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GLORIA observations of pollution tracers C_2H_6, C_2H_2, HCOOH, and PAN in the North Atlantic UTLS region

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Summary

The Gimballed Limb Observer for Radiance Imaging of the Atmosphere (GLORIA) is an imaging Fourier Transform Spectrometer (iFTS) using a 2-dimensional detector array to record emission spectra in the mid-infrared region with high spectral resolution. We report results from the Wave-driven ISentropic Exchange (WISE) aircraft campaign carried out from Shannon (IRL) in autumn 2017.

- Pollutant species like C_2H_6, C_2H_2, HCOOH, and PAN which are produced at distinct source regions near the ground and transported to remote regions due to their atmospheric lifetime of several weeks were observed with high vertical resolution.
- Enhanced volume mixing ratios of these molecules were detected along some parts of the flight track (mainly during flight #2) in the upper troposphere and lowermost stratosphere (UTLS). The origin of this pollution is mainly Asia and North America
- Simulations of the Chemistry Climate Model EMAC and the ECMWF CAMS reanalysis also show enhanced values in the region GLORIA does. However, apart from PAN (EMAC), simulations by CAMS and, to a lesser extent, EMAC, underestimate the elevated amount of observed VMR features.

GLORIA instrument

Instrument: Cryogenic limb and nadir emission imaging FTIR spectrometer[1,2]. GLORIA is designed to operate on various high altitude research platforms (aircraft and stratospheric balloons).

Spectral coverage: 750-1450 cm^{-1}, unapodized spectral resolution: 0.0625 cm^{-1} (OPD_{max} = 8.0 cm, ~ 12 s per interferogram).

Data: Vertical profiles (limb mode) of temperature and trace gases from the middle troposphere up to flight altitude (lowermost stratosphere); vertical resolution: < 2 km; cloud index threshold[3] = 2.

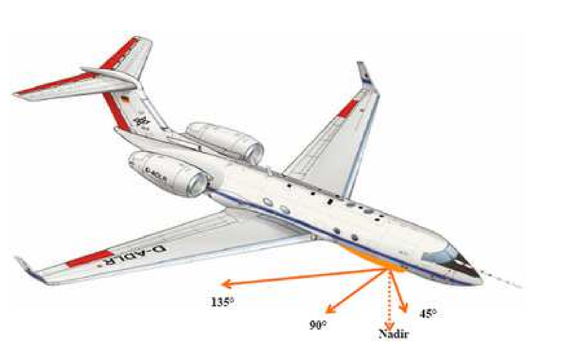


Fig. 1. The GLORIA instrument is located inside a belly pod aboard the DLR HALO aircraft.

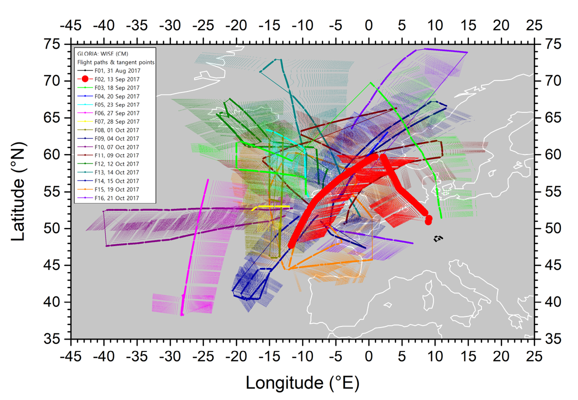


Fig. 2. Flight paths and GLORIA tangent altitudes during the WISE campaign in autumn 2017 (F02 = flight #2: red).

GLORIA data analysis

Radiative transfer: Calculated with KOPRA[4] (Karlsruhe Optimized and Precise Radiative transfer Algorithm).

Retrieval calculations: Least squares fitting procedure KOPRAFIT[5] using a Tikhonov-Phillips regularization approach which was constrained with respect to a first derivative a priori profile of the target species.

Fitted parameters: Temperature, gases, continuum or offset and scale, wavenumber shift.

Error estimation: Random noise as well as covariance effects of fitted parameters; temperature errors; pointing inaccuracies; errors of non-simultaneously fitted interfering gases; spectroscopic data errors (1σ).

