Assessing aViation emission Impact on local Air quality at airports: TOwards Regulation - AVIATOR

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Literature survey on particulate matter, comprising UFP microphysical and chemical transformation processes and connected regulatory issues

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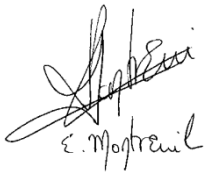
WP LEADER	31/01/20	Dr. Emmanuel Montreuil	
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EXECUTIVE SUMMARY

Emissions from aircraft have adverse effects on the air quality in and around airports, contributing to public health concerns within neighbouring communities. AVIATOR will adopt a multi-level measurement, modelling and assessment approach to develop an improved description and quantification of the relevant aircraft engine emissions, and their impact on air quality under different climatic conditions.

This deliverable provides a survey on comprehensive literature available on ultrafine particles and measurements in airport vicinity. After an initial introduction to ultrafine particles selected results from scientific literature and presented on particulate emissions and ambient measurements downwind from runway in airport vicinity. A section on regulatory issues is included and a comprehensive list of references concludes this deliverable.

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LIST OF ABBREVIATIONS

In this deliverable a number of abbreviations are used.

APU	Auxiliary power unit
CAEP	Committee on Aviation Environmental Protection (within ICAO)
GSE	Ground support equipment
ICAO	International Civil Aviation Organization
MAB	Madrid Air Basin
MMA	Madrid Metropolitan Area
NPF	New particle formation
nvPM	non-volatile particulate matter
PM	Particulate matter
PNC	Particle number concentrations
PNSD	Particle number size distribution
SOA	Secondary organic aerosols
UFP	Ultrafine particles
VOC	Volatile organic compound
WP	Work package

LITERATURE SURVEY ON PARTICULATE MATTER

1. Introduction

Emissions from aircraft have adverse effects on the air quality in and around airports, contributing to public health concerns within neighbouring communities. AVIATOR will adopt a multi-level measurement, modelling and assessment approach to develop an improved description and quantification of the relevant aircraft engine emissions, and their impact on air quality under different climatic conditions.

Aircraft main engines and APU emit volatile precursors of PM and non-volatile PM in form of soot or black carbon with diameter below 100 nm (UFP) and volatile material. In the process of dilution of the exhaust with ambient air, the particles are coated by volatile species and new volatile particles are created from a range of gaseous sulphates and organic precursors (Kärcher et al., 2000; Timko et al., 2013). Superimposed on these processes is the overall physical dynamics of the exhaust flow. While there is an increasing amount of measurement data and theoretical estimates of the mass and number emission of nvPM, major gaps exist in understanding and modelling the subsequent aging processes and formation of particles from volatile and semi-volatile PM. These processes can have strong impact on the resulting pollutant concentration in and around an airport.

Within AVIATOR, work package WP5 "Modelling of plume microphysics, chemistry and dynamics" aims for a deeper understanding of these effects by comprehensive modelling towards more realistic dispersion calculations for key pollutants emitted from aircraft main engines and APU, with focus on non-volatile and volatile UFP (particle composition, number concentration and size distribution). This deliverable contains a comprehensive survey of relevant literature on measurements in airport vicinity with a particular focus on ultrafine particles. This deliverable might be updated during the course of the project, if identified to be efficient in terms of, e.g. new literature or novel regulatory issues.

2. Scientific framework of ultrafine particles

In this survey, we provide an overview on studies dealing with measurement of particulate matter in airport vicinity, with a particular focus on ultrafine particles. In the following we use the term aircraft emission uniformly when authors suggest that emissions originate from aircraft engines, while recognizing that a clear source attribution to engine exit emissions is in general not provided.

2.1. General clarification

Atmospheric particles such as UFP (ultrafine particles < 100 nm) can affect atmospheric chemistry, human health and climate (e.g., Atkinson et al., 2014; Kulmala et al., 2016a, 2004). Descriptions on relevance of ultrafine particles in the atmosphere and characteristics on measurements can be found in recent scientific literature, presenting results from atmospheric measurement campaigns, e.g. Agudelo-Castaneda et al. (2019). It has been reported that UFP have the potential to contribute to an increase in the health impact of aerosols because their fine size allow UFP to penetrate and deposit in the deeper respiratory system, or even penetrate the pulmonary epithelium and olfactory nerve (HEI, 2013) and reach the cardiovascular system and hence other body organs.

