containing elemental carbon and organic matter > 95% and water-soluble inorganic components as sulphates < 5%. Most relevant for measurement purposes is the thermal stability of the carbonaceous matter.



According to their thermal stability, various fractions of the deposited particulate matter are oxidised at different temperatures: semi-volatile organic compounds evaporate at  $T < 250^{\circ}\text{C}$ , non-volatile organic compounds from  $250^{\circ}\text{C}$  to approx.  $430^{\circ}\text{C}$  and black or elemental carbon burns off at  $T > 430^{\circ}\text{C}$  (Petzold et al., 2005). The thermal analysis of the carbonaceous matter demonstrates the insignificance of organic matter with respect to the volatile fraction if volatile particulate matter is related to volatilisation temperatures of approx.  $250^{\circ}\text{C}$ . Discussing volatile particulate is then exclusively related to sulphuric acid particles which make up less than 5% of the total particle mass.

### *Emissions at Ground Conditions*

At engine operation conditions relevant for air quality monitoring purposes, the elemental carbon fraction of total carbon varies from 10% at idle to > 70% for power settings > 30%, approaching nearly 100% close to take-off, clearly indicating the large contribution of organic matter to particle mass at idle conditions (Petzold and Schröder, 1998; Herndon et al., 2007a,b). Burning high sulphur fuel (FSC = 3000 ppb) results in a sulphate mass fraction of 7% at idle, while for low FSC, sulphate was below the detection limit (Petzold and Schröder, 1998). For approx. 70% thrust, the sulphate fraction decreases to 4% for high FSC fuel and remains below the detection limit for low FSC fuel. On the other hand, formaldehyde as the key tracer for oxidised hydrocarbons amounts up to  $1.2 \text{ g kg}^{-1}$ , acetaldehyde to  $0.33 \text{ g kg}^{-1}$  at low power setting (Knighton et al., 2007). Emissions of gaseous hydrocarbon compounds are thus dominating the emissions of carbonaceous matter at idle. In contrast to cruise conditions, the separation of volatile and non-volatile organic matter is a critical issue for engine emissions at ground conditions

## **4.1.2 Number**

### *Emissions at Cruise Conditions*

The number of particles emitted from various subsonic aircraft engines or formed in the exhaust plume per unit mass of burned fuel varies from  $2 \times 10^{14}$  to  $3 \times 10^{15} \text{ kg}^{-1}$  for non-volatile particles (mainly black carbon or soot) and is of the order  $2 \times 10^{17} \text{ kg}^{-1}$  for all particles including volatile particles >3 nm in diameter at plume ages of a few seconds (Schumann et al., 2002, and references given there). Concerning the carbonaceous particulate matter which forms the non-volatile fraction of the emitted particles, emission indices with respect to black carbon vary from 0.7 down to  $0.01 \text{ g kg}^{-1}$  in terms of mass and from 20 down to  $2 \times 10^{14} \text{ kg}^{-1}$  by number, strongly depending on engine technology or certification year, respectively. The emission factors determined during the PARTEMIS combustors tests at simulated cruise conditions for so-called “old” and “modern” operation conditions fit well into the scheme of data from airborne measurements. The difference between “old” and “modern” operation conditions were an increase in combustor inlet temperature by 200K and an increase in combustor inlet pressure from 7.05 to  $8.2 \times 10^5 \text{ Pa}$  (Wilson et al., 2004).

Concerning the formation of volatile particles in the expanding and cooling plume downstream the nozzle exit plane, two influencing factors have to be considered. Results from the PARTEMIS study (Petzold et al., 2005, and references given there) demonstrate the impact of combustion particles present in the exhaust gas and the fuel sulphur content. The fuel sulphur content influenced particles with sizes smaller than  $0.02 \mu\text{m}$ . The emission of particles in the combustion particle mode with diameters  $> 0.02 \mu\text{m}$  was not affected by the fuel sulphur content while the emission of ultrafine particles with diameters  $< 0.01 \mu\text{m}$  depended crucially on the fuel sulphur content. Another factor quenching the nucleation of new particles in a cooling exhaust gas is the presence of a large aerosol surface area which acts as a sink for the condensable gases emitted from the combustor/engine. A model study on volatile particles formation during

PARTEMIS demonstrated that the particles form at the dilution point in the sampling line when the hot exhaust gas is mixing with cold dilution air (Vancassel et al., 2004).

### *Emissions at Ground Conditions*

First experimental test studies on engine particulate emissions operated from idle to taxi were performed in 1996 during the DLR co-ordinated SULFUR 4 studies using a Rolls-Royce/SNECMA M54h Mk501 engine (Petzold and Schröder, 1998; Petzold and Döpelheuer, 1998). Extensive studies were conducted during the NASA EXCAVATE study in 1999 (NASA, 2005) and during the NASA APEX study on a CFM56-2C1 engine (NASA, 2006; Wey et al., 2007) in 2004 and during follow-up measurements APEX-2, APEX-3 and airport studies. The data set on aerosol properties emitted during ground operation developed in the past 5 years. The results from these experiments are available as NASA Reports (NASA, 2005; NASA, 2006), and most recently as series of papers in the Journal of Propulsion and Power (Wey et al., 2007; Yelvington et al., 2007; Lobo et al., 2007; Knighton et al., 2007) and as a set of extended abstracts of the Conference on Transport, Atmosphere, and Climate, held in Oxford on 26-29 June 2006 (Herndon et al., 2007a; Herndon et al., 2007b; Whitefield et al., 2007; Hagen et al., 2007).

All experiments showed consistently that engines emit high concentrations of organic aerosols at low power settings. At idle, the aircraft emit much higher levels of organic aerosols than of black carbon particles. The BC mass emission indices increase significantly in going from idle to cruise power. Emissions were lowest at medium power settings. Observed increases range from a factor of 20 for a smaller old-technology engine (Petzold and Döpelheuer, 1998) to a factor of 40 for modern clean engines of the CFM56 family (Lobo et al., 2007). Absolute numbers depend on the engine type. Typical values found during the studies of CFM56 engines using standard fuel are  $< 60 \text{ mg kg}^{-1}$  at power settings  $\leq 70\%$  and  $> 200 \text{ mg kg}^{-1}$  at power settings  $> 85\%$ . Respective values for the old-technology engine are  $60 \text{ mg kg}^{-1}$  at idle and  $330 - 350 \text{ mg kg}^{-1}$  at power settings  $\geq 70\%$  power (Petzold and Döpelheuer, 1998).