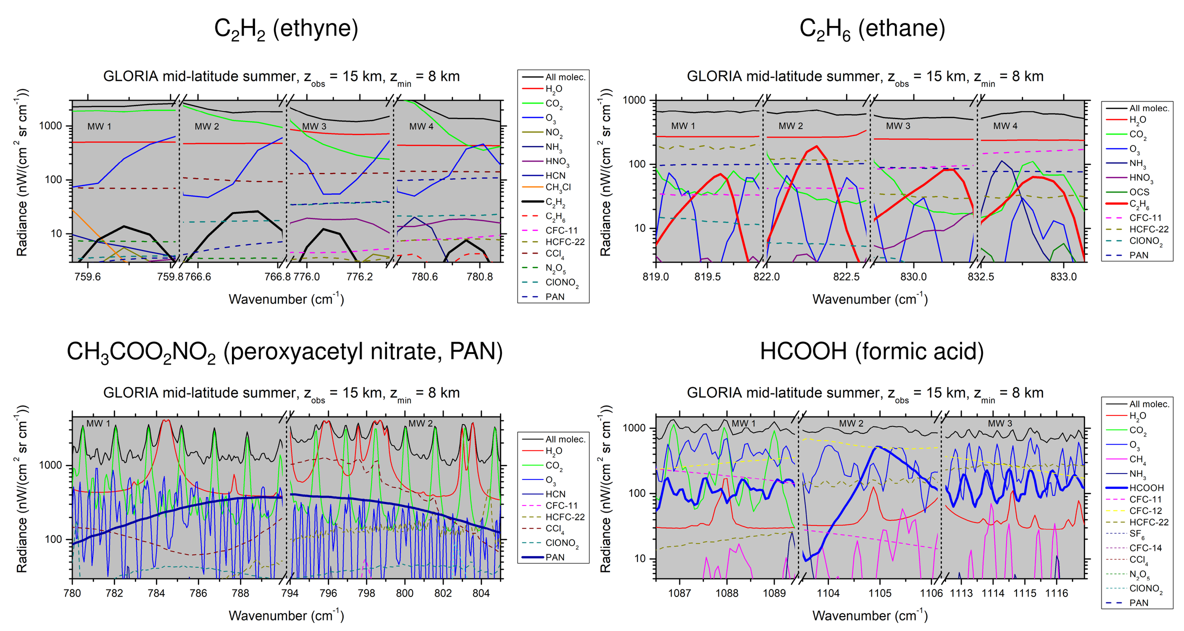


Fig. 3. Radiance calculations in different microwindows (with spectral resolution of GLORIA) for a mid-latitude standard atmosphere. Emissions of individual species contributing to the combined spectrum (all molecules) are shown.

