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Review: Particulate matter emissions from aircraft

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Abstract: The contribution of aircraft operations to ambient ultrafine particle (UFP) concentration at and around airports can be significant. This review article considers the volatile and non-volatile elements of particulate matter emissions from aircraft engines, their characteristics and quantification and identifies gaps in knowledge. The current state of the art emission inventory methods and dispersion modelling approaches are reviewed and areas for improvement and research needs are identified. Quantification of engine non-volatile particulate matter (nvPM) is improving as measured certification data for the landing and take off cycle are becoming available. Further work is needed: to better estimate nvPM emissions during the full flight; to estimate non-regulated (smaller) engines; and to better estimate the emissions and evolution of volatile particles (vPM) in the aircraft exhaust plume. Dispersion modelling improvements are also needed to better address vPM. As the emissions inventory data for both vPM and nvPM from aircraft source improves, better estimates of the contribution of aircraft engine emissions to ambient particulate concentrations will be possible.

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1. Introduction

Particulate matter emissions from aircraft engines contribute to ambient concentrations of ultrafine particles in and around airports together with other combustion sources including road traffic. The impact of emissions on ambient concentrations from an airport, for which aircraft engine is a main source, differs from airport to airport due to the different relative contribution of other sources such as road traffic, and due to pollutant mix differences, chemical characteristics and size distribution [1]. Particulate matter, particularly the ultrafine component made up of small particles with an aerodynamic diameter of less than 0.1 μm, is widely considered a health hazard [2]. Aircraft gas turbine engines result in direct emissions of 'non-volatile' (nvPM), also described as black carbon (BC) 'soot' emissions. A number of research projects such as AAFEX [3], SAMPLE [4] and APRIDE [5] on the topic of aircraft particulate matter emissions have been undertaken in recent years in Europe and North America. These projects have contributed to the better understanding of these emissions and together with recognition of the health concerns regarding particulate emissions from aircraft engines e.g. [2] and [6], the International Civil Aviation Organization (ICAO) agreed in 2019 to the aircraft engine emission stand-

ards for nvPM (both mass and number), providing regulations for aircraft engine emissions (see Section 2.1). In addition to local air quality impacts, particles emitted from aircraft engines can affect climate and cloudiness in a number of ways [7]. There are several on-going projects such as AVIATOR [8] and ACACIA [9], that are taking measurements, linking these to modelling and assessing the particulate impacts on local air quality and climate.

At the high temperatures in the engine exit exhaust, particulate emissions mainly consist of ultrafine nvPM, the geometric mean diameter of these particles typically ranges from 15 nm to 60 nm. nvPM is formed in the combustor of the aircraft engine, and these emissions are considered 'primary' emissions. nvPM emissions are the main focus of this review. Volatile particles (vPM) condense and agglomerate in the exhaust plume or at a later stage in the ambient atmosphere and, due to their evolution in the aircraft exhaust plume, are more difficult to quantify, measure and assess. However, together with nvPM, vPM contributes to total measured ambient concentrations of particulate matter (the ambient measurements do not usually distinguish between them) which are compared with current local air quality health guidelines [10].

In this review, we consider the quantification of emissions and concentrations of nvPM from aircraft engines. In the following, we provide the regulatory background for aircraft engine emissions, examining the role of combustion technology and fuel composition in the formation of nvPM (Section 2); the nvPM emissions from non-regulated (smaller) aircraft engines are reviewed in Section 3; Section 4 of the review considers vPM emissions; Section 5 provides an overview of emission inventory methodologies; and Section 6 considers the dispersion modelling literature and methods. Finally, we summarize the knowledge gaps which could help evaluation of aircraft particulate emissions and the contribution made by aircraft engines and thus help guide future mitigation options.

2. nvPM Emissions from Regulated Aircraft Engines

2.1. Engine Emission Regulations

Regulated aircraft engines are those required by ICAO to be certified for their emissions performance, namely turbofan and turbojet engines of maximum rated thrust at sea level greater than 26.7 kN. These engines are typically used in commercial passenger and freight aircraft as well as in larger business jets. Standards limiting the LTO (landing and take-off) emissions of smoke number, nvPM (as maximum mass concentration), unburnt hydrocarbons (HC), carbon monoxide (CO) and oxides of nitrogen (NO_x) from turbojet and turbofan aircraft engines are contained in Annex 16 Volume II [11] to the Convention on International Civil Aviation, Doc 7300. Before 2016, the only emission standard related to PM emissions was the Smoke Number regulation, which effectively requires the engine emission to be invisible. However, in recognition of the growing health concerns regarding ultrafine PM and especially nvPM, the CAEP/10 nvPM mass concentration certification standard was agreed to at ICAO-CAEP (Committee on Aviation Environmental Protection) in 2016, which also required the LTO nvPM mass and nvPM number emission indices (EIs) to be reported.

Using standardized data collected under the CAEP/10 certification procedures, the CAEP was able to develop new emission standards for nvPM, and at the ICAO-CAEP 11th meeting (in February 2019), new emission standards for both nvPM mass and nvPM number and the corresponding additions and amendments (Chapter 4) to Annex 16 Volume II were agreed to. These standards include new regulatory limits for nvPM mass and nvPM number applying to both in-production and new engine types from 1 January 2023. The new engine emissions standards are indicative for LTO total nvPM

mass and nvPM number emissions per kN of rated thrust. As part of this agreement, the nvPM mass concentration standard is now considered to preserve the exhaust plume invisibility and there was also agreement to end the Smoke Number Standard applicability for engines of rated thrust greater than 26.7 kN beginning 1 January 2023.

