

ERC Instrument for Chemical composition of Aerosols - Aerosol Mass Spectrometer (ERICA-AMS) Data – Readme

This is the read me file for the ERICA-AMS dataset during ACCLIP 2022 campaign including the mass concentrations of non-refractory particle compounds (nitrate, sulphate, ammonium and organic).

Author(s)

- Lead: Oliver Eppers¹

Email: o.eppers@mpic.de; Phone: +49 6131 305 5212; ORCID: 0000-0001-9487-4603

- Corresponding authors: Oliver Appel^{2,1}, Philipp Brauner¹, Antonis Dragoneas¹, Fatih Ekinci^{2,1}, Franziska Köllner^{2,1}, Stephan Borrmann^{2,1}

¹Particle Chemistry Department, Max Planck Institute for Chemistry, Mainz, Germany

²Institute for Atmospheric Research, Johannes Gutenberg University, Mainz, Germany

1.0 Data Set Description

The data set provides mass concentrations of non-refractory particle compounds (nitrate, sulphate, ammonium and organics), measured onboard the NCAR GV aircraft during the ACCLIP campaign in July/August 2022. This data set is the second submission (Revision 1) and was last checked on March 24, 2023. The data set include measurements from research flights 2 - 12 (August 4 - August 26, 2022). The location of the research aircraft during the measurements can be found in the navigation, state parameter and microphysics dataset of the NCAR GV measurements (NSF/NCAR GV Team, 2023).

The status of the data is final, except the file from the second research flight (August 4, 2022). The latter one is not selected for in-cloud measurement periods due to missing cloud flag. We will provide the revised version as soon as the cloud information is available.

The frequency of data collection is around 0.1 Hz based on the measurement cycle frequency of ERICA-AMS. However, there are irregular time shifts every 5 minutes due to a time lag while generating a new file for the data acquisition.

2.0 Instrument Description

The ERICA instrument (ERC Instrument for the Chemical Composition of Aerosols) combines two commonly adopted methods for in situ real-time mass spectrometric analysis of aerosol particles. The first method is the laser desorption ionization (LDI) method followed by the thermal desorption with subsequent electron-impact ionization (TDI) as implanted in the Aerodyne aerosol mass spectrometer (AMS) (Drewnick et al., 2005; Canagaratna et al., 2007). The ERICA has been described in detail by Hünig et al. (2022) and Dragoneas et al. (2022).

This data set refers to the ERICA-AMS (ERICA aerosol mass spectrometer) measurements based on the thermal desorption with subsequent electron impact ionization method. In analogy to the Aerodyne AMS principle (Canagaratna et al., 2007), the incoming aerosol particles hit a tungsten vaporizer, where the non-refractory components are evaporated at around 600°C. A filament provides electrons (70 eV) for electron impact ionization of the vapor molecules emanating from the vaporizer. The generated cations are injected into a compact time-of-flight mass spectrometer (C-ToF-MS, Tofwerk AG, Switzerland) and eventually detected by its MCPs. A shutter unit upstream of the vaporizer switches between an open and closed position every 5 seconds. Thereby, the particle beam hit the tungsten vaporizer during the first 5 seconds, whereas only background air reaches the vaporization region for the closed shutter position. The size range for particle detection in the ERICA-AMS is assumed to be the same as published in Xu et al. (2017) for the deployed aerodynamic lens type at the inlet of ERICA ranging from particle diameters of 120 nm to 3.5 μm (d_{50} cutoff diameters).

3.0 Data Collection and Processing

The ionized vapour is extracted by an extraction pulser working at a frequency of 80 kHz, resulting in a maximum m/z of above 280. The data acquisition card averages 32 000 extractions to one raw spectrum that is then transmitted to the computer and represents 0.4 s of measurement. One measurement cycle of the ERICA-AMS consists of 10 s or 25 raw spectra. 11 raw spectra (4.4 s) are taken while the shutter is open as representative for the aerosol measurement, another 11 raw spectra (4.4 s) as background measurement (i.e. residual vacuum signal) while the shutter is closed. Three raw spectra (1.2 s) are needed for the switching of the shutter and are discarded due to an undefined shutter position. One data file is recorded every five minutes. In TofWare 2.5.7 we adjusted the settings to comply with the ERICA-AMS data. Executing "setigorooption pounddefine=tw_ACSM_diffSticks_dontNorm28" and recompiling as well as activating the button "Background is blocked beam (adv)" prevents TofWare from normalization of each spectrum to m/z 28. This normalization is only practical in ACSM instruments, which the software was designed for. Due to our inhouse-built shutter unit, the software is unable to read the shutter position, thus

