ATom: Actinic Flux and Photolysis Frequencies from CAFS Instrument, 2016-2018, V2

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Documentation Revision Date: 2021-11-17

Dataset Version: 2

Summary

This dataset contains actinic flux and photolysis frequencies for photodissociation reactions for a variety of chemical species during the four ATom campaigns. Spectrally resolved actinic flux was measured by the down- and up-welling Charged-coupled device Actinic Flux Spectroradiometers (CAFS) from approximately 280-650 nm. Photolysis frequencies were calculated from the actinic flux and published cross sections and quantum yield values for atmospherically relevant molecules. Solar radiation drives the chemistry of the atmosphere, including the evolution of ozone, greenhouse gases, biomass burning, and other anthropogenic and natural trace constituents.

This is Version 2 of this dataset. This version contains the most current iteration of CAFS-JV (photolysis frequency) data files and is the initial release of CAFS-FLUX-N and CAFS-FLUX-Z (actinic flux) files. Additional details can be found in Section 8. Dataset Revisions.

There are 134 data files in ICARTT (*.ict) format included in this dataset.

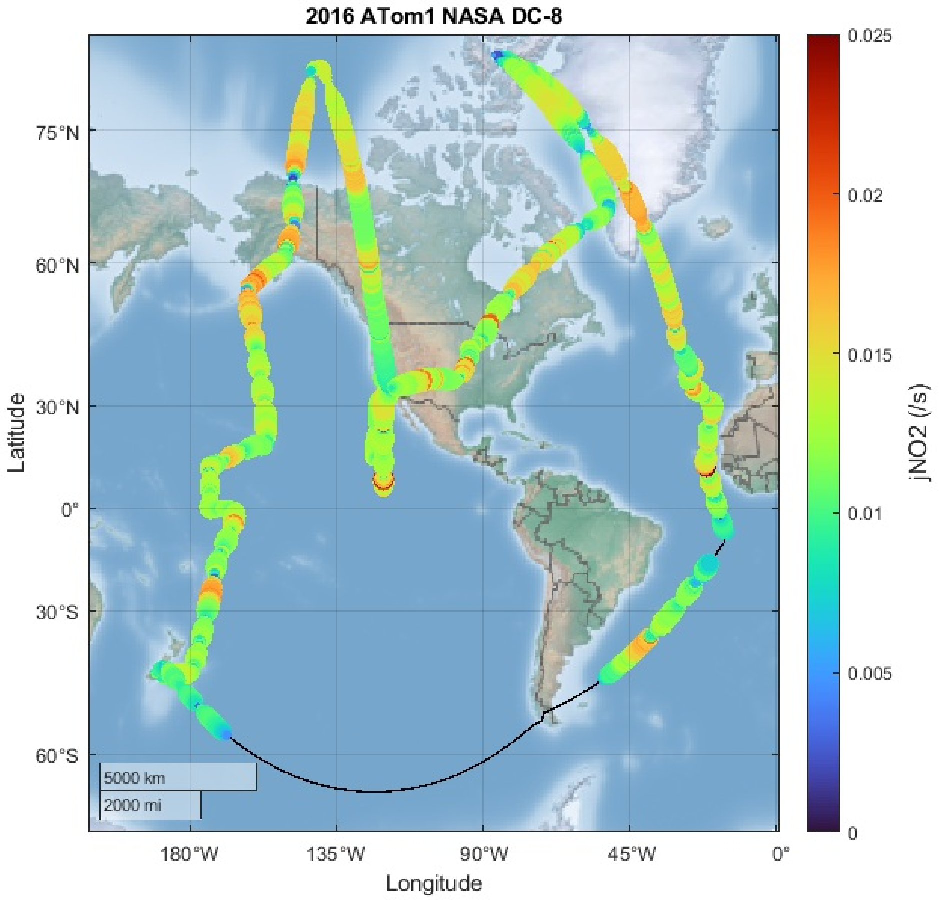

Figure 1. Charged-coupled device Actinic Flux Spectroradiometer (CAFS) derived total NO2 photolysis frequencies (jNO2) during ATom 1. The flight track (black line) is visible during nighttime operations when jNO2 is below detection limits.