Certification requirements include the need to report data under ISA (International Standard Atmosphere) conditions (except for the absolute humidity which is set to 0.00364 kg water/kg dry air), with a defined fuel specification and following specified test conditions requirements. Engine manufacturers report the emissions data alongside some engine characteristics to ICAO. These data are stored and made publicly available through the ICAO Aircraft Engine Emissions DataBank (EEDB). The databank, which covers all engine types whose emissions are regulated and the information provided by the engine manufacturers, is hosted by the European Union Aviation Safety Agency (EASA) on behalf of ICAO [12].

The certification process involves running the engine on a test bed at each thrust setting defined by the standard ICAO LTO cycle (100% for take-off, 85% for climb, 30% for approach and 7% for taxi/ground idle) [11]. The results of the engine emissions certification are the fuel flow (kg/s), the Emissions Index (EI) for NO_x, HC and CO (mass of emissions per kg fuel), the measured smoke number, the measured nvPM mass concentration and will now also include the EI for nvPM mass and nvPM number. These values allow for the calculation of emission data for each pollutant such as: emission rate (g/s); values of total LTO emission per rated thrust (total mass per kN and total nvPM number per kN); and the maximum Smoke Number.

2.2. Combustion Technologies

2.2.1 Pollutant formation in combustion chamber

As discussed, environmental concerns over aircraft engine emissions have resulted in the implementation of ICAO-CAEP emission regulations (see previous Section 2.1) which are part of the certification process for the engine. The combustion technology has had to evolve to control these regulated pollutants in addition to the principal imperatives of safety and operability. In the current engine designs that are now in service, the environmental focus of the combustion design has been controlling NO_x emissions whilst improving fuel efficiency. However, as the relevance of nvPM emissions has increased, the design of combustors has to consider both NO_x and nvPM emissions as well as fuel efficiency and, of course, all within the safety and operability constraints (altitude re-light, turbine inlet temperature, combustion efficiency, combustion instabilities, thermal load, etc) which bound the main design decisions.

Modern engines designed for subsonic aeroplanes generally tend to easily achieve the CO and unburnt HC emissions regulations; these pollutants are now of such low concentrations that they are no longer considered to be of much concern in urban or around airport locations. The focus of the following sections is to examine the most recent design features of combustors that affect the emissions of nvPM and NO_x, which are of most current environmental concern. The two main modern combustion technologies are covered: the most widespread, Rich-burn, Quick-quench, Lean-burn (RQL) and Lean Burn (LB) technologies.

2.2.2 Rich-burn, Quick-quench, Lean burn (RQL) technologies

In conventional RQL combustion chambers, fuel is atomized into a swirling and recirculating flow at the swirl fuel injector and the airflow is set to keep the primary

flame zone fuel rich. Flow exiting the primary rich zone is then quickly diluted, or “quenched”, to a uniform lean mixture. To minimize the formation of NO_x , the RQL design facilitates a fast passage through the high temperature zone where the fuel air ratio is stoichiometric and the NO_x formation potential is high [13]. Compared to previous generation combustion chambers, RQL technologies have significantly reduced NO_x emissions [14].

The formation of nvPM mainly occurs in the primary zone, close to the fuel spray, where fuel and air are not well mixed, and the intermediate zone reduces gas temperature by addition of a small amount of air to promote the complete oxidation of CO and soot particles [13]. However, there is an inherent trade-off in the RQL combustors in that the oxidation and removal of soot particles (formed in the rich primary zone) occurs in the hottest zone, where NO_x formation is highest. So, the control of nvPM at the same time as reducing NO_x provides some challenges to the RQL design, which was conceived originally for NO_x control. Furthermore, it is technologically difficult to cool the primary zone walls (no direct injection of air; double wall principle is often used in practice). Designers strive to minimize both nvPM and NO_x , most recently employing staging to separately optimize low and high-power conditions [14].

2.2.3 Lean Burn (LB) combustors

In a lean-burn combustor, where the fuel to air ratio is lower than the RQL type combustor, the peak temperatures are not as high. As a result, NO_x emissions are lower provided that the overall outlet temperature is not above about 1800 K [14]. At the same time, the excess air leads to a lower nvPM production [14]. However, a difficulty with hydrocarbon fuels is that they will not burn if the fuel air ratio is far below stoichiometric value and lean flames are inherently unstable. For the demanding operating conditions of aero combustors, a pilot zone is required for stability particularly during low power operation. Conditions in this pilot zone are like a small rich-burn combustor, producing nvPM, but because of the small size of the pilot zone and the small fuel-flow through it, the amount of pollutants is relatively small. Except during pilot-only operation, downstream lean-burn regions promote burn out of any particles formed by the pilot, so that levels of nvPM should be expected to be low [14]. Lean burn technology has utilized partial premixing such as the TAPS technology [15].

2.2.4 Future Technology and nvPM Control

Both LB and RQL combustors were designed to control NO_x emissions in modern combustors, where temperature and pressures are increasing, and both technologies have been successful in controlling thermal NO_x production. However, Lean Burn technology also has resulted in very low emissions of nvPM compared to most RQL combustors [14].

The most recent technology step in nvPM and NO_x emissions reduction has been the introduction of lean-burn combustors (GE’s TAPS in GenX and CFM LEAP engines). Although first entering service in late 2011, the technology remained the product of only one engine manufacturer on one large engine family until a second family was introduced in mid-2017, with combustion technology essentially by the same company. These two engine families are the only lean-burn engines in service at the time of writing [12].

Further developments in premixing technology to reduce nvPM include: i) the Lean-burn Direct Injection (LDI) [16] and ii) the less mature multi-point injection technology being developed by NASA [17]. The latter is an evolution of the existing lean-burn concept but demonstrates an increased staging flexibility. NASA research into

multi-point injection technology is focused on significantly increasing the number of injectors to allow better premixing and vaporization of the fuel and air [16]. There are, however, challenges associated with this technology and the ICAO-CAEP Independent Expert Integrated Review panel (IEIR) considered that it was unlikely that the technology would be ready for entry into service in the next 15 years [14].