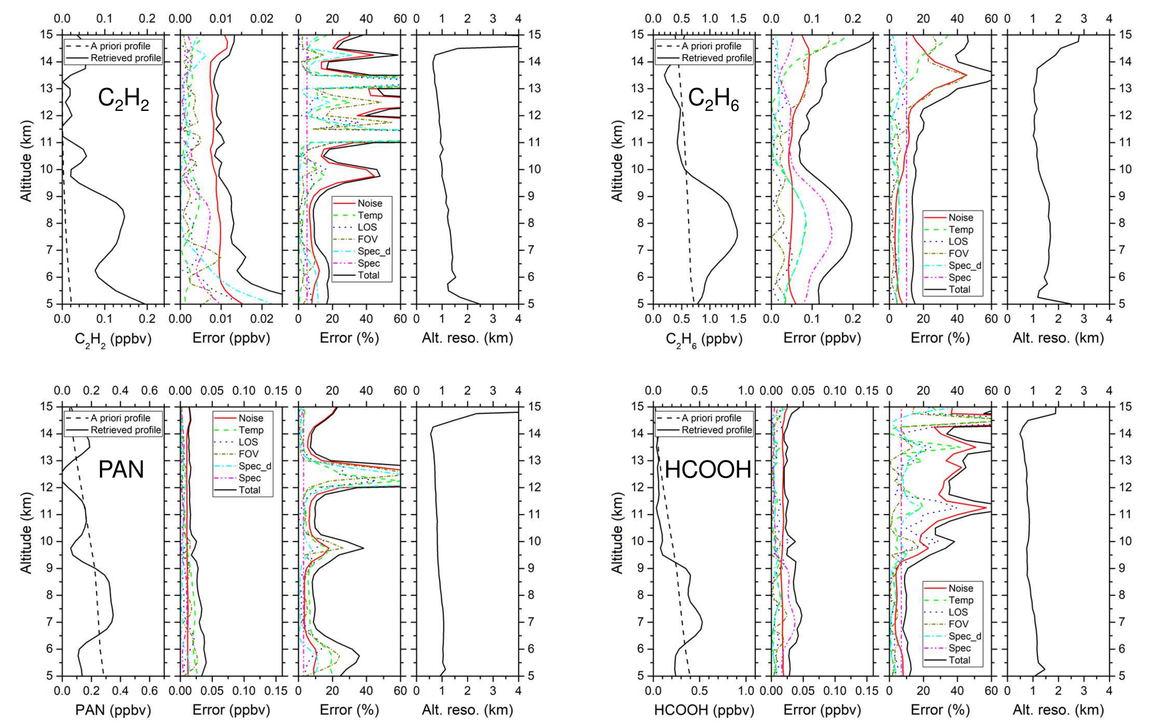


Fig. 4. Retrieved vertical VMR profiles (and a priori profiles) for flight #2 (2017-09-13) at 16:55:07 UTC together with absolute and relative errors and altitude resolution.

Sources and sinks of pollutant species

C_2H_2 (ethyne):
Source: combustion of biofuels, biomass- and fossil fuel burning[6]
Loss: reaction with OH[6]
Lifetime: ~ 2 weeks[6]

C_2H_6 (ethane):
Source: biomass burning, natural gas losses[7], fossil fuel production and biofuel use[8]
Loss: reaction with OH[8]
Lifetime: ~ 2 months[7]

$CH_3COO_2NO_2$ (peroxyacetyl nitrate, PAN):

$CH_3COO_2 + NO_2 + M \leftrightarrow CH_3COO_2NO_2 + M$ (R1)

Source: CH_3COO_2 (peroxyacetyl) is mainly produced by oxidation of acetaldehyde and photolysis of acetone and methylglyoxal (all directly emitted or produced from non-methane volatile organic compounds)[9]
Loss: Thermal decomposition via (R1)[9]
Lifetime: 1 h at 298 K, a few months in the cold upper troposphere[9,10,11]

HCOOH (formic acid):
Source: biogenic emissions, biomass burning and fossil fuel combustion; secondary photochemical production from anthropogenic and biogenic precursors[12]
Loss: wet and dry deposition and oxidation with OH[13]
Lifetime: 1-2 days (boundary layer), few weeks (free troposphere)[14]

Model simulations

Chemistry Climate Model EMAC

(ECHAM v5.3.02 / MESSy v2.53 [15,16] Atmospheric Chemistry)

- Resolution T106 (~1.1°x1.1°) with 90 hybrid p-levels up to 0.01 hPa.
- Multi-year simulation with a time step of 4 min. Nudging of model dynamics towards ERA-Interim[17] analysis (below 1 hPa).
- Chemistry from the troposphere to the lower mesosphere, kinetic and photochemical data from the IUPAC[18] and JPL-11[19] compilations.
- Photochemical reactions of precursor substances important for the build-up of PAN[9] were integrated.
- 2nd run: NMVOC emissions enhanced by a factor of two[20].

ECMWF CAMS reanalysis

(Copernicus Atmosphere Monitoring Service[21])

- 3-dimensional 3-h analyses and forecasts (T255, ~1.1°x1.1°, 60 levels) of global atmospheric composition (aerosols and chemical species).
- Available meteorological and atmospheric composition observations are included in the ECMWF 4D-Var data assimilation system[22].