Emission indices in terms of particles by number are  $< 1 - 5 \times 10^{15} \text{ kg}^{-1}$  for total particles and  $0.16 \text{ to } 3 \times 10^{15} \text{ kg}^{-1}$  for non-volatile particles. Emissions of non-volatile particles are greatest at idle and take-off thrust settings and are at a minimum at power levels corresponding to approach. The variation of the emission of non-volatile particles is strongly correlated to the variation of BC mass emissions. Observations in the exhaust of other engines during APEX 2 fit into this picture.

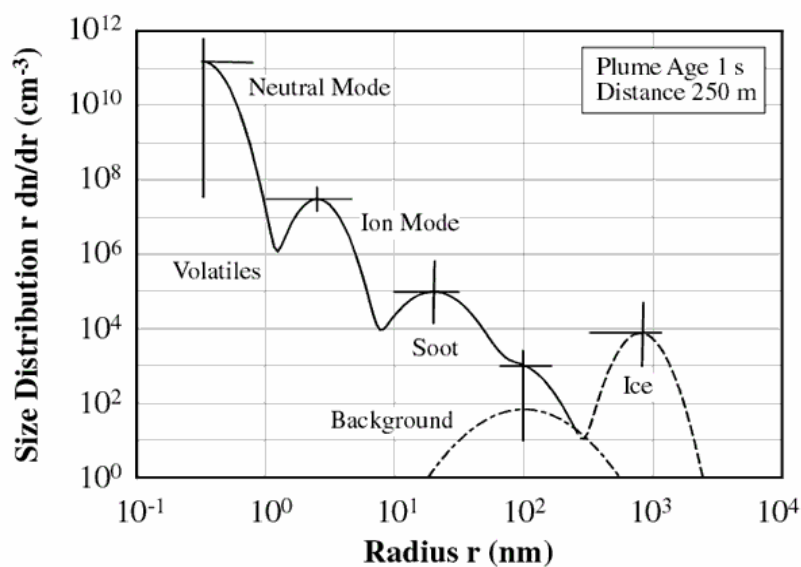
Moving the sample extraction point from the engine exit plane downstream to a distance of 10 – 30 m significantly increases the number of volatile particles, i.e., total particle emission indices were typically a factor of 10 higher at 25 to 35 m than at 1 m downstream of the exhaust plane, indicating that significant numbers of new particles form within the exhaust plume as it cools and dilutes. The concentration of sulphate aerosol increased considerably as sampling took place progressively further downstream of the exhaust plane, suggesting that sulphate particles form and undergo rapid growth within aircraft exhaust plumes. At larger sampling distances, the influence of the engine operation conditions on the emission of particles by number is significantly reduced.

### 4.1.3 Size Distribution

During combustion small non-volatile carbonaceous particle agglomerates are formed with typical diameters of 30–60 nm and geometric standard deviation of the size distributions of about 1.55–1.70 (Hagen et al., 1996, 1998; Petzold et al., 1999, 2003; Popovicheva et al., 2004; Petzold et al., 2005a; Delhaye et al., 2007). Details are engine dependent (Brundish et al., 2007; Dakhel et al., 2007). Larger agglomerates may grow up to sizes larger than 100 nm in diameter. Like any other carbonaceous particles forming during incomplete high-temperature combustion processes, particles emitted from gas turbines are fractal agglomerates which are made up from small spherical primary particles. Transmission electron microscopy analyses from CMF 56 type engine emissions show spherical primary particles of  $10.2 \pm 0.1$  nm in diameter (Delhaye et al., 2007). The primary particles clearly exhibit a spherical shape made out of concentric, size-limited, graphene layers arranged in an “onion-like” structure (Popovicheva et al., 2000, Delhaye et al., 2007).

Plume measurements (Schumann et al., 2002) show a clear trend of modern engines emitting smaller particles (CFM56 types: modal diameter  $\approx 25$  nm) than older engines (PW JT3D-3B: modal diameter  $\approx 100$  nm). During the combustor test rig studies of PARTEMIS (Petzold et al., 2005a), mean modal diameters are 37–41 nm for old engine and 39–45 nm for modern engine conditions. The geometric standard deviation of the particle number size distribution is 1.69 (1.66–1.73) in both cases. As was demonstrated during PARTEMIS, the turbine section of the engine does not influence the microphysical properties of the combustion particles, i.e., particle number, size and black carbon mass properties are determined mainly by the conditions inside the combustion chamber.

Figure 4-1 shows the approximate size distributions (solid line) versus radius for volatile particles (neutral and chemi-ion modes) and soot (primary and secondary modes), when no contrail is produced by the aircraft. If a contrail is formed, these size distributions change (not shown), and ice particles are created (dashed line). Size distributions of soot and ice particles have been measured in situ. Mean sizes and numbers of ion mode particles have been deduced from particle counter measurements, and corresponding size distributions of volatile particles have been inferred from simulation models.



**Figure 4-1:** Size distribution of various aerosol types present in young jet aircraft exhaust plumes (adapted from Kärcher, 1998)

Bars indicate the approximate range of variability resulting from variations of the fuel sulphur content, engine emission parameters, and ambient conditions, as suggested by evaluation of current observations and modelling studies. An approximate background aerosol size distribution is included for comparison (dot-dashed line).

Particle sizes are smallest at idle (diameters <20 nm) and largest at take-off (diameter <40 nm) (Wey et al., 2007). Non-volatile particles emitted by the engine exhibited a log-normal size distribution that peaked between 15 and 40 nm, depending on engine power. During APEX 2, values between 25 nm (7% thrust) and 41 nm (85% thrust) were found for the non-volatile combustion particle mode. Samples collected 30 m downstream of the engine exit plane exhibited a prominent nucleation mode, indicating that secondary aerosols composed of sulphuric acid and low-volatility organic species formed rapidly within the plume as it expands.

The mode at higher power settings coincides with the combustion particle mode observed during PARTEMIS and the cruise studies, while the particles at low power settings are obviously dominated by particles from gas-to-particle conversion processes according to their small size and to the low fraction of BC mass. The size distributions measured for the non-volatile fraction during APEX 2 coincide well with the combustion mode measured during PARTEMIS (Petzold et al., 2003).

The analysis of particulate matter in plumes advected from taxiing aircraft or from aircraft at take-off revealed a mono-modal size distribution at taxi with the mode centred at 16 nm at a bi-modal size distribution at take-off with modes centred at 10 nm (nucleation mode) and approx. 35 nm (combustion particle mode) (Whitefield et al., 2007). Number and mass emission indices were  $38 \times 10^{15} \text{ kg}^{-1}$  (taxi) and  $12 \times 10^{15} \text{ kg}^{-1}$  (take-off), and  $180 \text{ mg kg}^{-1}$  (taxi) and  $360 \text{ mg kg}^{-1}$  (take-off). The values are higher than those observed during the engine tests which may be caused by humidity uptake of the particles during advection to the sampling site.