2.3. Fuel Composition and PM Emissions

Sustainable Aviation Fuel (SAF) in this context includes sustainable fuels made from biogenic wastes and residues (biofuels), fuels produced through Power-to-Liquid technologies (synfuels), as well as fossil fuels engineered for improved environmental performance. Sustainable biofuels and synfuels are subject to sustainability criteria such as those used in the European Union Renewable Energy Directive and in ICAO's Carbon Offsetting and Reduction scheme for International Aviation (CORSA). Both types of sustainable fuel are lower in aromatic hydrocarbon compounds, including naphthalene and their sulphur content may reach – in pure form – near-zero values.

There have been several measurement campaigns that have quantified reductions in emission of particulate matter behind aircraft engines at the ground, especially comparing biogenic SAF with conventional kerosene [5,18–21].

An overview of fuel composition effects on nvPM emissions can be summarized as follows: the combustion of 'pure' or blends of SAF results in lower nvPM emissions when compared to the same Jet A-1 baseline due to the lower aromatic content in the fuel. The change in the nvPM emissions characteristics varies with engine type and operation [5,18–27]; the observed reduction in soot number and mass emissions is greatest at low engine thrust conditions and decreases with increasing thrust. In addition, SAF combustion results in: a reduction in the mean particle size distributions [5,18–27]; a shift in chemical composition with a sizeable decrease in the emission of organics and sulphates [3,22–32]; an alteration in the nvPM morphology with an increased prevalence of amorphous outer shell structures [33–35]; an increase of hygroscopic growth factor and hygroscopicity parameter with fuel sulphur content and engine thrust and a decrease with dry particle diameter [34]; and a shift in optical properties with reduced absorption, scattering and extinction coefficients [35]. In terms of vPM emissions, SO₂ emissions – a precursor of vPM in the exhaust plume – are directly proportional to the sulphur content of the fuel.

3. nvPM Emissions from Non-Regulated Engines

3.1. Introduction

ICAO requires that turbofan engines of maximum rated thrust at sea level greater than 26.7 kN be certified for their emission performance (Section 2.1). There are however other classes of engines that do not fall under this regulated category. These non-regulated engines include the following: i) small turbofan engines with rated thrust below 26.7 kN and often used on business jets and small private jets; ii) military turbofan engines; iii) auxiliary power units (APUs); iv) turboprop engines; v) turboshaft engines mainly used on helicopters; and vi) piston (or reciprocating) engines. For these unregulated engines, there are limited publicly available data, exposing a knowledge gap in aviation environmental impact assessment and mitigation. It should be noted that on a global scale, nvPM emissions from non-regulated engines are significantly lower compared to the regulated ones, and on a local scale this is likely to be true around large airports too. However, non-regulated engines may be significant emission sources at airports that mainly service aircraft listed i) to vi) above and therefore, could be a concern

for the local population around these airports. Here, we present a summary of available data with respect to the emission profiles and emission performance of unregulated engines. Unlike the regulated engines, available data in this case are often not reported according to the ICAO-prescribed LTO cycle. Emissions are primarily reported in pollutant units (grams or number of particles) per kilogram of fuel used, i.e. the EIs, and/or concentrations in grams or number per volume of gas sampled. A collection of the available data has been prepared as an Excel file with databanks for unregulated engines, and a compilation of additional data tables will be published over the course of the year 2022 as part of the RAPTOR project (<https://aviation-pm.eu>). For non-regulated engines (e.g., piston or turboshaft engines), emission factors were collected from a variety of sources.

3.2. Emission profiles of non-regulated engines

A summary of the literature on emission profiles from non-regulated engines can be found in Table 1. One main theme is the lack of publicly available data. Where data were available, there is no clear and consistent standardized measurement method or power setting across the different studies. Engine properties for which emissions were measured in some cases are unknown, for instance, APUs. A standardized test program including information allowing for example loss corrections would improve the quality and usability of available data for air quality modelling and development of inventories. In addition to missing engine characteristics, particulate emission data are limited for most engine classes. For military turbofan, turboprop and turboshaft engines, these data are primarily from the 1970s and 1980s; methods applied are relatively outdated. For instance, Spicer's group concluded in the 1980s that tested engines were clean based on the formerly used methods allowing smoke and particulate measurement. Those filtration methods miss the ultrafine particles that are now believed to have severe health impacts. Also, there are scarcely any data, gaseous and particulate, available for the civilian counterpart to aircrafts engines in military application.

Analysis of nvPM data for two small turbofan engines [36] showed that overall, the nvPM characteristics (e.g., increase of Geometric Mean Diameter (GMD) with thrust, GMD as a function of nvPM number / mass ratio) are in the range of those found for large engines. Both engines would nominally pass the CAEP/10 and CAEP/11 in-production standards (if the limit line were theoretically and linearly extended down to sub 26.7 kN).

Few of the studies presented in Table 1 measured PM mass and number concentrations. In some cases, only the concentration and/or the smoke number (SN) were presented. Due to differences in the maximum rated thrust of the different engine types, the different methods for reporting engine power, inconsistencies across studies with respect to what power settings were measured, a quantitative comparison of the different engines cannot be discussed. However, one can discuss the trends in PM emissions observed as follows:

- Turbofan-type engines: Durdina et al. [36] observed maximum nvPM mass emission indices (EI) at approach and minimum at idle; a decrease in PM number EI was observed, with slightly higher PM number at idle than at approach. Like the small turbofan engines, APUs showed decrease in PM number and PM mass with increasing power from Honeywell GTC P85 engines [22,37]. Crayford and Johnson [38] observed lower SN at low power than at higher power from a Rolls Royce Artouste MK113 engine. For military turbofan engines, Spicer et al. [39] observed increase in SN with increasing power, like the Artouste MK113.

