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The Time of Flight – Aerosol Mass Spectrometer (C-ToF AMS) is developed by Aerodyne Research Inc. (Billerica, USA) for fast (real-time) ground-based studies of the physical and chemical properties of nonrefractory (< 300-600°C) aerosols with particle diameter (d_n) of 80 - 1000nm (Jayne et al. 2000; Drewnick et al., 2005; Canagaratna et al., 2007^{*}). Since September 2007 the first C-ToF AMS (in the following F-AMS) of this type in France was adapted by the Laboratoire de Météorologie-Physique, University of Clermont-Ferrand, for the airborne application to perform first test flights onboard the French ATR-42 in January 2008. For aircraft-based measurements a pressure controlled inlet (PCI) (similar design as described by Bahreini et al. 2008^{*}) was used to maintain constant pressure conditions (~377 hPa) upstream of the aerodynamic lens (AL) as the aerosol transmission through the AL strongly depends on the pressure gradient along the AL (Zhang et al., 2002 * & 2004 *; Liu et al. 2007*). The major part of the F-AMS is placed in one ATR-42 standard rack. Two remaining AMS controller boxes need less than ¹/₂ of a second ATR rack.

Post-flight Ionization efficiency (IE) calibrations have been performed prior and after the campaign as well as five times during the campaign period. The resulting IE showed a good stability and reproducibility (< 6% decrease, caused by detector's ageing, throughout the campaign). The data shown here are corrected by collection efficiency (CE) factors which were obtained according to Crosier et al. 2007^{*}. All flight data were treated according to procedures provided within the data processing software "Squirrel" * and in accordance to the standards defined and currently used by the Aerodyne AMS operators community (http://cires.colorado.edu/jimenez-group/wiki/index.php/Field_Data_Analysis_Guide).

Figure 1: Major part of the F-AMS mounted inside an ATR-42 rack.

For EUCAARI/IMPACT the F-AMS was operated in alternating mode of Mass Spectrometer (MS) measurements (aerosol chemical composition) and Particle Time-of-Flight (PToF) measurement (aerosol size distribution) delivering a full MS & PToF cycle every 25 seconds. This study is exclusively focused on the MS measurements which could have been proven by one in-flight comparison with an other AMS of the same type onboard a second aircraft (cf. next section). Altogether 7 clear-sky and 14 cloud-related flight have been performed. For aerosol measurements the F-AMS sampled aerosol via an isokinetic and isoaxial, shrouded inlet, developed by Meteo France. In-cloud measurements, when the aerosol sampling was switched to a CVI inlet, are currently still in analyzing process.



Figure 2: F-AMS versus UK-AMS. Correlation of chemical species measured during 35 minutes wing –to wing maneuver on May 09

For the total mass concentration (TMC) the correlation yields the UK-AMS to measure 31% lower mass loadings compared to the F-AMS (not shown herein). The most likely factor causing this discrepancy is the different aerosol inlet systems used on board the BAe-146 (Rosemount -inlet, Crosier et al. 2007^{*}) and the ATR-42 though the measurements agree to within stated tolerances. The TMC measured on board the ATR-42 was calculated by combining results of a Scanning $\stackrel{\square}{\rightarrow}$ Mobility Particle Sizer (SMPS) and a Passive Cavity Aerosol Spectrometer Probe (PCASP) for particles with $d_p < 800$ nm. The comparison yields the SMPS-PCASP-systems generally to measure about 30% more than the F-AMS. The F-AMS detects only the non-refractory (at 600°C) particles while the SMPS-PCASP also considers refractory aerosol material. Thus it is expected that the SMPS-PCASP exceeds the AMS measurement. However, SMPS-PCASP does not provide an optimal reference for the AMS as the measurement principles of both are not equivalent. This comparison is only used to exclude that the F-AMS fully overestimates the TMC.

Airborne C-ToF AMS measurements during EUCAARI/IMPACT on board the French ATR-42

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For in-flight data quality proof the ATR-42 flight on May 09 was dedicated to a comparison, for instrumental proof, with an AMS of the same type operated by the University of Manchester onboard the British FAAM BAe-146. Differing from the F-AMS the British AMS (UK-AMS) was not equipped with a PCI. A wing-to-wing manoeuvre was performed for a duration of about 35 minutes at two different flight altitudes – 14 minutes at 1.2 km (asl), followed by about 7 minutes of ascent and a further 14 minutes at 2.7 km (asl). Thus, the instruments measured inside as well as outside the atmospheric boundary layer, covering a range of aerosol loading into the AMS of almost two orders of magnitude between 0.2 and 10 µg m⁻³ total. The figure 2 shows the time series plots and the correlation between both instruments according to aerosol chemical species of Organics, Nitrate, Sulphate, and Ammonium. Chloride which is also measured by the AMS is not shown (for both AMS below the detection limit). Both instruments agree within 20 % for Nitrate, Sulphate and Ammonium. Organics measured with the F-AMS, particularly for enhanced particle loading, yields up to two times higher values than the UK-AMS which is most likely caused by an higher F-AMS vaporizer temperature (~600°C) compared to 350°C the UK-AMS was operated with.