Citation

Hall, S.R., and K. Ullmann. 2021. ATom: Actinic Flux and Photolysis Frequencies from CAFS Instrument, 2016-2018, V2. ORNL DAAC, Oak Ridge, Tennessee, USA. https://doi.org/10.3334/ORNLDAAC/1933

Table of Contents

1. Dataset Overview

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Project: Atmospheric Tomography Mission

The Atmospheric Tomography Mission (ATom) was a NASA Earth Venture Suborbital-2 mission. It studied the impact of human-produced air pollution on greenhouse gases and on chemically reactive gases in the atmosphere. ATom deployed an extensive gas and aerosol payload on the NASA DC-8 aircraft for systematic, global-scale sampling of the atmosphere, profiling continuously from 0.2 to 12 km altitude. Flights occurred in each of four seasons over a 4-year period.

Related Publication

Hall, S. R., K. Ullmann, M. J. Prather, C. M. Flynn, L. T. Murray, A. M. Fiore, G. Correa, S. A. Strode, S. D. Steenrod, J.-F. Lamarque, J. Guth, B. Josse, J. Flemming, V. Huijnen, N. L. Abraham, and A. T. Archibald. 2018, November 28. Cloud impacts on photochemistry: building a climatology of photolysis rates from the Atmospheric Tomography mission. Atmospheric Chemistry and Physics 18:16809–16828, https://doi.org/10.5194/acp-18-16809-2018

Related Datasets

Hall, S.R., and K. Ullmann. 2019. ATom: L2 Photolysis Frequencies from NCAR CCD Actinic Flux Spectroradiometers (CAFS). ORNL DAAC, Oak Ridge, Tennessee, USA. https://doi.org/10.3334/ORNLDAAC/1714.

- Version 1 of this dataset. Now superseded and available only upon request.

Hall, S.R., K. Ullmann, M.J. Prather, C.M. Flynn, L.T. Murray, A.M. Fiore, G. Correa, S.A. Strode, S.D. Steenrod, J.-F. Lamarque, J. Guth, B. Josse, J. Flemming, V. Huijnen, N.L. Abraham, and A.T. Archibald. 2019. ATom: Global Modeled and CAFS Measured Cloudy and Clear Sky Photolysis Rates, 2016. ORNL DAAC, Oak Ridge, Tennessee, USA. https://doi.org/10.3334/ORNLDAAC/1651

- Related photolysis rate data.

Wofsy, S.C., S. Afshar, H.M. Allen, E.C. Apel, E.C. Asher, B. Barletta, J. Bent, H. Bian, B.C. Biggs, D.R. Blake, N. Blake, I. Bourgeois, C.A. Brock, W.H. Brune, J.W. Budney, T.P. Bui, A. Butler, P. Campuzano-Jost, C.S. Chang, M. Chin, R. Commane, G. Correa, J.D. Crounse, P. D. Cullis, B.C. Daube, D.A. Day, J.M. Dean-Day, J.E. Dibb, J.P. DiGangi, G.S. Diskin, M. Dollner, J.W. Elkins, F. Erdesz, A.M. Fiore, C.M. Flynn, K.D. Froyd, D.W. Gesler, S.R. Hall, T.F. Hanisco, R.A. Hannun, A.J. Hills, E.J. Hintsa, A. Hoffman, R.S. Hornbrook, L.G. Huey, S. Hughes, J.L. Jimenez, B.J. Johnson, J.M. Katich, R.F. Keeling, M.J. Kim, A. Kupc, L.R. Lait, K. McKain, R.J. Mclaughlin, S. Meinardi, D.O. Miller, S.A. Montzka, F.L. Moore, E.J. Morgan, D.M. Murphy, L.T. Murray, B.A. Nault, J.A. Neuman, P.A. Newman, J.M. Nicely, X. Pan, W. Paplawsky, J. Peischl, M.J. Prather, D.J. Price, E.A. Ray, J.M. Reeves, M. Richardson, A.W. Rollins, K.H. Rosenlof, T.B. Ryerson, E. Scheuer, G.P. Schill, J.C. Schroder, J.P. Schwarz, J.M. St.Clair, S.D. Steenrod, B.B. Stephens, S.A. Strode, C. Sweeney, D. Tanner, A.P. Teng, A.B. Thames, C.R. Thompson, K. Ullmann, P.R. Veres, N.L. Wagner, A. Watt, R. Weber, B.B. Weinzierl, P.O. Wennberg, C.J. Williamson, J.C. Wilson, G.M. Wolfe, C.T. Woods, L.H. Zeng, and N. Vieznor. 2021. ATom: Merged Atmospheric Chemistry, Trace Gases, and Aerosols, Version 2. ORNL DAAC, Oak Ridge, Tennessee, USA. https://doi.org/10.3334/ORNLDAAC/1925.