- Turboshaft: Measured turboshaft SN increased with increasing power, like the turbofan engines [40]. There were however differences in the particle mass and number EI. For small turboshaft engines, Drozd et al. [41], who measured at idle and cruise, observed lower PM mass EI at cruise than at idle from T63-A-700 engines. On the larger T700-GE engines, Corporan et al. [40] observed an increase in PM mass and number EI with increasing power. 284
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- Turboprop: In the case of turboprop engines (all groups measured emissions on T56-A-15; [42–44]), SN increased significantly from the lowest power level, low speed ground idle (LSGI), to the next power level, high speed ground idle (HSGI). At higher power settings than HSGI, SN did not increase much. PM number EI decreased with increasing power after HSGI, remaining relatively the same from LSGI through HSGI to flight idle (FI). PM mass EIs also decreased slightly with increasing power. 290
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There are no public turboprop emission data like the ones provided for jet engines in the EEDB. In addition, the non-public FOI data [45], which form a sort of standard for turboprop emissions, do not contain nvPM values or Smoke Numbers. Nevertheless, it would be a useful step forward to develop a public fuel flow (FF) and EI data set for turboprops including nvPM that would serve as a common reference. A unique and internationally agreed labelling (engine UID, like for the EEDB) for non-regulated engines would be helpful to share and exchange engine emission data and to assign engines to specific aircraft types. 297
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Table 1. Summary of Data on Emission Profiles of non-Regulated Engines

Studies	Type of Engine	Description of data	Measured / reported compounds
Durdina et al., 2019 [36]	Turbofan <26.7kN	Measured nvPM emissions from a Dassault 900EX carrying three Honeywell TFE731-60 engines were similar in profile to larger engine measurements.	nvPM mass and number, GMD
Klapmeyer and Marr (2012) [46]	Turbofan <26.7kN	Plume measurements, during regular airport operations, of NO _x , CO ₂ , and PM from Cessna C560 aircrafts carrying two Pratt & Whitney (PW) JTD15-5 engines during idle/taxi and at take-off.	NO _x , Particle number, CO ₂
ICAO EEDB [12]	Turbofan <26.7kN	Pratt & Whitney reported emissions from JT15D series (-1, -4, -5, -5A, -5B, -5C) and corrected as prescribed by ICAO. Allied Signal reported emissions from TFE731-2-2B and TF3731-3 engines	Reported on ICAO EDB; HC, CO, NO _x , SN
Spicer et al., 2009, 1992, 1989, 1987 [39,43,47,48]	Military turbofans	Military turbofan engines have different power modes than non-military turbofan engines including an afterburn power mode. However, excluding afterburn power mode for which emissions data are very scarce, military turbofan engine emission profiles are like other turbofan engines. Particle emissions measured as smoke numbers showed highest smoke numbers at 75% to intermediate power and lowest at idle to 30% of normal rated power. Measured airplane engines include F110, F101, F100-PE-100, TF41-42, TF30-P103, TF30-P109.	(JP-4 fuel; [43]: JP-8 + 100) HC, CO, NO _x , SN

<p>Bulzan et al., 2010; Crayford and Johnson, 2011; Khandelwal et al., 2019; Kinsey et al., 2012 Lobo et al., 2015</p> <p>[3,22,37,38,49]</p>	<p>APUs</p>	<p>Generally, APUs show similar CO and HC emission profiles to larger turbofan engines. Observed NO_x emissions were different; while some studies observed no change in NO_x emissions others observed some increase in NO_x emission with increasing power [37,38]. Particle mass EIs decreased with increasing power demand for GTCP85 series [3,22,37], whereas a Rolls Royce Artouste Mk113 APU had higher PM mass concentration (mg/m³) at full power than at idle [38]. Lobo et al. (2015) observed lowest PM number EIs at highest power. Kinsey et al.'s (2012) study was inconclusive in PM number EIs as different research groups in the same campaign showed different particle number EI profiles; some were u-shaped with maximum at highest power, others showed no variation with power. For the Kinsey et al.'s group (2012), using Fischer Tropsch fuel (FT; synfuel) reduced PM number and mass EI, and had a clear profile of decreasing EIs with increasing exhaust gas temperature. Crayford et al. observed higher smoke number (SN) and PM number concentrations (number/cm³) at full power than at idle [38].</p>	<p>[37](JP-8, and FT-2): HC, NO_x, CO, nvPM mass and number</p> <p>[38]: HC, CO, NO_x, SN</p> <p>[3] (JP-8, and FT-2): SO₂, HC, CO, NO_x, nvPM mass and number</p> <p>[22] (Jet A1): nvPM mass and number</p> <p>[49] (Jet A1): CO and NO_x</p>
<p>Cain et al., 2013; Corporan et al., 2007, 2010, 2004; Drozd et al. 2012 ; Kinsey et al., 2019</p>	<p>Turboshaft engines (primarily used on helicopters)</p>	<p>Variable observations were made for particulate emissions probably due to differences in sampling methods. There was a general agreement in particle number emissions. Particulate number and mass emissions</p>	<p>[50] (JP-8): CO₂, CO, PM mass and number, particle size distribution (PSD)</p> <p>[21,40,42,51,53](JP-8, FT): GMD, SN, CO, NO_x, PM mass and number</p>