2.2. Nature and origin of ultrafine particles

The origin of UFP can be related to specific emission sources (mostly combustion sources), such as road traffic, shipping, industrial sources (e.g., Charron and Harrison, 2003; Cheung et al., 2013; Johnson et al., 2014), as well as airports (Keuken et al., 2015a, 2015b) or newly produced within the atmosphere by homogeneous nucleation processes of gaseous compounds. Thus, UFP may be of both primary and secondary origin.

Nucleation is known as well as gas to particle conversion or new particle formation (Kulmala, 2003). This phenomenon is observed everywhere in the atmosphere (Kulmala et al., 2004) and has been widely studied in the last 25 years. The main aim of most of the studies done in this domain was to find the specific compounds and mechanisms behind new particle formation.

The first compound found to be strongly involved in atmospheric new particle formation was sulfuric acid, but despite its key role in this phenomenon, the particle formation rates observed could not be explained with the concentration of sulfuric acid found in the atmosphere (Sipila et al., 2010). The sources of sulfuric acid present in the atmosphere can be anthropogenic, for example SO_2 coming from transport emissions, or biogenic, for example dimethyl sulfide (DMS) coming from oceans (Jamriska et al., 2008; Kumar et al., 2010; Morawska et al., 2008, Chang et al. 2011).

The second compound found to be a key player in new particle formation together sulfuric acid was ammonia. It was able to stabilize molecular clusters formed by sulfuric acid (Ortega et al. 2008), increasing new particle formation rates (Kirkby et al., 2011). Although the observed enhancement of particle formation rates by these two compounds, the observed formation rates were still far from those observed in the atmosphere. As the stabilizing effect observed was linked to the acid-base interaction between the sulfuric acid and the ammonia, other bases present in the atmosphere, as amines, were investigated. Experiments performed in the frame of CLOUD experiment at CERN showed that indeed amines were able to stabilize sulfuric acid much more than ammonia, leading to formation rates close to those observed in the atmosphere (Almeida et al., 2013). The only question still open is the high new particle formation rates observed in environments are the levels of sulfuric acid were low, like for example boreal forest. In a recent work done as well in the frame of CLOUD experiment, a novel new particle formation mechanism, without the participation of sulfuric acid was discovered. Highly oxidized molecules (HOMs) coming from oxidation of biogenic monoterpenes were found to form new

particles when concentration of sulfuric acid was low (Kirkby et al., 2016). Although this work is limited to biogenic organic compounds, is most likely that anthropogenic organics compounds as well able to follow the same mechanism.

3. Particle measurements including UFP

3.1. Measurement of particles from aircraft

Aircraft are important sources of submicron particles, as shown by studies on emissions from jet engines (Kinsey et al., 2010; Mazaheri et al., 2011; Vander Wal et al., 2014)

Lobo et al. (2015) describe the results of the physical characterization of aircraft engine PM emission measurements conducted during the Delta-Atlanta Hartsfield Study at the Hartsfield-Jackson Atlanta International Airport. Engine exit plane PM emissions were sampled from on-wing engines on several in-service commercial transport aircraft from the fleet of Delta Airlines. They found size distributions to be lognormal in nature with a single mode. The geometric mean diameter was found to increase with increasing engine thrust, ranging from 15 nm at idle to 40 nm at take-off. PM number- and mass-based emission indices were observed to be higher at the idle conditions (4% and 7%), lowest at 15%–30% thrust, and then increase with increasing thrust. In an earlier study Lobo et al. (2007) present results from emission measurements at the engine exit plane (1m) as well as locations 10 and 30 m downstream. They found the soluble mass fraction to be increasing with distance from engine exit plane and with increasing aromatic and sulphur content.