#### 4.1.4 Conclusion

Conclusions from engine testing in test rigs and exhaust measurements at under-wing engines reveal that:

1. The non-volatile combustion particle fraction is correlated to engine technology. This fraction is stable in the sense that it does not depend on sample handling as long as particle coagulation and losses to the sample line walls are avoided. This fraction also does not undergo strong transformation processes in the near-field plume which is relevant for air quality considerations.
2. Particulates are composed of light-absorbing black carbon, accessible by simple and robust optical methods, by organic compounds of variable thermal stability, and to a small extent of sulphates originating from the fuel sulphur. The total mass can be measured to the needed accuracy only with labour-intensive gravimetric methods. On-line particle-mass sensitive methods show a strong variation in determined mass emissions and are not yet mature for the considered application. On-line methods for measuring the light-absorbing fraction are robust and stable, but focus exclusively on the key component black carbon.
3. The consideration of volatile particulate matter emissions suffers from the lack of knowledge of the contributing compounds. Volatile particulate mass contributes substantially to total particulate mass only for low to medium thrust conditions which, however, are most relevant for airport air quality issues.
4. There is a lack of understanding what metrics should be used to characterise potential hazardous effects from particulate matter from aircraft. A similar statement holds for the climate effect of aircraft-related particulate matter.

**Potential precisely accessible particle properties are particle number, particle size and particle mass. Particle number and size, because the scientific debate on health effects of particulate matter focuses on insoluble nanometer-sized particles by number or surface. Particle mass, because current air quality regulations focused on particle mass.**

## **4.2 Measurement Methods**

The following chapter is based on the work prepared by the SAE E-31 committee within the past years and published in the Aerospace Information Report AIR 5892 on Non-Volatile Exhaust Particulate Measurement Techniques (SAE, 2004) and related Technical Annexes which are in preparation for publication.

Non-volatile particulate matter measurement methods can be divided into two general approaches: measurement of the total particle mass and measurement of size and number density of particles. The first approach includes methods that measure the total mass of emitted particles without distinguishing size or number of particles emitted. One technique samples the exhaust stream and collects particulate matter on a filter, which is analyzed for the collected particle mass. Another technique probes the exhaust flow optically to quantify scattering material in situ without requiring the exhaust to be sampled and transported to the measurement system. Other techniques use oscillating microbalances, which measure particle mass based on the change in the natural frequency of the vibrating element as particles are collected. Current regulatory interest in stationary source particle emissions is directed toward total mass measurements and the total mass measurement techniques are discussed in the context of this regulatory interest.

The second general approach distinguishes the size and number of individual particles. The techniques for measuring number and size distribution have been used extensively in atmospheric research and have been refined for use in measuring aircraft engine exhaust. This type of measurement provides a number density and a particle size distribution derived from a sampled exhaust stream. The measurement of these parameters can be used, with a value of particle density and information on the particle morphology, to estimate the total mass of the particles. This approach offers considerably more information about the emitted particles, but it comes at the expense of a more complex and costly measurement system.

### **4.2.1 Mass Measurement**

Measuring the mass of emitted particulate matter may be performed in several ways, as summarized in Table 4-1. Filter methods are the oldest and most well known but are being replaced by optical methods that are non-intrusive and provide immediate results wherever possible. Microbalance methods and real-time optical methods also provide immediate results, however, as any filter method, they require extractive sampling.

SAE E 31 has proposed the selection of Gravimetric Analyses and Microbalance methods for total mass measurement and optical absorption photometry for black carbon measurement. The recommendation is based on an in-depth analysis of method maturity, accuracy and precision on one hand and on the appropriateness of the measured parameter on the other hand. Details of this method assessment will be published as a Technical Annex to the SAE Aerospace Information Report AIR 5892.

Gravimetry uses time-averaging methods to sample the emitted particles on appropriate fibrous filters or membrane filters. Since a considerable fraction of aircraft engine exhaust particles is smaller than 0.1  $\mu\text{m}$  in diameter, filter materials of high sampling efficiency in this size range are required. Common practice is to use glass fibre, quartz fibre, or Teflon filters for exhaust particle

sampling that show a filtration efficiency of > 99% in the relevant size range. Applying gravimetry, the sampled particulate matter is analyzed gravimetrically by weighing the filter before and after loading, which yields the total particle mass. For mass concentration measurements, also the sampled volume has to be recorded precisely.

Potential on-line methods for the determination of total particulate matter mass emissions are the tapered element oscillating microbalance (TEOM) and the quartz crystal microbalance (QCM). Both these methods have been used successfully in similar evaluations of automotive internal combustion engines, with the TEOM also recently introduced for measurements in the aerospace industry. In the TEOM instrument, a hollow vibrating tube equipped with a small in-line filter is installed inside a heated chamber. The change in harmonic oscillation measured by a sensitive frequency counter over a prescribed averaging time is proportional to the additional PM mass collected on the filter. The TEOM mass values must be corrected for pressure loss across the filter and total sample flow through the TEOM. In general, the TEOM has been shown to correlate reasonably well to the total filter method as determined over extended averaging periods, but it has high instrumental noise making shorter term measurements problematic. The harmonic oscillator principle used in the QCM is similar to the TEOM except that the collected PM is deposited on the crystal element itself using an electrostatic precipitator.

Aerosol absorption photometry analyses the modification of filter optical properties such as transmittance or reflectance caused by the particles deposited on the filter matrix. Optical absorption methods are highly suitable for the measurement of combustion particles because the key component black carbon is a very efficient absorber of light in the visible spectral range. Since total particulate matter contains also a fraction of non-absorbing aerosol components, absorption photometry cannot be used for the measurement of total particle mass. However, black carbon makes up the largest and most stable fraction of aerosol components in the exhaust of a gas turbine engine. Besides its specific sensitivity to the main combustion product black carbon aerosol absorption photometry has the further advantage of being operated continuously such that time-resolved recordings of particle emissions are possible.