Figure 3: Total mass concentration measured by AMS and combined from SMPS-PCASP measurements for particles with $d_p < 0.8 \ \mu m$. Correlation for duration of ~3 hours during the flight on May 09.



meteorological during the first 12 days over Central Europe was dominated by a stable anticyclone effecting a $_{50^{\circ}N}$ blocking of long-range aerosol transport into sampling area. This stable air mass was neither influenced by cloud formation nor 45° precipitation. 5 flights have been performed during the time of the prevailing anticyclonic situation between May 5 and 11 (1st clearsky period). Two further clear-sky flights have been performed on

3.0 a

25

20

1.5

- 20

Figure 4: ATR clear-sky flight tracks during EUCAARI/ IMPACT mainly within air sectors Box A or B May 28 & 29 after cyclone-driven air mass exchange and periodic precipitation events (2nd clear-sky period).

Some result from clear-sky aerosol measurements are shown in figures 6a - c. The comparison of up- and downwind Rotterdam (RTM) shows slightly different boundary layer (BL) conditions. Upwind (East) of RTM the BL is not clearly stated neither by Θ nor by the TMCs. Downwind (West of RTM - North Sea and Channel) the BL is sharply separated from the free troposphere (FT) in the Θ profiles and in the TMCs at ~1.5km altitude. Below 1.5 km, in both cases, the TMCs are of the same order of magnitude and show Organics and Nitrate as dominating species, indicative for the aerosol's anthropogenic (industrial) origin. Below 1km and downwind RTM Sulphate and Nitrate are enhanced compared to upwind composition which suggests to be originated by fuel burning and/or the petro-chemical industry, located in the vicinity of RTM, possibly also influenced by ship traffic. Above 1.5km, generally, Nitrate decreases while Organics and Sulphate remained dominant. Here, aerosols most likely are originated from fuel burning and are transported from Eastern Europe into sampling areas (Figure 5). The 💆 1.5 TMC plot of figure 6c) shows both median profiles from Ξ figure 6a) and b) together with data of one flight on May 28, after cyclone-driven air mass exchange. The to comparison with the median profiles demonstrates the strong tropospheric vertical mixture on May 28 compared to earlier measurements (nearly constant TMC of ~10 µg m⁻³ from ground level up to 3km). The chemical composition shows Sulphate to be most prominent while Nitrate already at ~0.5 km decreases and plays no dominating role aloft.

Close to ground the Sulphate content is of the same order of previous measurements but between 1.5-2.2km Sulphate exceeds by one order of magnitude - above 2.3 km still by a factor of five. Figure 5 shows the Mediterranean Sea as the air mass origin

(Sardinia, Corsica, North Italy) moving within 24

hours via Rhône Valley and West-France or across

the Alps into the sampling area.



Crosier et al. (2007), by airborne studies in the FT above the Po Valley and Adriatic Sea, as well as Schneider et al. 2004, at Crete (MINOS), found Sulphate-dominated aerosol in significant quantities originated from fossil fuel burning in South-East Europe forming the regional pollution and characterizing a typical Mediterranean pollution aerosol – as shown with this study – with impact also on Central-European air quality.

Beside this, further analyses of organic mass spectra (related to the ageing state of sampled aerosol) and single features with high potential to be caused by penetrating ship emission plumes at 0.3km flight altitude above the North Sea are focused by current investigations.

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* All publications referred to herein are public available at http://cires.colorado.edu/~jjose/ams-papers.html



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different altitude. calculated with HYSPLIT



Figure 6: TMC (left column), aerosol chemical composition (mid-left), fractional chemical composition (mid-right) and the potential temperature Θ (right column). Panels a) 3 flights upwind RTM b) 4 flights downwind RTM as the wind mainly approached the air space of measurements from Eastern Europe (Poland Czech Republic, cf. figure 5) and c) one single flight on May 28 - air mass transported from South