[40–42,50–52]	(concentrations and EIs) and geometric mean diameter (GMD) increased with increasing power. General emission profiles of emissions of CO, NO _x , and HC are like those of turbofan engines. PM emissions were significantly reduced with FT fuel.	[41](JP-8, FT): PM mass, CO, CO ₂ , HC [52] (JP-8, FT): GMD, CO, CO ₂ , HC
Chan et al., 2013; Cheng et al., 2008; Corporan et al., 2008; Spicer et al., 2009 [43,44,53,54]	Turboprop engines (primarily on military aircraft)	Emission measurements were primarily conducted on turboprop engines for military purposes as in the T56 series III engines on C-130 Hercules (C-130H) aircraft. Power in turboprop engines is reported as shaft horsepower (shp). The gaseous emission profiles observed for the T56 series engines are like those of turboshaft engines. Particle number and mass emissions tended to decrease with an increase in power. [54] (JP-8): CO, NO _x , CO ₂ , SO _x [53] (JP-8): SN, PM number and mass, GMD, CO, NO _x , CO ₂ [43] (JP8): CO, NO _x , OC [44] (F-34, 50-50 F34/Camelina-HEFA blend) : PM number and mass, NO _x , CO, HC

4. Volatile Particulate Matter (vPM)

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As the exhaust leaves the engine, the hot combustion exhaust gases cool down and liquid droplets and condensation nuclei of sulphates and nitrates are formed, on the surfaces of which further substances such as water and organics can condense. Shortly after formation, these particles have typical diameters of a few nanometers. Both the number and mass of particles change significantly during transport due to processes such as agglomeration, condensation, and evaporation on a time scale from seconds to several tens of minutes. An effective emission rate can be derived from the number of particles in the cooled exhaust gas. These particles are referred to as volatile or semi-volatile ultrafine particles but here, the simplified term volatile particulate matter (vPM) is used.

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Timko et al. [28] report on measurements during the AAFEX campaign. Particles (sum of vPM and nvPM) were measured at distances between 30 m and 300 m behind a CFM56 engine for different power settings and types of fuel. At distances with moderate dilution, as compared to the smaller distance (30 m) from the engine exit, more particles were measured as condensation nuclei were formed in the plume. The study also observed that nucleation of particles accelerates with increasing fuel sulphur content, as these new particles are generated largely from sulphates. About one order of magnitude more particles were observed for low power while the difference was less pronounced for higher powers. The authors deduce that the PM evolution strongly depends on the ratio of particle precursors (sulphate and organics) to soot; more nvPM particles are generated at higher powers and lead to a greater interaction between precursors and nvPM with more coating of nvPM with sulphuric acid etc and thereby to a smaller total number of newly formed particles as compared to lower powers.

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Wong et al. [55,56] report on a detailed microphysical modelling of the formation of organic and sulphuric coatings on nvPM particles and then investigated, using microphysical modelling, the role of organic emissions in the formation of volatile particles in the near field (distances less than 1000 m) behind a CFM56 engine at 7% maximum thrust. Soot coating and accordingly particle growth was fastest after some cooling of the exhaust gas and before dilution becomes dominant (distances between 100 m and 500 m). Soot coating was mainly by organics with low vapour pressure. Higher ambient temperatures resulted in smaller soot coating due to the higher saturation vapor pressure. The nucleation of new particles in aircraft plumes at ground conditions at a distance of 1000 m was solely determined by the binary nucleation of sulphuric acid and water. The presence of organic vapours did not promote nucleation of new particles but, via condensation, affected the mass composition and the size of the nucleated aerosols. At ambient temperatures above 285 K, most of the sulphuric acid mass condensed as soot coating. For organic species, condensation was suppressed at higher temperatures, with almost no condensed organics at temperatures above 295 K. According to the model results, nucleation mode particles have a higher organics mass fraction than soot coatings for high ambient temperatures or low ambient humidities and a higher sulphate mass fraction otherwise.

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Peck et al. [57] modelled the evolution of surrogate organic emissions in an aircraft exhaust plume for low and high-power settings of the engine. They found that the nucleation mode particles have a sulphate core, but their mass is mostly organic coating. At low engine power, there was only slow condensation of organics on a few soot particles. At high power, there were more soot particles and hence more organic material condensed.

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Yu et al. [58] studied the mode-specific, semi-volatile chemical composition of particulate matter based on the AAFEX data at a distance of 30 m behind a CFM56 engine at different power settings. At low power, the mass of vPM was mainly organic material, while at high power it was roughly half organic and half other material.

In summary, the measurements and modelling show that non-volatile particles emitted from aircraft engines at ground are mainly soot which becomes coated over the first hundreds of metres mainly with sulphuric acid and water; the diameter and composition of these particles change during the course of transport but their number remains more or less constant. Volatile particles are mainly created by the binary nucleation of sulfuric acid and water; their mass and size increase due to coating with sulfuric acid, water, and organics. Ambient temperature and humidity affect the mass composition of volatile and (to less extent) non-volatile particles.

5. Emission Inventories

An emission inventory typically comprises a dataset with emission amounts, in terms of mass, for the most relevant air pollutant and greenhouse gas species, split up by source type, time and location. Different levels of spatial and temporal resolution are possible, depending on the scope and purpose of the inventory. Aircraft emissions inventories usually report only those emissions related to the LTO cycle for the aircraft, which covers activities up to 3000 feet (914 meters), which coincides with the typical order of magnitude for the height of the neutrally stratified atmospheric mixing layer. Emissions below this altitude are expected to be the dominant influence for local ground-level air quality parameters. The 3000 feet boundary is also used as the cut-off altitude for reporting national emissions for air pollution under EMEP (Gothenburg Protocol) and the EU National Emission Ceilings Directive (2016/2284/EU), so national emission inventories often pay limited attention to what happens at higher altitudes.