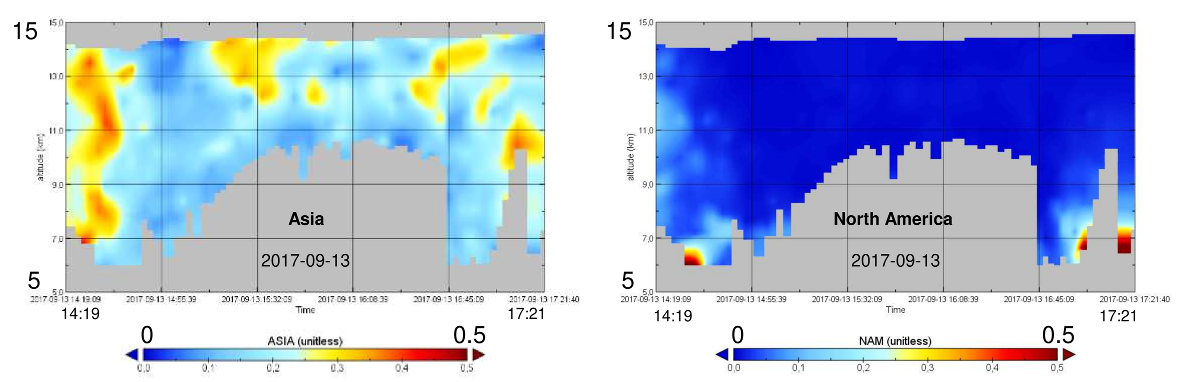
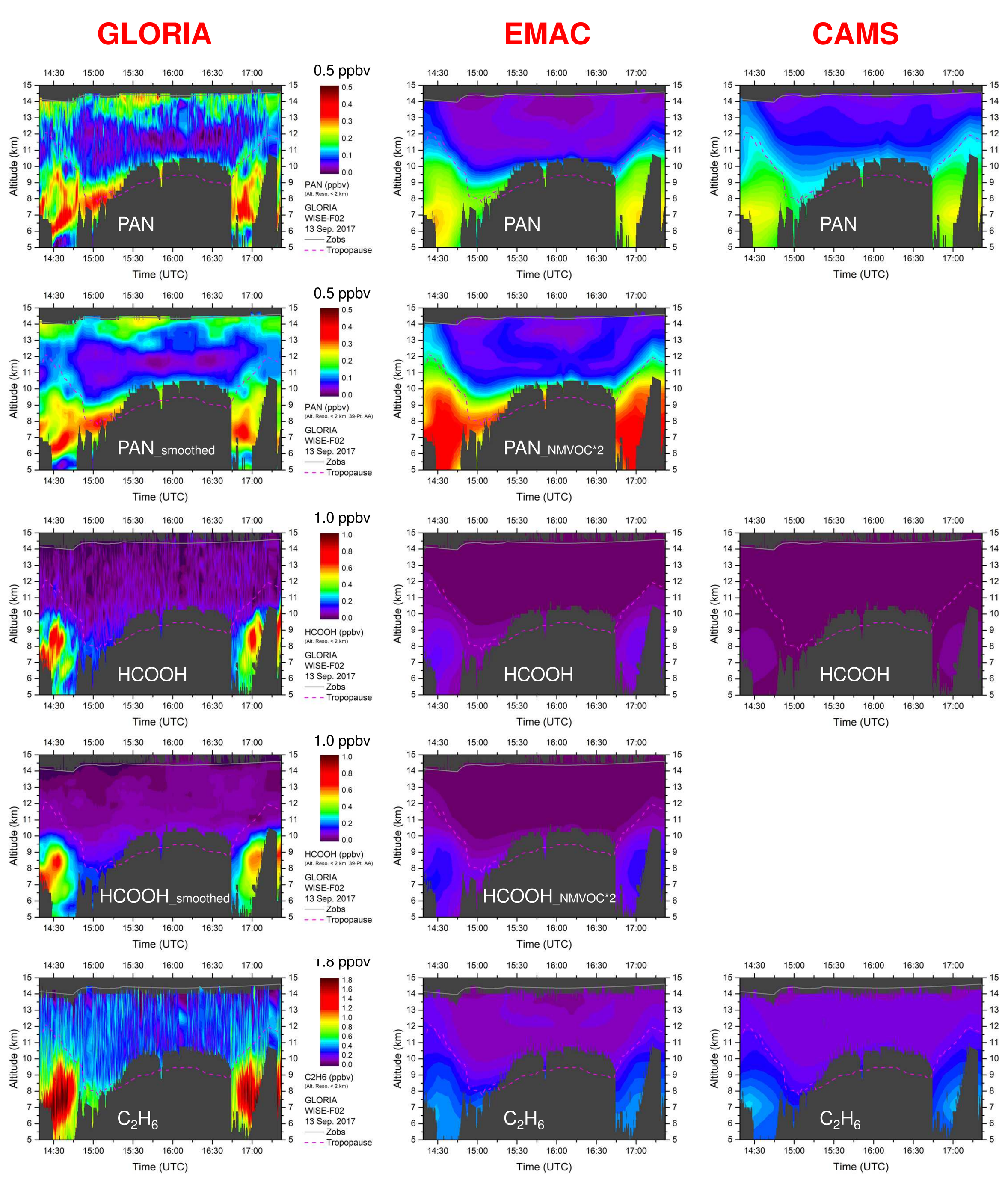