Lobo et al. (2015) present emissions measurements in what they call an advected plume study where over 300 exhaust plumes generated by a broad mix of commercial transports were sampled 100–350 m downwind from aircraft operational runways during normal airport operations. The range of values measured at take-off for the different engine types in terms of PM number-based emission index was between $7 \times 10^{15} – 9 \times 10^{17}$ particles/kg fuel burned, and that for PM mass-based emission index was 0.1 – 0.6 g/kg fuel burned. PM characteristics of aircraft engine specific exhaust were found to evolve over time as the exhaust plume expands, dilutes with ambient air, and cools.

Beyersdorf et al. (2014) study the formation of volatile aerosols (defined as any aerosol formed as the plume ages) in more detail. Tests were performed at varying ambient temperatures (−4 to 20 °C). They reported that at idle, particle number and volume emissions were reduced linearly with increasing ambient temperature, with best fit slopes corresponding to -8×10^{14} particles (kg fuel)$^{-1}$ °C^{-1} for particle number emissions and −10 mm^3 (kg fuel)$^{-1}$ °C^{-1} for particle volume emissions. They indicated that temperature dependency of aerosol formation could have large effects on local air quality surrounding airports in cold regions. Aircraft-produced aerosol number concentrations in these regions would be much larger than levels expected based solely on measurements made directly at the engine exit plane. They reported that the majority (90% at idle) of the volatile aerosol mass formed as nucleation-mode aerosols, with a smaller fraction as a soot coating. Conversion efficiencies of up to 2.8% were measured for the partitioning of gas-phase precursors (unburned hydrocarbons and SO_2) to form volatile aerosols and they reported that highest conversion efficiencies were measured at 45% power.

Miracoli et al. (2011) report on experiments which investigate the effects of photo-oxidation on fine particle emissions from an in-use CFM56-2B gas turbine engine mounted on a KC-135 Stratotanker airframe. Emissions were sampled into a portable smog chamber from a rake inlet installed one-meter downstream of the engine exit plane of a parked and chocked aircraft. This chamber was then exposed to sunlight and/or UV lights to initiate photo-oxidation. They performed separate tests at different engine loads (4, 7, 30, 85%) and found that photo-oxidation created substantial secondary particulate matter (PM), greatly exceeding the direct PM emissions at each engine load after an hour or less of aging at typical summertime conditions. Specifically, they reported that after several hours of photo-oxidation, the ratio of secondary-to-primary PM mass was on average 35 ± 4.1, 17 ± 2.5, 60 ± 2.2, and 2.7 ± 1.1 for the 4, 7, 30, and 85% load experiments, respectively. They concluded that the composition of

secondary PM formed strongly depended on load. The authors reported that at 4% load, secondary PM was dominated by secondary organic aerosol (SOA), while at higher loads, the secondary PM was mainly secondary sulphate.

Miracoli et al. (2011) present that a traditional SOA model that accounts for SOA formation from single-ring aromatics and other volatile organic compounds, underpredicts the measured SOA formation by ~60% at 4% load and ~40% at 85% load when investigating photo-oxidation. They reported that large amounts of lower-volatility organic vapours were measured in the exhaust; they represented a significant pool of SOA precursors that were not included in traditional SOA models. They concluded that these results underscored the importance of accounting for atmospheric processing when assessing the influence of aircraft emissions on ambient PM levels. Models that did not account for this processing would likely underpredict the contribution of aircraft emissions to local and regional air pollution.

Trueblood et al. (2018) report on deployment of a robust hygroscopicity tandem differential mobility analyzer (HTDMA) from a CFM56-2C1 aircraft gas turbine engine. They found that Growth Factor (GF) and the hygroscopy parameter (κ) increased with fuel sulphur content and engine thrust condition, and decreased with increasing dry particle diameter. The highest GF and κ values were found in the smallest particles, typically those with diameters of 10 nm.

3.2. Measurements of particles at urban sites and airport vicinity

Similarly measurements at and near airports show that aircraft are important sources of submicron particles (Hudda et al., 2014; Keuken et al., 2012; Westerdahl et al., 2008), reporting increases in particle number concentrations of UFP near airports (e.g. Westerdahl et al., 2008; Hu et al., 2009; Klapmeyer et al., 2012; Hsu et al., 2012a,b; Brilke et al., 2019).