5.1 Airport Emission Inventories

At airport level, the compilation of an emission inventory is typically motivated by the permitting process, which requires an Environmental Impact Report investigating continued or planned activities at the airport in question. Other potential reasons for compiling an emission inventory are to perform benchmarking or to monitor emission trends in the light of emission mitigation plans or actions. Since individual airports are not subject to international regulations covering the regular reporting of emissions by country, these emission inventories do not have a fixed format, scope or methodology. The scope of the inventory will likely be influenced by the local issues that are thought to be most important. When considering only a single airport, it is usually possible to include more detailed information in the analysis and perform a more comprehensive modelling of the emissions. The emission inventory will typically include all significant emission sources at the airport and cover a full calendar year and a range of pollutant species, such as carbon dioxide (CO₂), carbon monoxide (CO), nitrogen oxides (NO_x), sulphur oxides (SO_x), particulate matter, and hydrocarbons (HC) or non-methane volatile organic compounds (NMVOCs).

Celikel et al. [59] investigated the influence of different emission models on the inventory of an airport, and Fleuti and Maraini [60] carried out a sensitivity study of how assumptions and applied levels of detail on aircraft traffic affect the overall emission. In summary, aircraft are the dominant source of emissions including particulate matter at an airport. The calculation of aircraft emissions are found to be standardized to a large extent by the ICAO engine emission databank, in contrast to vehicle emissions (airside

traffic, ground support equipment), which show a larger variation across different data-banks. Model-specific assumptions on climb profiles and their assignment to aircraft types or more generalized aircraft groups can strongly influence the resulting emissions. More complex emission calculations for aircraft (performance models) show a tendency to decrease the emissions of NO_x, PM10 and CO₂ and to increase the emissions of HC and CO (more realistic take-off thrusts below 100 % maximum). The authors concluded that total aircraft emission up to 914 m is not a suitable quantity for describing the role of aircraft in the context of regional air quality because the near-ground impact decreases with emission height and most other sources emit only at ground level.

5.2 Regional level (Country, District)

EU Member States are subject to emission legislation [61] requiring the annual reporting of air pollutant emissions by sector, including the aviation sector. Currently, these national inventories require only the reporting of emission in terms of mass, not in terms of particle numbers and/or particle size distributions. Although the country-level emission inventory will distinguish between the different airports in the countries, it is constructed at a national level and not by combining separate airport level inventories. This is motivated by the requirement that a single consistent and transparent methodology is used for all years and sources. The full requirements on data and methodologies used are specified in the CLRTAP guidelines [62] and the EMEP/EEA Emission Inventory Guidebook [63]. For the highest level of detail (Tier 3) recommended by the Guidebook, information on the aircraft type and destination of each flight is used in combination with more detailed aircraft/engine- and LTO phase-specific emission factors. The Guidebook notes the availability of emission factors for CO, NO_x, HC from the ICAO engine emission databank (EEDB) [12] and lists the First Order Approximation v.3 (FOA3) method for deriving PM emission factors from the EEDB reported smoke number. Note that from version 28 onwards, the EEDB also contains nvPM emission indices for certified in production engines [12]. Calculating emissions of climb, cruise and descent activities is not mandatory and the methods described in the EMEP/EEA Guidebook are less detailed for these flight phases. The Guidebook provides an emission calculator spreadsheet which estimates fuel burn for the non-LTO phase (Cruise/Climb/Descent) which is combined with the relevant emission indices for NO_x. A similar method is used in the emission calculator for nvPM mass emissions [63].

The U.S. Environmental Protection Agency (EPA)'s National Emissions Inventory (NEI) compiles estimates of air emission (criteria air pollutants (CAPs), precursors of CAPs and hazardous air pollutants (HAPs)) from a variety of sources including aircraft. Aircraft emissions (including PM10 and PM2.5) at airports (LTO portions of aircraft operations, ground support equipment) are reported as point source and non-point source [64]. These were estimated from ~20,000 airports and include the use of US Federal Aviation Administration (FAA)'s Aviation Environmental Design Tool (AEDT) [65].

5.3 Global level inventories

The ICAO-CAEP produces full flight global aviation inventory for greenhouse gases (CO₂, NO_x) and local air quality emission (NO_x) every three years and the international aviation totals are provided in the ICAO Environmental Report (Environmental Trends) [66]. The inventory is based on emission results from the US FAA AEDT model. These results usually compare well with the other two models (the European IMPACT and the UK FAST models) used in ICAO-CAEP analyses [67]. The 2022 report will include the 2018 annual estimate for full flight and LTO nvPM mass, in addition to post-Covid forecast years of 2028, 2038 and 2050.

Other published results provide the following estimates: the global LTO black carbon (BC) emissions are ~ 0.00083 Tg/year and ~ 0.00074 Tg/year for 2005 and 2015 respectively [68], while the full-flight estimate for 2018 was 0.0093 Tg/year [7]. These could be compared to other other anthropogenic sources 4.8 Tg/year for 2000 [69].