**Table 4-1:** Methods for the Measurement of Particle Mass Concentrations in Aircraft Engine Exhaust (SAE AIR 5892)

Measurement Method	Measurement	Analysis
Gravimetric analysis	Total particulate matter, total mass	off line, filter samples
Microbalance	Total particulate mass	on-line, extractive
Combustion of filter samples and CO <sub>2</sub> detection	Total carbonaceous mass, TC	off line, filter samples
Combustion analysis including OC/EC separation	Organic carbon (OC), elemental carbon (EC), OC + EC = TC	off line, filter samples
Optical absorption photometry	Black carbon (BC), BC $\cong$ EC	on-line, filter samples time resolution $\geq$ 1 min
Laser induced incandescence	Black carbon (BC)	on-line, in situ
Transmissometry	Opacity	on-line, in situ
Light scattering	Forward scattering	on-line, in situ

#### 4.2.2 Measurement of Size and Number Density

Physical characterization of particles from gas turbine engine exhaust can be achieved with the use of commercially available particle size and number density measurement systems. These instruments are widely accepted by industry, academia, and government research organizations. They can be operated as laboratory or mobile systems under the difficult conditions encountered near the exit planes of combustors or turbine engines. These instruments provide real-time or near real-time results for size distribution and on-line data to assess particle loading. The concerns of sample mishandling and extensive time delays associated with off-line, post-test analysis have been eliminated. With this extractive sampling technique, special consideration must be given to the sampling systems to minimize modification of the exhaust particles as they are transported to the diagnostic instrumentation.

Measurement of the physical characteristics of combustion particles from aircraft turbine engines and research combustors requires that the diagnostic instruments be designed to sample from a wide range of pressure ( $1 - 62 \times 10^5$  Pa) and temperature conditions (220 K to 2400 K). They must be capable of measuring particle number densities (number of particles per unit volume of exhaust gas) that range from background levels as low as  $0.5 \text{ cm}^{-3}$  to exhaust emission levels as high as  $1.0 \times 10^9 \text{ cm}^{-3}$  and particle diameters ranging from a few nanometers ( $\sim 3 \text{ nm}$ ) to  $>10 \text{ }\mu\text{m}$ . The particle number density in turbine engine exhaust is determined by using one or multiple condensation nuclei counters (CNC). These counters provide real-time particle number density measurements and a means of rapidly assessing particle emissions from a combustion source.

In a CNC, a particle-laden sample is passed through a heated saturator where an alcohol (usually n-butanol) evaporates into the sample stream and saturates the flow. The saturated sample passes through a cooled condenser where the alcohol condenses on the particles to generate larger particles (droplets) that can be counted with an optical detector. The detector counts the particles in either the single-pulse mode for number densities of less than  $10^4$  particles per  $\text{cm}^3$  or in the photometric mode (light scattering) for particle number densities of  $10^4$  to  $10^7$  particles per  $\text{cm}^3$ . The sample entering the counter is usually diluted with particle-free air that is added to the sample flow at or within a few centimeters of the probe tip. This dilution minimizes particle-to-particle interaction or coagulation, respectively, and gas-to-particle conversion via particle nucleation and condensation of vapours on particle nuclei, and prevents the saturation of the PM instrumentation. The CNC data has to be corrected for this dilution to obtain the actual particle concentration in the exhaust gas.

A differential mobility analyzer (DMA) in combination with a condensation nuclear counter can be used to determine particle size distributions in turbine engine exhaust. An exhaust sample is passed through a bipolar ion charger that imparts a Boltzmann charge distribution to the particles. The charged and neutral particles enter the DMA, which contains a high voltage rod charged to provide a precise negative potential. The particles are separated or classified by size according to their mobility in the electric field. By changing the voltage in the DMA, a size spectrum is acquired. Bipolar charger/DMA/CNC systems are commercially available and can measure particle sizes from 2-1000 nm in diameter using analyzers with different ranges. The size distribution for larger diameter particles ( $>300 \text{ nm}$ ) is determined with an optical particle counter (OPC) that uses light scattering techniques.

The direct measurement of particle size distribution and number density allows the determination of the mean geometric diameter and particle distribution half width. In addition, emission indices with respect to particle number, surface area, and mass can be computed. The particle surface area and mass require assumptions of morphology and density, which may be a source of error in mass calculation.

## Chapter 5 Benchmarking for a Limiting Value

### 5.1 Comparison with Approaches for Vehicles

In December 2005, the European Commission presented a long-awaited proposal for 'Euro 5' emission standards for cars and vans (COM(2005)683 final). Member States and the European Parliament are considering adding a 'Euro 6' step to the proposal, in order to achieve long-term certainty and stability.

#### How have specific PM emissions of diesel passenger cars evolved in the past?

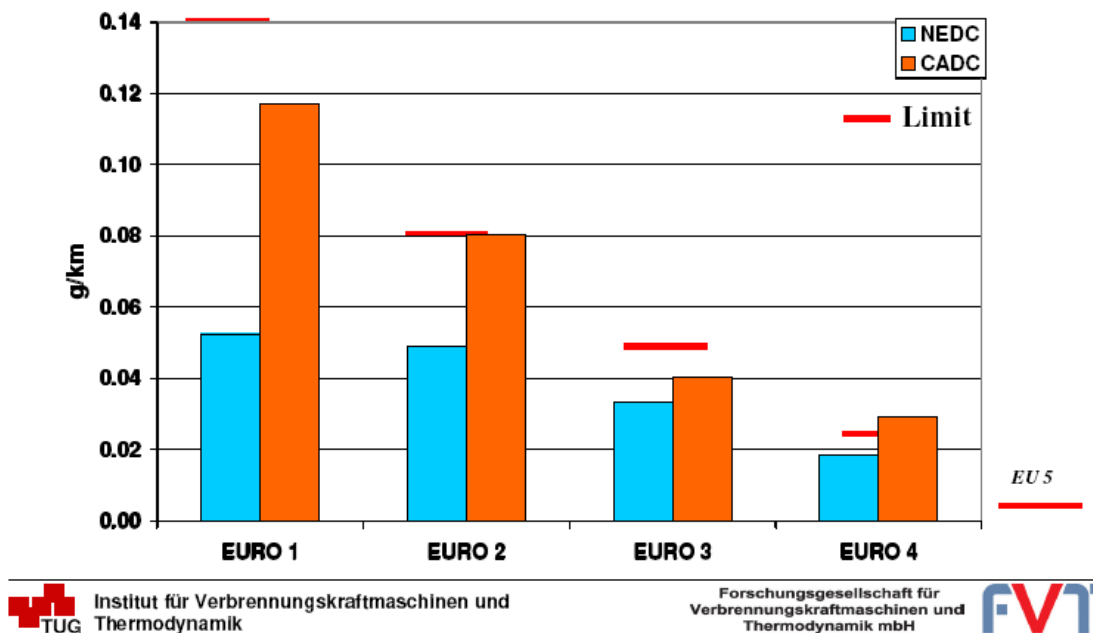


Figure 5-1: Differences between PM emission limits for diesel cars (Hausberger, 2006)

Figure 5-1 shows emissions measured under the official test cycle (NEDC) and emissions measured under a test cycle designed to reflect 'real world' driving (CADC). From Figure 5-1 it can be concluded, that PM emissions have been impacted by the regulation.