we set it manually to 1-12 (Total) and 14-24 (Bkgnd), switch buf 13. The triggering of the shutter directly by the ADQ precludes potential shifts in the time scheme. We also insert the sample flow rate into the instrument of $1.45 \text{ cm}^3 \text{ s}^{-1}$ and the IE calibration factor of $4415 \text{ ions pg}^{-1}$. This value was determined by the average of four IE calibration measurements with ammonium nitrate during and after the campaign. In addition to ammonium nitrate, measurements with ammonium sulphate were performed. The calibration procedure is described in more detail in the supplemental material of Appel et al. (2022). From these calibration measurements, the relative ionization efficiency (RIE) of ammonium and sulphate was obtained with $\text{RIE}(\text{NH}_4) = 4.188$ and $\text{RIE}(\text{SO}_4) = 1.053$. In order to convert the raw spectra into an integrated unit mass resolution (UMR) spectrum, also called stick spectrum, an m/z calibration is needed to identify the signal peaks. We use a three-parameter fit of the form $p_1 \cdot m^{p_3} + p_2$ to the masses of CH^+ ($m/z = 13.0078$), O_2^+ (31.9898), SO_2^+ (63.9619), $^{182}\text{W}^+$ (181.948), $^{184}\text{W}^+$ (183.951) and $^{186}\text{W}^+$ (185.954). A time series of calibration values for each data point was used. TofWare calculates a time series of stick spectra according to the predefined values. The aerosol signal is represented by the "diff" spectrum, which is the difference of the measurement with an open shutter and a closed shutter.

The attribution of the UMR signals to the chemical species of interest (e.g. nitrate, organic, ammonium, sulfate) is done by means of a so-called fragmentation table (Allan et al., 2004).

The determination of detection limits is based on the method described in S1.3 in the supplemental material of Appel et al. (2022). The precision of the data is determined based on the zero filter measurements during the individual flights. In more detail, the precision is defined as the 2σ variation of the mass concentration during a filter measurement. The accuracy of the ERICA-AMS data is $\sim 30\%$ based on Canagaratna et al. (2007) and Middlebrook et al. (2012).

The collection efficiency (CE) of the vaporizer is depending on e.g. the chemical constituents of the aerosol particle. Middlebrook et al. (2012) developed a method to estimate the collection efficiency based on the measured chemical composition. We used this method to correct the acquired mass concentrations, since a high ammonium nitrate fraction as well as very acidic aerosol can be found in the ATAL and lower stratosphere, respectively. However, during the majority of the flights, the measured ammonium mass concentrations were below the detection limit. Thus, the CE was assumed to be 0.5 for those periods with low ammonium content.

We removed data from the zero filter measurements during the flights as well as data that were collected during in-cloud measurement periods based on the cloud flag provided by Sarah Woods. The cloud flag is included in the navigation, state parameter and microphysics dataset (NSF/NCAR GV Team, 2023).

For research flights 1, 13 and 14, no final data from ERICA-AMS are available, yet, due to signal reflection issues in the AMS raw spectra. With further extensive data evaluation being necessary for these flights, the final data might be available in a future revision of the ERICA-AMS data set.

4.0 Data Format

The ERICA-AMS datasets for the individual ACCLIP flights are provided in the ICARTT format. The filename for the ERICA-AMS products from ACCLIP 2022 are given in the following format:

ERICA-AMS_AerosolComp_GV_20220804_RF02_R1.ict

ERICA-AMS is the acronym for the instrument, AerosolComp is the abbreviation of the data product type (aerosol chemical composition), GV is the platform (NCAR GV aircraft), 20220804 is the date of take-off time given as YYYYMMDD format (UTC), RF## is the flight mission number during ACCLIP campaign, R1 is the Revision no. 1.