5.4 Methods to derive PM emissions

5.4.1 LTO nvPM mass and number emission estimates

As in-production engines (>26.7 kN) are now beginning to be certified against the CAEP/11 nvPM mass and nvPM number regulations, emissions are being reported to the ICAO EEDB [12]. The relevant emission indices from these certification measurements can be used directly for building LTO emission estimates for this part of the larger engine fleet. As the certification process moves forward and more emissions data are reported, the measured LTO emission indices will be increasingly applied. However, certification data are not yet available for all in production engines and some older (out of production) engines will not be subject to nvPM certification (though these older engines will continue to remain in the fleet for some years to come). Therefore, there continues to be a need for estimates of nvPM mass and number emissions to be calculated from smoke number measurements. In the most recent ICAO Doc 9889 Airport Air Quality Manual (v.2) [70], a new methodology, the First Order Approximation method v.4 (FOA4), is presented to estimate non-volatile PM emissions from the reported engine smoke number, as well as estimate the non-volatile particle numbers and particle size distribution. The methodology is based on the work done in CAEP/11 and previously published by Agarwal et al. [68]. Other studies e.g. [71,72] use a mass-to-number conversion based on the Fractal Aggregates (FA) approaches [4,73,74]. It is also possible that these estimation methods from smoke number measurements could be applied to smaller non-regulated (<26.7 kN thrust) turbofan engines where smoke number measurements may be available (but not nvPM mass and nvPM number measurements).

5.4.2 Full flight nvPM mass and number emissions

As discussed, it is common practice to calculate aircraft emissions up to 3000 ft and to only consider emissions during the LTO cycle in an airport emissions inventory. However, SO_2 , NO_x and nvPM emissions contribute to climate impacts and some studies have shown that emissions of NO_x and nvPM during cruise may also have an impact on ground level air quality [75]. In view of these factors, there is also a need to consider the emissions of NO_x and nvPM at higher altitudes. In global inventories to date, measurements of LTO NO_x emissions have been used to estimate NO_x emissions during non-LTO phases using methods such as the Boeing Fuel Flow Method 2 (BFFM2) [76] and the DLR method [77]. Similar methods adapting LTO emissions to represent emissions at higher altitude conditions are also under consideration for nvPM emissions. For example, in ICAO-CAEP, a new methodology is being developed for calculating full flight emissions. The method consists of 4 steps: the first one being the definition of the relevant cruise combustor conditions; the second the determination of suitable ground reference data; the third the interpolation of the LTO cycle data; and the fourth the correction of the interpolated data from ground reference to cruise conditions. While the proposed method is considered fit for purpose, some additional uncertainties prevent the method from achieving similar accuracy as the cruise NO_x prediction methods, e.g. the BFFM2. These uncertainties result from the wider variety of curve shapes of nvPM emissions characteristics compared to NO_x emissions, and the lower amount of measured cruise

emission data available for validation. It is recommended to further analyse, validate and, if appropriate, revise the method as further analysis takes place.

5.4.3 vPM emissions

Aviation vPM is largely considered to be made up of particles formed from sulphur (S) present in the fuel, see Section 4. Condensable organic material in the exhaust then condenses on these sulphate nuclei (and also on nvPM), coating and adding to their mass.

Volatile PM is formed from the fuel sulphur via oxidation of SO₂ (S-IV) to SO₃ (S-VI) and subsequent hydration, in the exhaust plume, of SO₃ to H₂SO₄. In ICAO Doc 9889 [70], the EI mass is estimated from the fuel sulphur content and the conversion rate of S-IV to S-VI (ϵ). A typical conversion rate would be around 2%, with a range of $\epsilon = 0.5$ –6%. A constant ϵ for an engine is assumed which means that the EI does not vary by power setting. Emissions of SO₂ can be calculated based on the fuel sulphur concentration, e.g., current evidence suggests that the globally averaged aviation fuel sulphur concentration is in the range 400–800 ppm [78,79]. Therefore, assuming a fleet average fuel sulphur concentration of 600 ppm, the corresponding EI of SO_x as SO₂ yields a value of 1.2 g/kg-fuel.

Measurements of condensable organics in the engine exhaust are very limited. Based on the assumption that condensable organics are directly related to unburned hydrocarbons, ICAO Doc 9889 recommends that an estimate is made by scaling the engine's reported ICAO hydrocarbon (HC) EI to those of other engines in the database [70]. Making a second assumption that modern engines behave in a similar manner, the HC ratio can be multiplied by the volatile organic PM mass EI for the CFM56-2- C1 engine which was measured during NASA's Aircraft Particle Emissions Experiment 1 (APEX1). The result is an EI for mass that is both engine and power-setting specific for the volatile organic PM.

The behaviour and characteristics of vPM are clearly more complex than nvPM and there are significant approximations currently being made in the calculation of effective vPM mass and number EIs. Further field work and modelling studies, such as those being undertaken under the European Horizon 2020 Project AVIATOR (<https://aviatorproject.eu>), will help to better characterize the estimation and role of vPM in aircraft exhaust plumes, their contribution to ambient concentrations of PM and ultimately health impacts.

6. Dispersion modelling

Dispersion models provide a 3-dimensional concentration distribution of pollutants for subsequent time intervals. They are an important tool to supplement measurements that take place at a few, specific locations. Dispersion modelling is also used to investigate future scenarios or to study specific processes of atmospheric dispersion. The range of dispersion models extends from simple analytic solutions based on physical conservation laws to complex numerical algorithms. ICAO provides with the Airport Air Quality Manual Document 9889 [70] an extensive description of emission and dispersion calculation methodologies at airports, in particular for aircraft main engines and APU. The 2nd edition includes a calculation methodology for nvPM mass and number emission indices (FOA4) based on work in the CAEP/11 cycle (Section 5). ICAO Doc 9889 has been continuously improved in the past cycles including CAEP/12 and the current CAEP/13 cycle, with an update expected in 2022.

ICAO-CAEP evaluated several dispersion models, which are designed for local air quality (LAQ) studies at and around airports [80]. The aim was to investigate which tools are sufficiently robust, transparent, and appropriate for CAEP analyses and to understand potential differences in modelling results. The use of multiple approved models provided CAEP insight into model-dependent sensitivities of the results. Some of these models are routinely applied in scientific projects and airport assessment procedures. In the Project for the Sustainable Development of Heathrow (PSDH), several dispersion models were evaluated and compared against measurements [81]. A further summary of aspects relevant for airport dispersion modelling is provided with focus on Los Angeles Airport [82]. Finally, an extended guidance for a model-based quantification of the contribution of airport emissions to local air quality can be found in the ACRP Report 71 [83].