For vehicles, the limited and thus the measured particle property is the mass of particulate matter per kilometre. However, there is ongoing activity to include the particle number, too. A standard for particle numbers should be in place ultimately by 1 January 2010. Such a standard is necessary to ensure that manufacturers do not choose technical solutions to meet the PM standard that allow through large amounts of ultrafine particles – that weigh almost nothing, but have damaging health effects. An example is an open particle filter. Diesel cars with filters do not show a correlation between PM mass and PM numbers, i.e. some filters eliminate ultrafine particles much better than others.



In order to include particle number in the certification process, a Particle Measurement Programme (PMP) has been set up (ECE, 2007). The objective of the PMP was to develop and demonstrate new particle emissions measurement methods suitable for use in a type approval environment to supplement or replace the existing particulate mass measurement. The aim was to deliver measurement methods with improved sensitivity at low particle emissions levels (e.g. those delivered by wall-flow diesel particulate filter equipped engines/vehicles). These methods should enable setting of limit values to ensure adequate control of emissions of ultrafine particles.

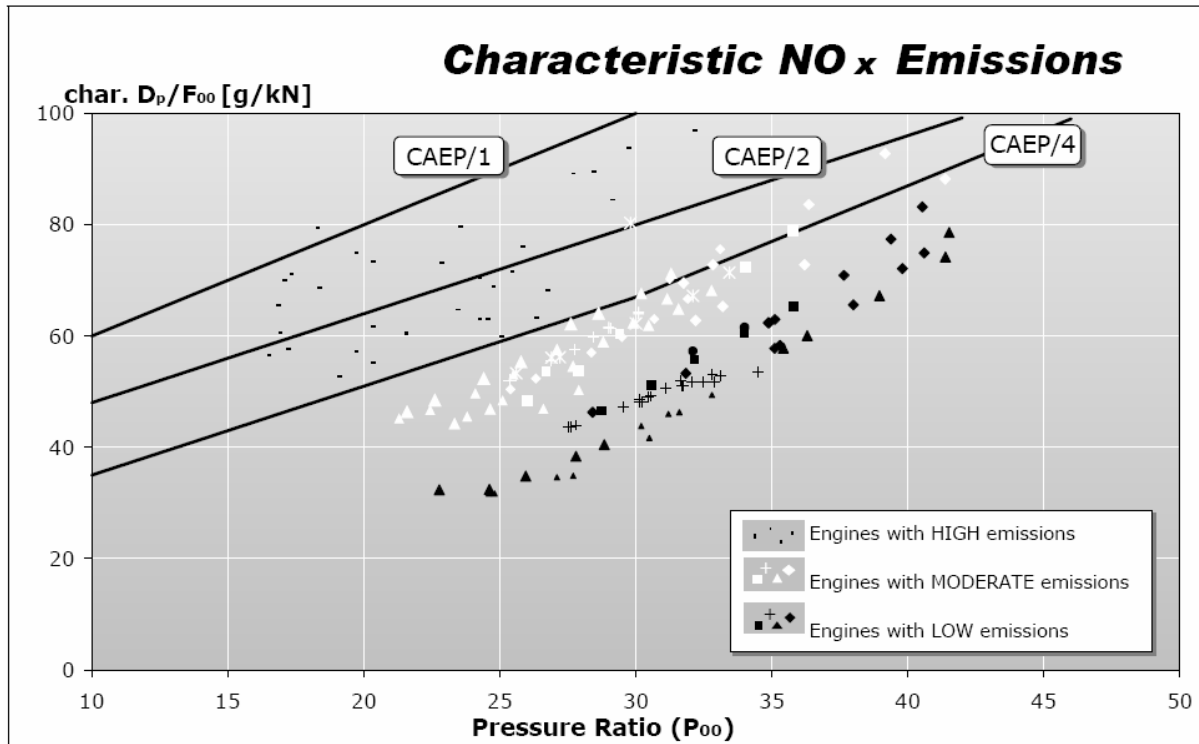
**The PMP validation exercise has demonstrated that the particle number measurement method is a far more sensitive indicator of particle emissions performance than even the revised particulate mass measurement. Indeed particle number is sufficiently sensitive to indicate changes in the fill state of a Diesel Particle Filter following regenerations. There is no evidence that the mass method is sensitive enough to indicate this.**

## **5.2 Approaches for other Aircraft Emissions**

Aircraft are required to meet the engine certification standards adopted by the Council of ICAO. These are contained in *Annex 16 — Environmental Protection, Volume II — Aircraft Engine Emissions to the Convention on International Civil Aviation*. These were originally designed to respond to concerns regarding air quality in the vicinity of airports. As a consequence, they establish limits for emissions of oxides of nitrogen ( $\text{NO}_x$ ), carbon monoxide, unburned hydrocarbons, for a reference landing and take-off (LTO) cycle below 915 metres of altitude (3 000 ft). There are also provisions regarding smoke and vented fuel. ICAO has set limitations on emissions at their sources (stringency) on the basis of the best achievable technology by all manufacturers.

Limiting particle emissions is, therefore, an additional measure to other pollutant emissions. As a consequence, a limiting value for particles should follow in principle the approach used for other emissions. As all other emissions, it has to address the rated output of the aircraft engine. The ICAO regulatory parameter for gaseous emissions is expressed as the mass of the pollutant emitted during the landing/take-off (LTO) cycle divided by the rated thrust (maximum take-off power) of the engine ( $D_P/F_{00}$ ).

Moreover, for  $\text{NO}_x$ , the limit is determined by the pressure ratio of the engine, as shown in Figure 5-2. ICAO is currently considering alternative parameters on which to base future high altitude emissions controls, taking into account trends in emissions reduction technology, as well as the performance of the whole aircraft and its productivity. Particular attention is being given to  $\text{NO}_x$ . In the case of  $\text{CO}_2$ , it has been decided not to develop an ICAO standard, since  $\text{CO}_2$  production is directly related to fuel consumption and there is already intense economic pressure to keep fuel consumption to a minimum and, in addition, there would be significant difficulties in designing a certification condition.