- Data from all ATom instruments and all four flight campaigns, including aircraft location and navigation data, merged to several different time bases.

Wofsy, S.C., and ATom Science Team. 2018. ATom: Aircraft Flight Track and Navigational Data. ORNL DAAC, Oak Ridge, Tennessee, USA. https://doi.org/10.3334/ORNLDAAC/1613

- Flight path (location and altitude) data for each of the four campaigns provided in KML and CSV formats.

2. Data Characteristics

Spatial Coverage: Global. Flights circumnavigate the globe, primarily over the oceans

Spatial Resolution: Point measurements

Temporal Coverage: Periodic flights occurred during each campaign

Deployment	Date Range
ATom-1	July 29 - August 23, 2016
ATom-2	January 26 - February 21, 2017
ATom-3	September 28 - October 28, 2017
ATom-4	April 24 - May 21, 2018

Temporal Resolution: 3 seconds

Data File Information

There are 134 data files in ICARTT (*.ict) format included in this dataset. The ICARTT files conform to the ICARTT File Format Standards V1.1. The files are named *CAFS-**X**_DC8_**YYYYMMDD_R#***.ict, where ***X*** represents the measured variable, **YYYYMMDD** is the start date (in UTC time) of the flight, and **R#** is the file version or revision number.

Table 1. File names and descriptions.

File Name	Number of Files	Description
CAFS-FLUX-N_DC8_YYYYMMDD_R#.ict	44	Contain the nadir instrument spectral actinic flux densities (upwelling radiation during level flight)
CAFS-FLUX-Z_DC8_YYYYMMDD_R#.ict	45	Contain the zenith instrument spectral actinic flux densities (downwelling radiation during level flight)
CAFS-JV_DC8_YYYYMMDD_R#.ict	45	Contain photolysis frequencies that were calculated from the total actinic flux (zenith plus nadir) and are generally insensitive to aircraft orientation

Data File Details

Table 2. Variable names and descriptions for files named CAFS-JV_DC8_YYYYMMDD_R#.ict.