A variety of modelling studies have been carried out at Zurich Airport, among others comparisons of model-based emission inventories [59], comparisons of modelled and measured concentrations [84] and sensitivity studies [60]. Barrett et al. [85] investigated the impact of aircraft plume dynamics on the predicted local air quality and observed that measured, aircraft-induced concentrations in the vicinity of airports do not reveal the usual inverse dependence on wind speed but are related to the effects of plume dynamics. A simplified methodology to account for plume dynamics in the context of emission grids for airport dispersion modelling has been developed in a study by EUROCONTROL [86].

In summary, dispersion calculations for airports are complex because many different source groups need to be accounted for. For some groups, standardized emission databanks (ICAO engine emission databank, national vehicle emissions) or recommendations (APU emissions according to ICAO Doc 9889) are available, but for other groups (ground support equipment, stationary sources) airport-specific data must be gathered. The resulting concentrations do not only depend on the applied emissions, but also on the applied type of dispersion model, on the applied meteorological data, and, in the near field of aircraft operations, on the assumed dispersion dynamics of the aircraft engine exhaust. Comparisons with measurements are hampered by the fact that non-airport emission sources and background concentrations contribute to the measured concentrations. Although there exist several data sets that have been used for comparisons, up today there exists no generally agreed gold-standard for the validation of airport dispersion models.

7. Discussion

Current emission source regulations cover the nvPM emissions from aircraft engines (>26.7kN thrust) for both mass and number. Combustion technologies for current in production engines were generally designed mainly to consider NO_x emission regulations and fuel burn (in addition to a whole host of safety and operability considerations). nvPM mass and number regulations are now also being considered in advanced combustion technology developments but there are potential trade-offs between technologies and their emission performance. Fuel composition is also an important factor in determining nvPM emissions, and the composition of SAF with lower aromatic content and virtually zero sulphur content also tends to lower nvPM mass and number emissions. The increased use of SAF to reduce the overall life-cycle CO₂ emissions of aviation is thus likely to lead to lower nvPM emissions from aircraft engines.

Available data for several smaller non-regulated engines was collated and it was found that it is often not consistent with the ICAO EEDB structure and therefore difficult

to use for emission calculations for an aircraft fleet mix. Turboprop engine data was lacking especially in terms of nvPM emissions data.

As the ICAO EEDB becomes populated with nvPM mass and number certification data for larger in production engines, these measured LTO EIs can be used to directly estimate the engine nvPM mass and nvPM number emissions in inventory calculations. These data will eventually cover most of the larger engines in the fleet and will provide a consistent data source for LTO emissions, based on real measurements. Some older engine types that remain in operation will still require the use of alternative estimation methods above based on smoke number measurements. Smaller non-regulated engines will require assessment using methods outlined in the ICAO Doc 9889.

To calculate full flight nvPM emissions, further work is required to establish whether a similar type of approach currently used for estimating full flight NO_x emissions from LTO emissions is applicable. Consideration of vPM emissions is currently done by estimating the S and HC mass contributions to vPM, as described in ICAO Doc 9889 [70]. For the HC contribution, this method only uses the gaseous unburned organic emissions (EIHC) along with a power-dependent set of coefficients. The coefficients are based on the ratio of the EIHC to the measured condensed HCs at each of the ICAO LTO points. While this is a useful means of estimating the condensed HCs, it is based on a single engine and represents all the combustion inefficiencies by the EIHC for that one engine. Further research on improving the estimation of the contribution from this source of vPM is required.

The number of non-volatile particles (nvPM) emitted from aircraft engines can be assumed in a good approximation not to undergo transformation processes during the atmospheric transport on a local scale up to several tens of kilometres. Thus, nvPM number can be modelled to a good approximation as a passive tracer without transformation. The mass of aging nvPM particles may change during atmospheric transport due to coating with other material, but the mass of the original nvPM, which is in the UFP range, is usually very small if compared to PM₁₀ from other sources and is currently not of major interest. Accounting for volatile particles (vPM) in local atmospheric dispersion modelling is more challenging. The volatile particles are not emitted but they are formed during transport, with a complex change in diameter (condensation and growth of the particles, re-evaporation) and number (formation of nuclei, agglomeration), in particular, during the first seconds (nucleation) and minutes (before dilution takes over) of atmospheric transport.

In the simplest approach, vPM can be included in a dispersion calculation by means of an effective emission rate (number and mass), as for instance described in the FOA4 method in ICAO Doc 9889 for vPM mass [70]. Such an emission rate implicitly refers to a typical transport time or distance from the engine where it is valid. For vPM number, this approach is probably not so well suited in the near field (up to some 100 m from the engine), given the change of vPM number. A more sophisticated approach would be to account for the formation and change of vPM number and mass by suitable transformation rates of vPM, for example, between different diameter ranges. Beside defining realistic rates depending on engine power setting, exhaust gas properties and ambient conditions, a challenge is to account for the dynamics of both mass and number in such an approach.

8. Conclusions

A number of knowledge gaps are identified in this review together with areas for further investigation. The contribution of smaller non-regulated engine emissions to

nvPM emissions would be helped by more consistent reporting and database development. The estimation of emissions of nvPM for non-LTO parts of the flight (climb and cruise) needs further investigation. Estimating and modelling emissions of vPM emissions from aircraft needs further assessment to understand the contribution made to ambient ultrafine particle number concentrations.

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