**Figure 5-2:** Characteristic NO<sub>x</sub> Emissions (Hotes, 2002)

### 5.3 Potential Nature of a Limiting Value for Particulate Matter

The comparison with the vehicle approach results in two conclusions:

- (1) A limiting value will very likely have an impact on the technological evolution, as the industry is forced to meet new limits.**
- (2) The automotive industry increasingly focuses on particle numbers rather than particle mass.**

Concerning the nature of a limiting value, the principle of characteristic emission indices as it is used with other gaseous emission should also apply for particles. This means,

- (3) A limiting value will have to be related to the rated thrust, irrespective of the measured variable, i.e. mass and/or number.**

As there are no reliable information that would allow for a more detailed definition of a limiting value, no further dependencies with other variables can be concluded, as it is, e.g., the case with NO<sub>x</sub> and the pressure ratio of the engine. See Figure 5-2 for details.

## Chapter 6      **Fleet Development and Possible Production Cut-off**

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### **6.1      *Rationale***

As described in the preceding chapters, at the time of drafting this paper, no adequate measurement procedure was agreed upon and, thus, no definite limiting value could be defined. As a consequence of this, the impact of a possible limiting value on fleet development cannot be assessed.

However, it is commonly agreed that particle emissions are, among others, related to the total fuel burnt in an engine per unit of time. This holds true for both mass and number of particles, regardless of other factors that might impact the particle emission index in terms of size distribution, the composition of particles, or other particle properties.

With this assumption, a preliminary estimate on the future development of particle emissions can be given. This is based on the fact that future engine technology will aim at a reduction of specific fuel consumption and, as a consequence, on a reduction of particle emissions, too. Therefore, the following chapter will describe how the worldwide emitted particle emission would develop, taking into account both the introduction of new engine technology and the expected growth in traffic numbers, the first leading to a reduction, and the latter to an increase in overall emissions.

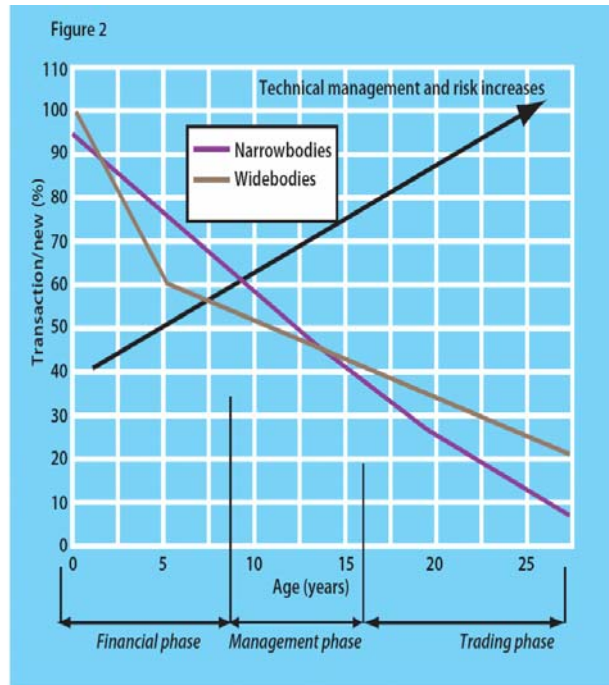
### **6.2      *Engine Life Cycle and Fleet Development***

Typically, an engine is designed to operate for at least 25 to 30 years. After this period, maintenance costs and the risk of an engine failure increase. Figure 6-1 shows engine values and life cycle over time. A new limiting value could only apply to engines certified in the future. This means for the impact of a limiting value that at the time of its introduction only a very small proportion of engines will be designed to comply with such a limiting value. Over time, this proportion will increase.

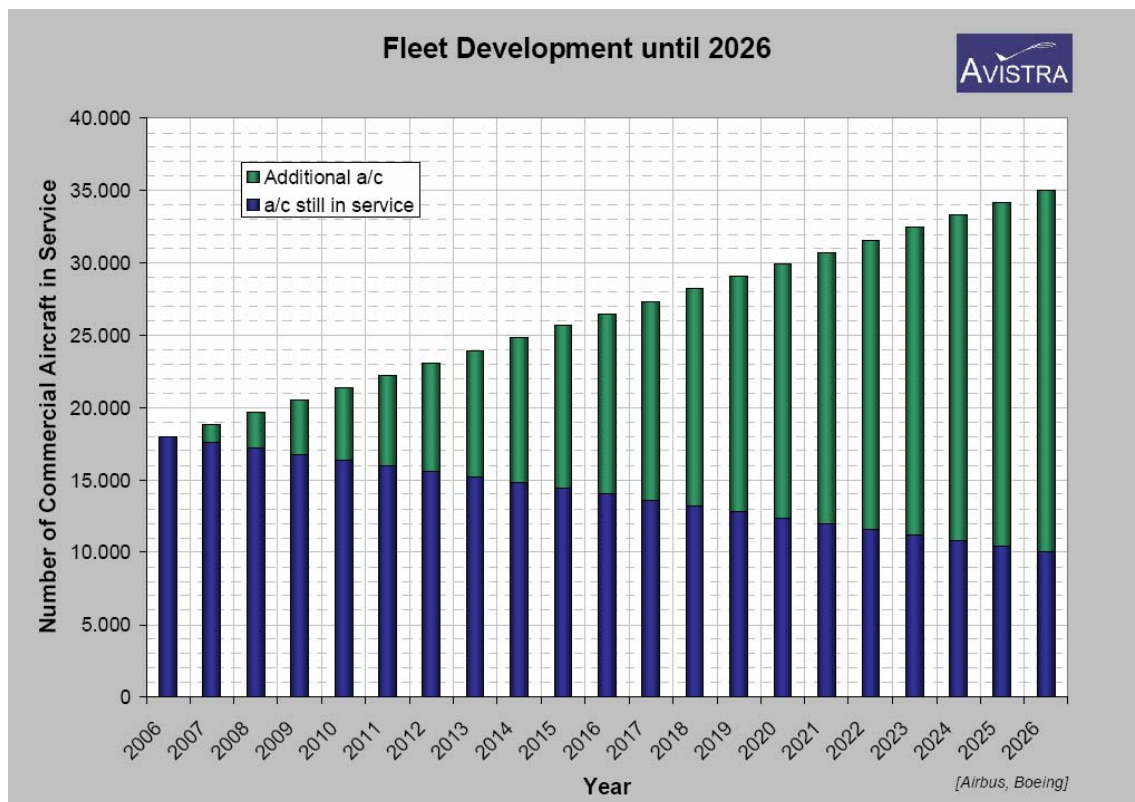
However, depending on the limit value, some engines might already comply with a possible new regulation. As no regulation exists so far due to the lack of sufficient knowledge on particle characteristics and adequate measurement procedures, the baseline scenario for an assessment of the impact of any future limit value is the status quo. This means, all future reduction potentials will be compared to state of the art engine technology in 2006. Taking this into account, the number of engines in service today will decrease over time, while at the same time new aircraft will be introduced.