Name	Units	Description
UTC_Start	seconds	Start time in seconds since 0000 UTC
UTC_Stop	seconds	End time in seconds since 0000 UTC
UTC_Mid	seconds	Midpoint of in seconds since 0000 UTC
j[O3->O2+O(1D)]	rate per second	Total in situ rate coefficient for photolysis of species
j[NO2->NO+O(3P)]	rate per second	Total in situ rate coefficient for photolysis of species
j[H2O2->2OH]	rate per second	Total in situ rate coefficient for photolysis of species
j[NO3->NO+O2]	rate per second	Total in situ rate coefficient for photolysis of species
j[NO3->NO2+O(3P)]	rate per second	Total in situ rate coefficient for photolysis of species
j[N2O5->NO3+NO2]	rate per second	Total in situ rate coefficient for photolysis of species
j[HNO2->OH+NO]	rate per second	Total in situ rate coefficient for photolysis of species
j[HNO3->OH+NO2]	rate per second	Total in situ rate coefficient for photolysis of species
j[HNO4->HO2+NO2(UV_VISonly)]	rate per second	Total in situ rate coefficient for photolysis of species (UV_VISonly)
j[CH2O->H+HCO]	rate per second	Total in situ rate coefficient for photolysis of species
j[CH2O->H2+CO]	rate per second	Total in situ rate coefficient for photolysis of species
j[CH3CHO->CH3+HCO]	rate per second	Total in situ rate coefficient for photolysis of species
j[C2H5CHO->C2H5+HCO]	rate per second	Total in situ rate coefficient for photolysis of species
j[CH3OOH->CH3O+OH]	rate per second	Total in situ rate coefficient for photolysis of species
j[CH3ONO2->CH3O+NO2]	rate per second	Total in situ rate coefficient for photolysis of species
j[CH3CH2ONO2->CH3CH2O+NO2]	rate per second	Total in situ rate coefficient for photolysis of species
j[CH3CO(OONO2)->CH3CO(OO)+NO2]	rate per second	Total in situ rate coefficient for photolysis of species
j[CH3CO(OONO2)->CH3CO(O)+NO3]	rate per second	Total in situ rate coefficient for photolysis of species
j[CH2=C(CH3)CHO->Products]	rate per second	Total in situ rate coefficient for photolysis of species
j[CH3COCH=CH2->Products]	rate per second	Total in situ rate coefficient for photolysis of species
j[CH3COCH3->CH3CO+CH3]	rate per second	Total in situ rate coefficient for photolysis of species
j[CH3COCH2CH3->CH3CO+CH2CH3]	rate per second	Total in situ rate coefficient for photolysis of species
j[CH2(OH)COCH3->CH3CO+CH2(OH)]	rate per second	Total in situ rate coefficient for photolysis of species
j[CH2(OH)COCH3->CH2(OH)CO+CH3]	rate per second	Total in situ rate coefficient for photolysis of species
j[CHOCHO->HCO+HCO]	rate per second	Total in situ rate coefficient for photolysis of species
j[CHOCHO->H2+2CO]	rate per second	Total in situ rate coefficient for photolysis of species
j[CHOCHO->CH2O+CO]	rate per second	Total in situ rate coefficient for photolysis of species
j[CH3COCHO->CH3CO+HCO]	rate per second	Total in situ rate coefficient for photolysis of species
j[CH3COCOCH3->Products]	rate per second	Total in situ rate coefficient for photolysis of species
j[Cl2->Cl+Cl]	rate per second	Total in situ rate coefficient for photolysis of species
j[ClO->Cl+O(3P)]	rate per second	Total in situ rate coefficient for photolysis of species
j[ClNO2->Cl+NO2]	rate per second	Total in situ rate coefficient for photolysis of species
j[ClONO->Cl+NO2]	rate per second	Total in situ rate coefficient for photolysis of species
j[ClONO2->Cl+NO3]	rate per second	Total in situ rate coefficient for photolysis of species

j[ClONO2->ClO+NO2]	rate per second	Total in situ rate coefficient for photolysis of species
j[Br2->Br+Br]	rate per second	Total in situ rate coefficient for photolysis of species
j[BrO->Br+O]	rate per second	Total in situ rate coefficient for photolysis of species
j[HOBr->OH+Br]	rate per second	Total in situ rate coefficient for photolysis of species
j[BrNO->Br+NO]	rate per second	Total in situ rate coefficient for photolysis of species
j[BrONO->Br+NO2]	rate per second	Total in situ rate coefficient for photolysis of species
j[BrONO->BrO+NO]	rate per second	Total in situ rate coefficient for photolysis of species
j[BrNO2->Br+NO2]	rate per second	Total in situ rate coefficient for photolysis of species
j[BrONO2->BrO+NO2]	rate per second	Total in situ rate coefficient for photolysis of species
j[BrONO2->Br+NO3]	rate per second	Total in situ rate coefficient for photolysis of species
j[BrCl->Br+Cl]	rate per second	Total in situ rate coefficient for photolysis of species
j[CHBr3->Products]	ratio	Total in situ rate coefficient for photolysis of species
jO3_dnwFrac	ratio	jO3_downwelling/(jO3_downwelling+jO3_upwelling)
jNO2_dnwFrac	ratio	jNO2_downwelling/(jNO2_downwelling+jNO2_upwelling)