List of parameters:

- Mass concentration of particulate nitrate (shortname: nitrate), unit: $\mu\text{g m}^{-3}$
- Detection limit of nitrate mass concentration (shortname: DL_nitrate), unit: $\mu\text{g m}^{-3}$
- Mass concentration of particulate sulphate (shortname: sulphate), unit: $\mu\text{g m}^{-3}$
- Detection limit of sulphate mass concentration (shortname: DL_nitrate), unit: $\mu\text{g m}^{-3}$
- Mass concentration of particulate ammonium (shortname: ammonium), unit: $\mu\text{g m}^{-3}$
- Detection limit of ammonium mass concentration (shortname: DL_ammonium), unit: $\mu\text{g m}^{-3}$
- Mass concentration of particulate organic (shortname: organic), unit: $\mu\text{g m}^{-3}$
- Detection limit of organic mass concentration (shortname: DL_organic), unit: $\mu\text{g m}^{-3}$

5.0 Data Remarks

For research flights 1, 13 and 14, no final data from ERICA-AMS are available, yet (see 3.0). For RF02, the data quality is only prefinal since in-cloud measurement periods could not be determined due to missing cloud flag. We will provide the revised version as soon as the cloud information is available.

For RF 03, 04, 05 and 07, the cloud flag was missing for some periods, in particular for the beginning and/or end of the flight. Therefore, the ERICA-AMS data might be affected by cloud artefacts during these periods.

Periods of zero filter measurements are removed from the dataset as well. In the following, an overview of the zero filter periods is given for the individual flights:

Flight no.	Time periods of zero filter measurements (UTC)
RF02	02:08:11 - 02:23:56 05:12:53 – 05:17:23
RF03	05:09:10 – 05:14:12 07:00:36 – 07:05:38
RF04	02:01:08 – 02:10:51 04:46:03 – 04:51:04
RF05	05:52:36 – 05:56:28 07:31:29 – 07:34:41
RF06	01:47:00 – 01:52:02 04:04:14 – 04:09:16
RF07	01:38:21 – 01:43:22 07:28:34 – 07:33:16
RF08	01:52:53 – 01:58:05 05:12:58 – 05:18:09
RF09	01:36:56 – 01:42:38 06:50:18 – 06:55:30
RF10	01:57:03 – 02:02:05 05:28:53 – 05:33:55
RF11	01:40:26 – 01:45:28 06:12:15 – 06:17:27
RF12	01:45:07 – 01:49:59 07:01:01 – 07:05:32

6.0 References

Allan, J. D. et al.: A generalised method for the extraction of chemically resolved mass spectra from Aerodyne aerosol mass spectrometer data, *Journal of Aerosol Science*, 35, 909-922, DOI: 10.1016/j.jaerosci.2004.02.007, 2004.

Appel, O. et al.: Chemical analysis of the Asian tropopause aerosol layer (ATAL) with emphasis on secondary aerosol particles using aircraft-based in situ aerosol mass spectrometry, *Atmos. Chem. Phys.*, 22, 13607–13630, DOI: 10.5194/acp-22-13607-2022, 2022.

Canagaratna, M.R. et al.: Chemical and microphysical characterization of ambient aerosols with the aerodyne aerosol mass spectrometer, *Mass Spectrom. Rev.*, 26, 185-222, DOI: 10.1002/mas.20115, 2007.

Dragoneas, A. et al.: The realization of autonomous, aircraft-based, real-time aerosol mass spectrometry in the upper troposphere and lower stratosphere, *Atmos. Meas. Tech.*, 15, 5719–5742, DOI: 10.5194/amt-15-5719-2022, 2022.

Drewnick, F. et al.: A New Time-of-Flight Aerosol Mass Spectrometer (TOFAMS) – Instrument Description and First Field Deployment, *Aerosol Science and Technology*, 39:7, 637-658, DOI: 10.1080/02786820500182040, 2005.

Hünig, A. et al.: Design, characterization, and first field deployment of a novel aircraft-based aerosol mass spectrometer combining the laser ablation and flash vaporization techniques, *Atmos. Meas. Tech.*, 15, 2889–2921, DOI: 10.5194/amt-15-2889-2022, 2022.

Middlebrook, A. M. et al.: Evaluation of Composition-Dependent Collection Efficiencies for the Aerodyne Aerosol Mass Spectrometer using Field Data, *Aerosol Science and Technology*, 46:3, 258-271, DOI: 10.1080/02786826.2011.62004, 2012.

NSF/NCAR GV Team: ACCLIP: Low Rate (LRT - 1 sps) Navigation, State Parameter, and Microphysics Flight-Level Data. Version 1.0 (Version 1.0) [Data set]. UCAR/NCAR - Earth Observing Laboratory. DOI: 10.26023/X2WP-S330-G807, 2023. Accessed 23 Mar 2023.

Xu, W. et al.: Laboratory characterization of an aerosol chemical speciation monitor with PM_{2.5} measurement capability, *Aerosol Science and Technology*, 51:1, 69-83, DOI: 10.1080/02786826.2016.1241859, 2017.