Figure 6-2 is based on numbers provided by Airbus and Boeing. While Boeing expects a disproportionate increase in point-to-point services, Airbus' forecast is based on the assumption that the major part of the global traffic growth will be handled by a few big hub airports ("A380 effect"). In consequence, the forecasts produced by Boeing and Airbus differ in terms of number of aircraft. Therefore, the data depicted in Figure 6-2 is based on a mixed view of both forecasts.

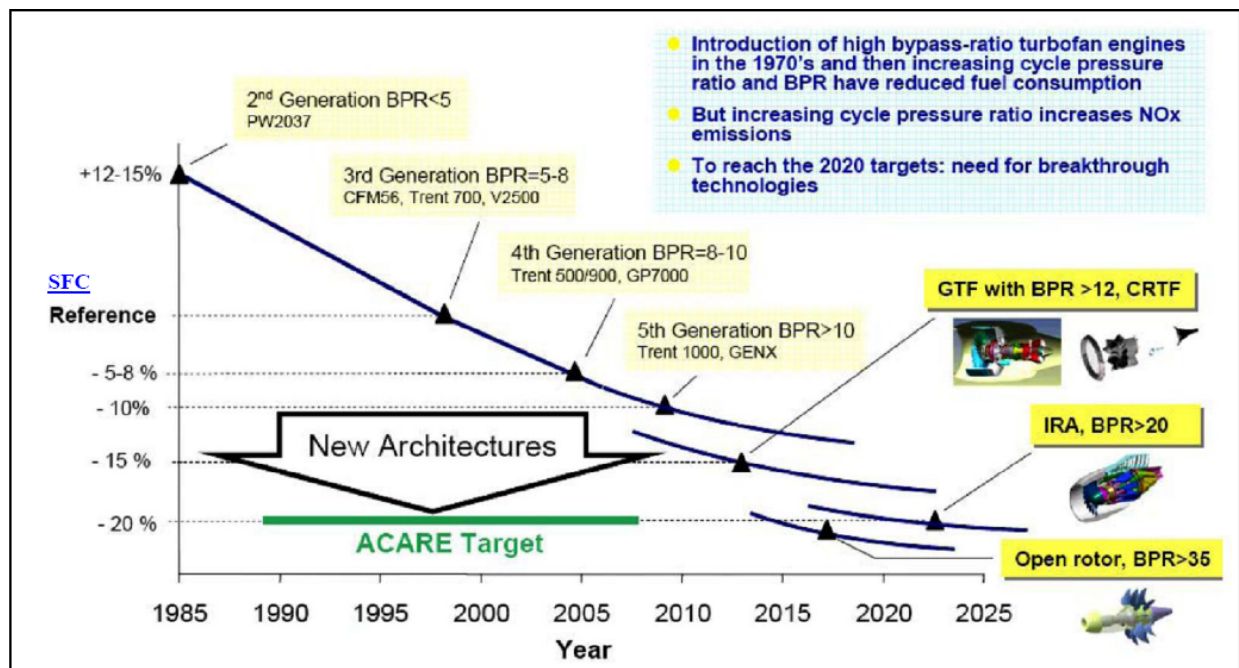
Over the next 20 years, 8,000 aircraft of the current worldwide commercial fleet will be replaced by new aircraft. In addition to this, the growth in air travel demand will be absorbed by another 17,000 aircraft, resulting in 25,000 new aircraft in 2026 compared to 2006. Only 10,000 aircraft of today's fleet will still be operating.