Table 3. Variable descriptions for files named CAFS-FLUX-N_DC8_YYYYMMDD_R#.ict and CAFS-FLUX-Z_DC8_YYYYMMDD_R#.ict.

Name	Units	Description
UTC_Start	Seconds	Start time in seconds since 0000 UTC
UTC_Stop	Seconds	End time in seconds since 0000 UTC
UTC_Mid	Seconds	Midpoint of in seconds since 0000 UTC
XXX.XXXX	photons/s/cm^2/nm/10^{10}	Spectrally resolved actinic flux density where XXX.XXXX (e.g. 296.6078) is the center wavelength in nm where the observation was taken, n=433. Note: 10^{10} scaling factor applied.

3. Application and Derivation

ATom builds the scientific foundation for mitigation of short-lived climate forcers, in particular, methane (CH$_4$), tropospheric ozone (O$_3$), and Black Carbon aerosols (BC).

ATom Science Questions

Tier 1

- What are chemical processes that control the short-lived climate forcing agents CH$_4$, O$_3$, and BC in the atmosphere? How is the chemical reactivity of the atmosphere on a global scale affected by anthropogenic emissions? How can we improve chemistry-climate modeling of these processes?

Tier 2

- Over large, remote regions, what are the distributions of BC and other aerosols important as short-lived climate forcers? What are the sources of new particles? How rapidly do aerosols grow to CCN-active sizes? How well are these processes represented in models?
- What type of variability and spatial gradients occurs over remote ocean regions for greenhouse gases (GHGs) and ozone-depleting substances (ODSs)? How do the variations among air parcels help identify anthropogenic influences on photochemical reactivity, validate satellite data for these gases, and refine knowledge of sources and sinks?

Significance

ATom delivers unique data and analysis to address the Science Mission Directorate objectives of acquiring “datasets that identify and characterize important phenomena in the changing Earth system” and “measurements that address weaknesses in current Earth system models leading to improvement in modeling capabilities.” ATom will provide unprecedented challenges to the CCMs used as policy tools for climate change assessments, with comprehensive data on atmospheric chemical reactivity at global scales, and will work closely with modeling teams to translate ATom data to better, more reliable CCMs. ATom provides extraordinary validation data for remote sensing.

4. Quality Assessment

Actinic flux is calibrated using NIST source lamps with certified uncertainties of 4% in the UV-B and 3% in the UV-A spectral regions. Additional uncertainties due to calibration geometry, radiometric power supplies, temperature stability, and wavelength assignment lead to total uncertainties of +/- 6% and +/- 5% at high signal. Optical uncertainties typically add +/-3% with larger values at high solar zenith angles (>80 degrees). Spectrometer stray light uncertainty affects the shortest wavelengths (<310 nm) requiring comparisons to double monochromator detectors and modeled actinic flux to provide corrections.

Photolysis uncertainties are dominated by cross-section and quantum yield determinations from published literature sources and are typically +/- 5-10% for cross-sections and greater than +/- 10% for quantum yields. These uncertainties are not consistently quantified and can be exacerbated by variability in temperature and pressure. Combined with measurement and calibration uncertainties, examples of high-sun total photolysis uncertainties include +/-15% for ozone and +/-12% for nitrogen dioxide.