Fig. 5. Surface air mass origin tracer calculations in the Lagragian chemistry transport model CLaMS[23,24] showing the fraction of air originating from different geographical regions on 2017-09-13.

Observations and simulations



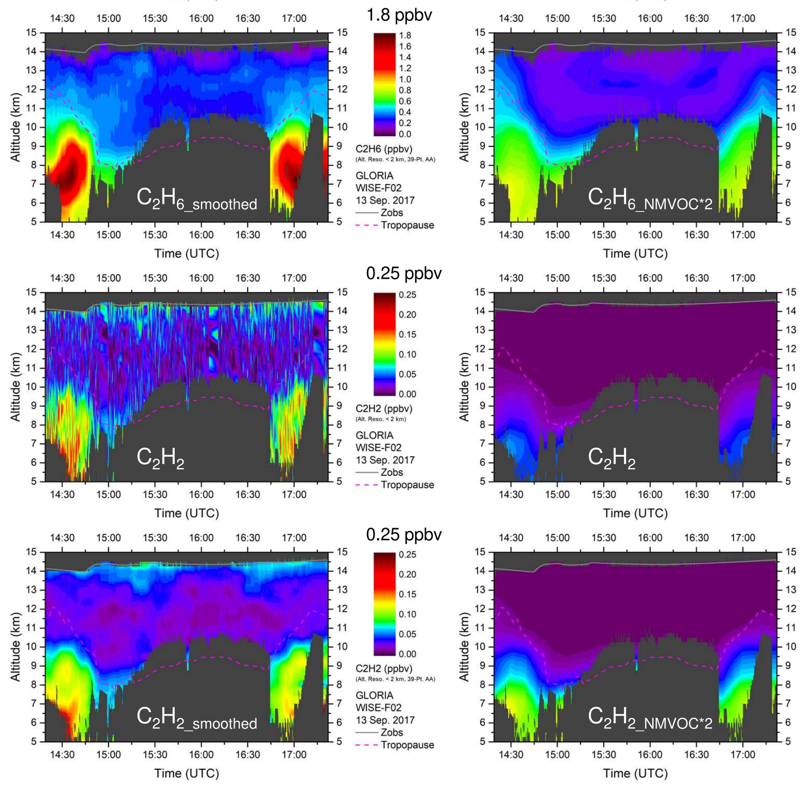


Fig. 6. GLORIA observations (directly retrieved and temporally smoothed for model comparison) of PAN, HCOOH, C_2H_6, and C_2H_2, for WISE flight #2 (left column) compared to EMAC simulations with and without raised NMVOC emissions (middle column) and CAMS assimilations (right column). Features of enhanced VMR are visible in the measurements. The origin of this pollution is mainly Asia and North America (see Fig. 5). EMAC and CAMS are also showing enhanced values in the region GLORIA does. Apart from PAN (EMAC), simulations by CAMS and, to a lesser extent, EMAC, underestimate the elevated amount of observed VMR features.

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