For example, Hsu et al. (2013) and Stafoggia et al. (2016) detecte substantial increases in total PNC at the airports of **Los Angeles (CA, USA) and Rome Ciampino (Italy),** respectively, in the few minutes after take-offs, especially downwind, while landings make only a modest contribution to ground-level PNC observations.

Hsu et al. (2014) observe that departures and arrivals on a major runway of **Green International Airport (Warwick, RI, USA)** have a significant influence on UFP concentrations in a neighbourhood proximate to the end of the runway.

In a study carried out at **Los Angeles International Airport (CA, USA)**, Hudda et al. (2014) conclude that emissions from the airport increase PNC by 4- to 5-fold at 8–10 km downwind of the airfield, while Shirmohammadi et al. (2017) reporte that the daily contributions of the airport to PNC are approximately 11 times greater than those from three surrounding freeways.

Costabile et al. (2015) report on measurement of Black Carbon (BC) aerosol in an urban airport vs. urban background environment in a **Mediterranean area (Rome**), while proposing a scheme to assess the ultrafine BC in the bulk aerosol.

Keuken et al. (2015) present total and size-resolved PNC, and black carbon concentrations, from measurements near **Schiphol airport** and report an elevation in ultrafine particles by a factor of 3 at 7 km downwind of Schiphol airport. Black carbon (BC) was measured as a sensitive indicator of the mass of PM in combustion emissions (Maricq, 2007). For the measurement site, take-off and climb-out from two specific runways, followed by taxiing and waiting at the gates, and landing at another runway were identified as the four most important sources of elevated PNC downwind of Schiphol at the Adamse Bos. The significance of PN emissions from takeoff and climb-out at Schiphol is in agreement with other studies (e.g. Mazaheri et al., 2011).

Hudda et al. (2016) reporte that average PNC were 2- and 1.33-fold higher at sites 4 and 7.3 km from **Boston (MA, USA)** airport when winds are from the direction of the airfield compared to other directions.

Masiol et al. (2017) report on sampling campaigns new **Heathrow airport** carried out during warm and cold seasons at a site close to the airfield (1.2 km), indicating that size spectra are dominated by ultrafine particles: nucleation particles (< 30 nm) are found to be about 10 times higher than those commonly measured in urban environment. However, they state that despite strong evidence that aircraft (and hence airports) are major sources of UFP, their fingerprint within the particle number size distribution (PNSD) may be difficult to identify. They justify this with four reasons: (i) the nature of semi-volatile compounds emitted by aircraft, (ii) the possible mechanisms of secondary aerosol formation, (iii) the dilution effect and (iv) the similar modal structures of other emission sources concurrently found in cities, such as road traffic (Masiol and Harrison, 2014).

A recent study by Brilke et al. (2019) reports on measurements at a measurement site with strong local pollution sources. Measurements are part of the A-LIFE study aiming to characterize new particle formation (NPF) in the Eastern Mediterranean region. They found nearby **Paphos airport** to be a large emission source for nucleation mode particles and the authors analysed the size distribution of the aircraft emission plumes at approximately 500 m from the main runway. Those events occurring during evening hours that were correlated with high NO and NO_2 concentrations (17 plumes) were averaged over the length of each single emission plume. Clearly, a substantial fraction of the particle size distribution is distributed in the sub-10 nm size region. The mode diameter of the aircraft emission plumes was inferred by averaging all 17 airport emission events, and the mode diameter of the averaged size distribution of all events is given at 12.6 nm in this recent study. Total particle concentration measured during this study exceeds 10^5 cm^{-3} during plume-impacted time intervals. Authors report that they meet the challenge of measuring sub-10 nm particles using the newly-developed DMA-train (Stolzenburg et al., 2017).