5. Data Acquisition, Materials, and Methods

Project Overview

ATom makes global-scale measurements of the chemistry of the atmosphere using the NASA DC-8 aircraft. Flights span the Pacific and Atlantic Oceans, nearly pole-to-pole, in continuous profiling mode, covering remote regions that receive long-range inputs of pollution from expanding industrial economies. The payload has proven instruments for in situ measurements of reactive and long-lived gases, diagnostic chemical tracers, and aerosol size, number, and composition, plus spectrally resolved solar radiation and meteorological parameters.

Combining distributions of aerosols and reactive gases with long-lived GHGs and ODSs enables disentangling of the processes that regulate atmospheric chemistry: emissions, transport, cloud processes, and chemical transformations. ATom analyzes measurements using customized modeling tools to derive daily averaged chemical rates for key atmospheric processes and to critically evaluate CCMs. ATom also differentiates between hypotheses for the formation and growth of aerosols over the remote oceans.

Charged-coupled device Actinic Flux Spectroradiometers

The Charged-coupled device Actinic Flux Spectroradiometers (CAFS) instruments measure in situ down- and up-welling radiation and combine to provide 4 pi steradian actinic flux density spectra from 280 to 650 nm. The sampling resolution is ~0.8 nm with a full width at half maximum (FWHM) of 1.7 nm at 297 nm. From the measured flux, photolysis frequencies are calculated for ~40 important atmospheric trace gases including O_3, NO_2, HCHO, HONO, and NO_3 using a modified version of the NCAR Tropospheric Ultraviolet and Visible (TUV) radiative transfer model.

The absolute spectral sensitivity of the instruments is determined in the laboratory with 1000 W NIST-traceable tungsten-halogen lamps with a wavelength-dependent uncertainty of 3–5%. During deployments, spectral sensitivity is assessed with secondary calibration lamps while wavelength assignment is tracked with Hg line sources and comparisons to spectral features in the extraterrestrial flux. The optical collectors are characterized for angular and azimuthal response and the effective planar receptor distance.

CAFS has an excellent legacy of performance on the NASA DC-8 and WB-57 platforms during atmospheric chemistry and satellite validation mission. These include AVE Houston 2004 and 2005, PAVE, CR-AVE, TC4, ARCTAS, DC3, SEAC4RS, KORUS-AQ, ATom, and FIREX-AQ. For FIREX-AQ, upgraded electronics and cooling reduced noise and allowed for a decrease to 1 Hz acquisition. Additional information can be found in Shetter and Mueller (1999) and on the ESPO CAFS Instrument webpage.

6. Data Access

These data are available through the Oak Ridge National Laboratory (ORNL) Distributed Active Archive Center (DAAC).

ATom: Actinic Flux and Photolysis Frequencies from CAFS Instrument, 2016-2018, V2

Contact for Data Center Access Information:

- E-mail: uso@daac.ornl.gov
- Telephone: +1 (865) 241-3952

7. References

Shetter, R.E., and M. Müller. 1999. Photolysis frequency measurements using actinic flux spectroradiometry during the PEM-Tropics mission: Instrumentation description and some results. Journal of Geophysical Research: Atmospheres 104:5647–5661. https://doi.org/10.1029/98JD01381

8. Dataset Revisions

Version	Release Date	Description
2.0	2021-11-17	Initial release of CAFS-FLUX-N and CAFS-FLUX-Z files. CAFS-JV files were updated to NASA standard chemical names, jN2O5 was updated, and jO3_dnwFrac and jNO2_dnwFrac were corrected.
1.0	2019-09-09	Initial release. Now superseded and available only upon request.

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Home

About Us
Mission
Data Use and Citation Policy
User Working Group
Partners

Get Data
Science Themes
NASA Projects
All Datasets

Submit Data
Submit Data Form
Data Scope and Acceptance
Data Authorship Policy
Data Publication Timeline
Detailed Submission Guidelines

Tools
MODIS
THREDDS
SDAT
Daymet
Airborne Data Visualizer
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Resources
Learning
Data Management
News
Earthdata Forum

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