**Figure 6-1:** Engine values and life cycle (Engine Year Book, 2007)



**Figure 6-2:** Fleet development until 2026

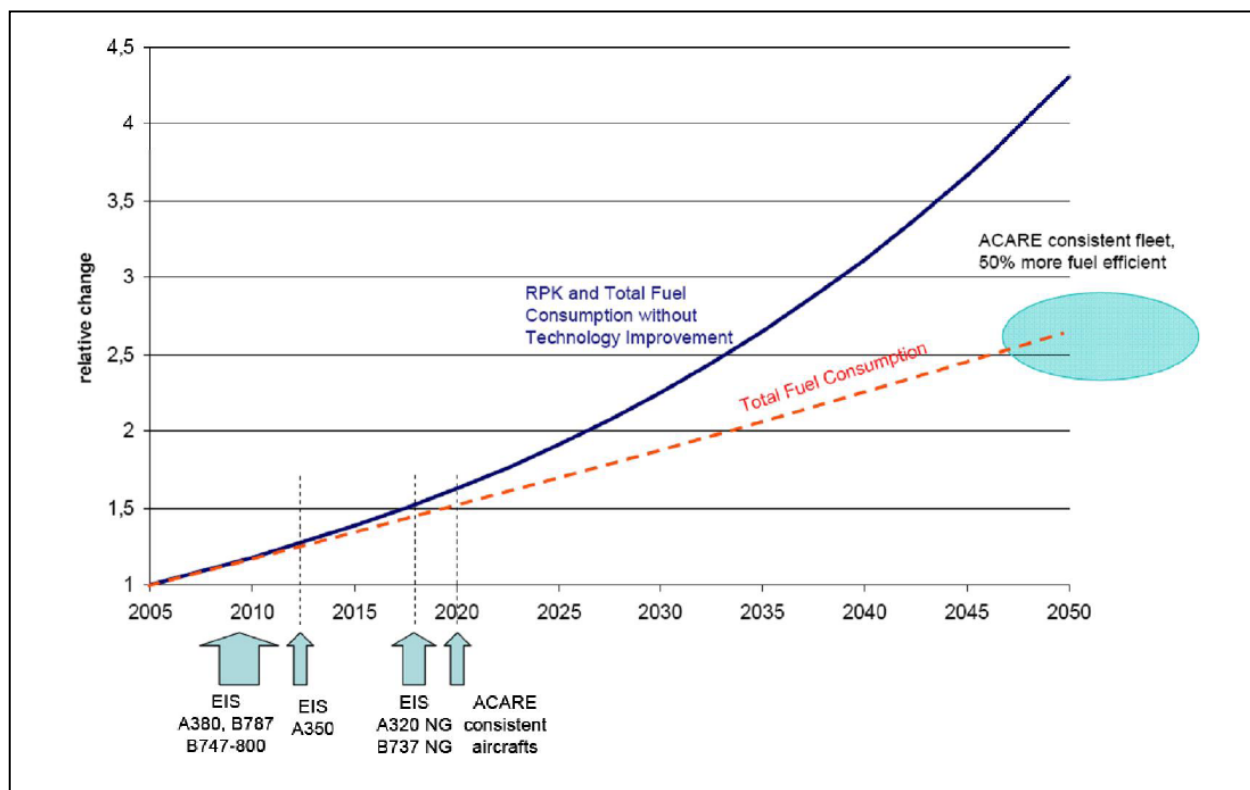


**Figure 6-3:** Reduction potential of SFC through new engine technology (Arndt, 2007)

With regard to engine technology, the main driving factor for environmentally friendly aircraft is still the specific fuel consumption, as this is directly linked to the operating costs of the airline. Figure 6-3 depicts the reduction potential in fuel consumption for the next 20 years. This is based on a prospected reduction related to engine technology of more than 20%. In addition to this, a further reduction potential of 25% from airframe improvements and 10 percent from more efficient ATM procedures is envisaged. This results in a total reduction potential of specific fuel consumption of up to 55% per PKT. However, due to overlapping effects and due to the fact that not all technological solutions are suitable for all kinds of engines (i.e. narrow-body, wide-body), the total effect for the entire world fleet will be lower.

Moreover, taking into account the expected growth in aircraft movements, this reduction potential is partly outweighed. The overall balance is schematically depicted in Figure 6-4. With the projected growth rate on the one hand and continuous advances in engine technology combined with new ATM procedures on the other, total fuel consumption will grow but at a lower pace than without the envisaged technology improvement.

Although the use of alternative fuels is discussed and currently being tested, these are not included in this calculation. However, Airbus predicts that about 25% of fuel used in aviation will come from alternative sources by 2025. Airbus just recently tested gas-to-liquid fuels on an A380 in the first flight of a commercial aircraft using the potential alternative to regular jet fuel. At the same time, Boeing tested a 20% biofuel mix of coconut and babassu oil in one of the four main fuel tanks of a B747. Boeing says that the industry had the potential to reduce emissions by 50% by 2050.



**Figure 6-4:** Change in total fuel consumption from 2006 to 2026 (Hüttig et al., 2008)

### 6.3 Potential Impact of a Limiting Value

The potential impact of any particulate matter emissions limiting value will have to be compared to the baseline scenario described above, where the emissions are exclusively related to fuel consumption. This means, that the goal of any new limiting value has to be a reduction of PM-emissions compared to this baseline scenario.

According to numerous forecasts, air traffic numbers will increase by 160% over the next 20 years in terms of revenue passenger kilometers (RPK). At the same time, through advances in engine technology and a constant fleet renewal, the total fuel consumption will only increase by 65% over the next 20 years. Thus, despite the increase in RPK by 160%, PM emissions will increase only by 65% only by the introduction of more fuel efficient aircraft. However, it will last at least until 2050 until the entire fleet is consistent with the new ACARE goals.

**Combining the effects of fuel efficiency improvement and increase in air traffic numbers, any limiting value for particulate matter emissions has to aim at a reduction of particle emissions growth to less than 65% over the next 20 years; otherwise this limiting value will be without impact on the environmental compatibility of aviation. The threshold value of 65% is set by the baseline scenario for the development of the aviation fleet, applying today's specific particle emissions.**

## Chapter 7 Report Conclusions

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The final chapter of this report summarises the conclusions drawn in the particular chapters in order to give a comprehensive presentation of the achieved results and recommendations:

### *Health Effects*

It is concluded from recent scientific studies that the surface area of ultrafine particles has to be considered as key particle property related to adverse health effects. Next, particle number is as well correlated to health effects, while the total mass of particles is linked to health effects only for extremely high particle loads. Therefore, current legislation for limiting PM<sub>10</sub> is not sufficient.

A limiting value for aircraft engine emissions could be defined independent from potential adverse health effects, as the rationale should be that aircraft engines emit as few particles (in mass and number) as possible.

WHO recommends that complete exposure/concentration-response relationships for different health endpoints provide better information for designing effective strategies to reduce adverse effects on human health.

### *Climate Effects*

In the very near future we cannot expect a robust and simple YES or NO answer to the question whether particles emitted from aircraft have a significant impact on cirrus clouds, heterogeneous chemistry, and thus on climate. However, research indicates that the climate effect of particle emissions is related closer to particle number than to particle mass.

### *Particle Metrics*

Potential precisely accessible particle properties are particle number, particle size and particle mass. Particle number and size, because the scientific debate on health effects of particulate matter focuses on insoluble nanometer-sized particles by number or surface. Particle mass, because current air quality regulations focused on particle mass.

### *Limiting Value Concept*

The Particle Measurement Programme of the European Commission (ECE, 2007) for ground-based emission sources (passenger cars and heavy duty diesel trucks) validation exercise has demonstrated that the particle number measurement method is a far more sensitive indicator of particle emissions performance than even the revised particulate mass measurement.

A limiting value will have to be related to the rated thrust, irrespective of the measured variable, i.e. mass and/or number, in order to match the concept of limiting values for gaseous emissions from aircraft engines.

Agreement is required whether or not any limiting value has to take fuel composition (sulphur content, bio fuels) into account since fuel properties may influence engine emission properties.

Combining the effects of fuel efficiency improvement and increase in air traffic numbers, any limiting value for particulate matter emissions has to aim at a reduction of particle emissions growth to less than 65% over the next 20 years; otherwise this limiting value will be without impact on the environmental compatibility of aviation. The threshold value of 65% is set by the

baseline scenario for the development of the aviation fleet, applying today's specific particle emissions.

Regardless of the nature of the limiting value, it can be designed in a way that already existing engines would already comply with it. In that case, the impact of such limiting value would be negligible. On the other hand, the design and the threshold of a new regulation dealing with particles have to take into account the feasibility in terms of engine technology. This means that the limiting value cannot be lower than a minimum standard already achievable today or at least within the next few years.

Therefore, a balance between the achievable and the desirable must be found. However, the prerequisite for finding such balance is the definition of a variable to be measured together with a standardised measuring procedure. Once these two parameters are set, an estimate on the technical feasibility together with a timeframe for the implementation of new technology can be given. Only then, the balance described above can be found and a limiting value be introduced.



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