The analysis yielded 9 NPF events in 27 measurement days from the combined analysis of the DMA-train, MPSS and trace gas monitors. Growth rate calculations were performed and a size-dependency of the initial growth rate (< 10 nm) is observed for one event case. Fast changes of the sub-10 nm size distribution on the timescale of a few minutes are captured by the DMA-train measurement during early particle growth and are discussed in a second event case. In 2 cases, particle formation and growth were detected in the nucleation mode size range which did not exceed the 10 nm threshold. This finding implies that NPF likely occurs more frequently than estimated from studies where the lower nanometre size regime is not covered by the size distribution measurements.

A recent review of aircraft and airport emissions (Masiol and Harrison, 2014) concluded that 'despite the increased attention given to aircraft emissions at ground level and air pollution in the vicinity of airports, many research gaps remain', including emissions and dispersion of submicron particles.

Generally, studies performed within or close to airports have reported increases in particles ranging from 4 to 100 nm in diameter and mostly distributed in the nucleation range (< 30 nm). Mazaheri et al. (2009) showed a main nucleation mode and an accumulation mode (40–100 nm) more evident during take-offs; Keuken et al. (2015) reported PNSD dominated by 10–20 nm particles in an area affected by emissions from **Schiphol airport** (the Netherlands); Hudda and Fruin (2016) found strong increases in particles smaller than 40 nm downwind of **Los Angeles International Airport**; Ren et al. (2016) showed that particles peaking at 16 nm dominate the PNSD at various distances from the runway of **Tianjin International Airport**, China; Masiol et al. (2016) reported that the fingerprint of aircraft emissions sampled under real ambient conditions at the airport of **Venice (Italy)** has a main mode at approx. 80 nm and a second mode in the nucleation range below 14 nm.

3.3. Field campaign in Madrid targeting UFP and ozone

Vertical soundings in a suburban environment in Madrid (Spain) during a field campaign in July 2016 demonstrate that ultrafine particles (UFP) are formed exclusively inside the mixed layer (Carnerero et al., 2018). As convection becomes more effective and the mixed layer grows, UFP are detected at higher levels. The morning soundings revealed the presence of a residual layer in the upper levels in which aged particles (nucleated and grown on previous days) prevail. The particles in this layer also grow in size, with growth rates significantly smaller than those inside the mixed layer. Under conditions with strong enough convection, the soundings reveale homogeneous number size distributions and growth rates at all altitudes, which follow the same evolution at the other stations considered in this study. This indicates that UFPs are detected quasi-homogenously in an area spanning at least 17 km horizontally. The NPF events extend over the full vertical extension of the mixed layer, which can reach as high as 3000 m in the area, according to previous studies.

Various studies have reported that the photochemical nucleation of new ultrafine particles in urban environments within high insolation regions occurs simultaneously with high ground ozone (O_3) levels. During this field campaign in July 2016, additionally atmospheric dynamics leading to summer O_3 episodes in the Madrid air basin (central Iberia) by means of measuring a 3-D distribution of concentrations for both pollutants were evaluated (Querol et al., 2018). The results demonstrate the concatenation of venting and accumulation episodes, with relative lows (venting) and peaks (accumulation) in O_3 surface levels. Regardless of the episode type, the fumigation of high-altitude O_3 (arising from a variety of origins) is found to contribute the major proportion of surface O_3 concentrations. Accumulation episodes are characterised according to Querol et al. (2018) by a relatively thinner planetary boundary layer (< 1500 m at midday, lower in altitude than the orographic features), light synoptic winds, and the development of mountain breezes along the slopes of the Guadarrama Mountain Range (located W and NW of the Madrid Metropolitan Area (MMA), with a maximum elevation of > 2400 m above sea level). According to the authors this orographic-meteorological setting causes the vertical recirculation of air masses and enrichment of O_3 in the lower tropospheric layers. When the highly polluted urban plume from Madrid is affected by these dynamics, the highest O_x (O_3+ NO_2) concentrations are recorded in the Madrid Metropolitan Area.

