Submicron aerosol composition measurements during ACRIDICON-CHUVA: Indications for secondary organic aerosol formation in the upper tropical troposphere

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ACRIDICON-CHUVA Workshop

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Instrument C-ToF-AMS

- Compact Time-of-Flight Aerosol Mass Spectrometer
- Sub-micron non-re refractory particle composition (approx. PM1)
- Species:
  - organics, sulfate, nitrate, ammonium, chloride
- Quantitative measurements
- Time resolution: 30 s
- Size range: 40 – 800 nm
- Expected results:
  - Particle composition
  - Mass concentration
  - Vertical profiles
Instrument C-ToF-AMS

Pressure controlled inlet

HASI
CVI

HV pulser
timing controller

preamp
averaging ADC
PC

ion reflector
MCP detector
orthogonal extractor

aerosol vaporizer

ionization chamber

particle ToF measurement

chopper

tungsten filament

aerosol inlet (aerodynamic lens)

turbo molecular pumps

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Instrument C-ToF-AMS
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- HASI
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- chopper
- particle ToF measurement
- aerosol vaporizer
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Vertical Profiles ACRIDICON-CHUVA Aerosol Measurements

- Organics
- Nitrate
- Sulfate
Vertical Profiles ACRIDICON-CHUVA Aerosol Measurements

Maximum at low altitudes

- Organics
- Nitrate
- Sulfate
Vertical Profiles ACRIDICON-CHUVA Aerosol Measurements

Decay at middle altitudes

Organics  Nitrate  Sulfate
Vertical Profiles ACRIDICON-CHUVA Aerosol Measurements

Increase at high altitudes for organics and nitrate but not for sulfate.
Vertical Profiles ACRIDICON-CHUVA Aerosol Measurements

- Increase at high altitudes for organics and nitrate but not for sulfate
- Vertical or long range transport?
- Cloud processing?
- Formation?
- Condensation?
Vertical Profiles ACRIDICON-CHUVA Aerosol Measurements – AC 11

AC 11: Outflow flight with tracer sampling

- Organics
- Nitrate
- Sulfate
Vertical Profile AC 11

ACRIDICON-CHUVA
AC 11, 16 Sept 2014
C-ToF-AMS
preliminary

- Altitude [km]
- Aerosol mass concentration [µg/m³]

- Organics
- Nitrate
- Sulfate
Flight Path AC11
Flight Path AC 11
colour coded with organic mass concentration
Flight Path AC 11
colour coded with organic mass concentration
Flight Path AC11

High altitude (11-12 km)

Outflow

Influence of Manaus
Time Series AC 11

Influence of Manaus
Influence of Manaus
Influence of Manaus:
Similar aerosol composition at low altitudes

HASI only

Influence of Manaus:
Similar aerosol composition at low altitudes
ACRIDICON-CHUVA
AC11, 16 Sept 2014
C-ToF-AMS
preliminary

HASI only

High altitude (11 – 12 km)
Time Series AC 11

High altitude (11 – 12 km):
Increase of organics and nitrate
Time Series AC 11

![Graph showing time series data for aerosol mass concentration and altitude over time. The graph includes lines for different categories such as Organics, Nitrate, Sulfate, Ammonium, and Chloride, with specific dates and times highlighted for Outflow and HASI only.]
Time Series AC 11

Increase of organics and nitrate at high altitudes

Outflow

HASI only
Mass Spectra

- Low altitudes: Manaus plume
  - Organics, NO$_3$, SO$_4$
  - Organic spectra: aged oxidized organic aerosol (OOA)

- High altitudes: 11-12 km
  - Organics, NO$_3$
  - Organic spectra: OOA, isoprene epoxydiol secondary organic aerosol (IEPOX-SOA)

- Outflow:
  - Organics, NO$_3$
  - Organic spectra: OOA, IEPOX-SOA
Aging of Organic Aerosol

- $f_{43}$: fraction of signal at m/z 43 to organic signal
- $f_{44}$: fraction of signal at m/z 44 to organic signal
Aging of Organic Aerosol

- \( f_{43} \): fraction of signal at m/z 43 to organic signal
- \( f_{44} \): fraction of signal at m/z 44 to organic signal
- Ratio of \( f_{43} \) and \( f_{44} \): information of oxidation state \( \rightarrow \) aging of organic aerosol

Ng et al. ACP 2010
Aging of Organic Aerosol
Aging of Organic Aerosol

Aged organic aerosol
Low altitude
Mass Spectra: aged OOA
$D_p \approx 200$ nm

Fresher organic aerosol
High altitude
Mass Spectra: OOA, IEPOX-SOA
$D_p \approx 100$ nm
Aging of Organic Aerosol

CVI

Residual particles: even younger organics
High altitude
Mass Spectra: Hydrocarbonlike organic aerosol (HOA)
IEPOX-SOA

- Isoprene EPOXidiol Secondary Organic Aerosol
- Oxidation of isoprene
- Source of isoprene: vegetation
- Indications for IEPOX-SOA in mass spectra at high altitudes
  - Marker peak: m/z 82
  - f82: fraction of signal at m/z 82 to organic signal

Robinson et al. ACP 2011; Hu et al. ACP 2015
IEPOX-SOA

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Robinson et al. ACP 2011; Hu et al. ACP 2015
Summary AC 11

- Increase of organic and nitrate mass concentration with altitude
- No such increase observed for sulfate
- Organic aerosol: younger aerosol at higher altitudes
  → aerosol formation (by condensation and/or nucleation) at higher altitudes
- Indications for Isoprene EPOXidiol Secondary Organic Aerosol (IEPOX-SOA)
- Cloud residues: even younger organic aerosol, mass spectra similar to HOA
Thank you for your attention!

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