A recent study (Saiz-Lopez et al., 2017) reporte an increase of 30–40% in ambient air O_3 levels, along with a decrease of 20–40% in NO_2, from 2007 to 2014 in Madrid, which may have led to large concentration increases of up to 70 and 90% in OH and NO_3, respectively, thereby changing the oxidative capacity of this urban atmosphere. They report that they do not know if this increase is due to a decrease in the NO titration effect or to the fact that O_3 formation is dominated by VOCs since urban areas are characterised by “VOC-limited” conditions, and a reduction in NO_x emissions might yield an increase in O_3 formation.

In addition to primary emissions, nucleation or new particle formation (NPF) processes give rise to relevant contributions to the urban ambient air UFP concentrations, mostly during photochemical pollution episodes in spring and summer (Brines et al., 2015, and references therein). Ambient conditions favouring urban NPF are high insolation, low relative humidity, available SO_2 and VOCs, and a low condensation sink potential (i.e. a relatively clean atmosphere with low surface aerosol concentrations; Kulmala et al., 2000, 2004; Kulmala and Kerminen, 2008; Sipilä, et al., 2010; Salma et al., 2016).

Querol et al. (2018) evaluate the temporal and spatial variability of O_3 and UFPs in the Madrid Air Basin (MAB, 4–20 July 2016) to investigate the causes of acute summer episodes of both pollutants, ozone and UFP, and possible inter-relationships.

4. Airport dispersion modelling

Dispersion modelling at and around airports is demanding. Aircraft (main engine and APU) and aircraft-related emission sources (GSE) resemble a complex source system with time- and space-varying emissions. In addition, there are other emission sources that may have a major impact on local air quality, in particular road traffic at and around the airport.

It is important to note that a dispersion modelling system is a model chain consisting of different, often independent parts: emission model, exhaust dynamics model, meteorological boundary layer model, wind field model, dispersion model, physical/chemical conversion model, deposition model. For airports, modelling of thrust-dependent emissions and exhaust dynamics of typically several 1000 to 100'000 movements consisting of different aircraft types with various space- and time-dependent pathways is challenging. In contrast to road traffic, exhaust dynamics affects the dispersion on a much larger scale.

Within ICAO, several dispersion modelling systems that allow emission and dispersion calculations for aircraft operations at and around airports have been evaluated and are in use for CAEP work (CAEP LAQ models), see ICAO environmental reports (ICAO 2007, 2010, 2013, 2016). Some of these models are routinely applied also outside ICAO for scientific studies, air quality forecasts, and airport licensing procedures (see e.g. Yamartino et al., 2004; Janicke et al., 2007; PSDH, 2006; Di Sabatino et al., 2011; Ruf et al., 2013; Lorentz et al., 2019; Voogt et al., 2019). Apart from these, there exists a variety of other models, like for example CFD models applied to aircraft and airports (Ghedhaïfi, 2010; Montreuil et al., 2018).

The ICAO document 9889 (ICAO, 2011, 2015), Airport Air Quality Manual, provides some guidance for modelling emission and dispersion from aircraft. Other guidance is provided by national standards. However, there is no international standard like for example in noise regulation and there is, at present time, no "gold standard" by which such a complex model system could be evaluated.

For ultrafine particles from aircraft, dispersion modelling has been hampered so far by the lack of standardized, engine-specific emissions. For non-volatile particles, major progress has been made over the last years (see e.g. ICAO Environmental Report 2019; Agarwal et al., 2019). For volatile particles, modelling of emission and further transformation during the atmospheric transport of volatile particles is subject to intensive current research.

5. Regulatory aspects

In terms of regulatory aspects for particulate matter particular considerations of measurements and pre-requisites for regulations have to be considered. Particle number counting instruments measuring ambient air particle concentrations usually count all UFP particles, irrespective of their origin and chemical composition. Such measurements cannot and must not be used to relate UFP concentration results to health effects. As particle number counters report total number of particles without any information on chemical composition and without knowledge of health impact of different species, the measured levels of UFP number concentrations represent not more than an inventory of the UFP concentrations observed at and near the airport.

UFP are very different in shape, from liquid droplets to solid complex, non-symmetrical structures. The sizing instruments report the so called “mobility diameter”, which does not take characteristics into account.

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