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ATom: Age of Air, ArN2 Ratio, and Trace Gases in Stratospheric Samples, 2009-2018

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Summary

This dataset provides calculated age of air (AoA) and the argon/nitrogen (Ar/N₂) ratio (per meg) from stratospheric flask samples and simultaneous high-frequency measurements of nitrous oxide (N₂O), carbon dioxide (CO₂), ozone (O₃), methane (CH₄), and carbon monoxide (CO) compiled from three airborne projects. The trace gases were used to identify 235 flask samples with stratospheric influence collected by the Medusa Whole Air Sampler and to calculate AoA using a new N₂O-AoA relationship developed using a Markov Chain Monte Carlo algorithm. The data span a wide range of latitudes poleward of 40 degrees in both the Northern and Southern Hemispheres and cover the period 2009-01-10 to 2018-05-21.

The N₂O, CH₄, CO, CO₂ and continuous H₂O measurements were compiled from the following project campaigns; HIAPER Pole-to-Pole Observations (HIPPO, 2008– 2011), Southern Ocean (ORCAS, 2016) and Atmospheric Tomography (ATom, 2016–2018). O₃ measurements were from HIPPO and ATom. Medusa air samples were used from all five HIPPO campaigns, ORCAS, and ATom campaigns 2–4. Samples were labeled as “stratospheric” if water vapor levels were below 50 ppm and either O₃ values exceeded 140 ppb or N₂O (detrended to a reference year of 2009) was below 315 ppb. These criteria identified lower stratospheric samples with high quality N₂O and δ(Ar/N₂) data.

There is one data file in comma-separated format (.csv) with this dataset.

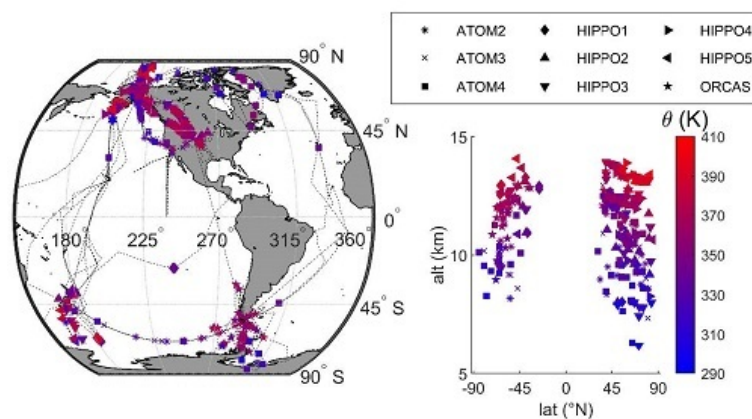


Figure 1. Horizontal (left) and vertical (right) distribution of airborne flask sample locations identified as being of stratospheric origin. Thin dashed black lines on the map illustrate the flight tracks of all 9 campaigns of the 3 airborne projects. Symbols indicate the campaigns during which the stratospheric samples were collected, and colors show the potential temperature at which the sample was taken. Source: Birner et al., 2020

Citation

Birner, B., M. Chipperfield, E.J. Morgan, and R.F. Keeling. 2020. ATom: Age of Air, ArN₂ Ratio, and Trace Gases in Stratospheric Samples, 2009-2018. ORNL DAAC, Oak Ridge, Tennessee, USA. <https://doi.org/10.3334/ORNLDAAC/1788>

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1. Dataset Overview

This dataset provides calculated age of air (AoA) and the argon/nitrogen (Ar/N₂) ratio (per meg) from stratospheric flask samples and

simultaneous high frequency measurements of nitrous oxide (N₂O), carbon dioxide (CO₂), ozone (O₃), methane (CH₄), and carbon monoxide (CO) compiled from three airborne projects. The trace gases were used to identify 235 flask samples with stratospheric influence collected by the Medusa Whole Air Sampler and to calculate AoA using a new N₂O-AoA relationship developed using a Markov Chain Monte Carlo algorithm. The data span a wide range of latitudes poleward of 40 degrees in both the Northern and Southern Hemispheres and cover the period 2009-01-10 to 2018-05-21.

The N₂O, CH₄, CO, CO₂ and continuous H₂O measurements were compiled from the following project campaigns; HIAPER Pole-to-Pole Observations (HIPPO, 2008– 2011), Southern Ocean (ORCAS, 2016) and Atmospheric Tomography (ATom, 2016–2018). O₃ measurements were from HIPPO and ATom. Medusa air samples were used from all five HIPPO campaigns, ORCAS, and ATom campaigns 2–4. Samples were labeled as “stratospheric” if water vapor levels were below 50 ppm and either O₃ values exceeded 140 ppb or N₂O (detrended to a reference year of 2009) was below 315 ppb. These criteria identified lower stratospheric samples with high quality N₂O and δ(Ar/N₂) data.

Project: [Atmospheric Tomography Mission \(ATom\)](#)

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 4 seasons over a 4-year period.

Related Publication:

Birner, B., Chipperfield, M. P., Morgan, E. J., Stephens, B. B., Linz, M., Feng, W., Wilson, C., Bent, J. D., Wofsy, S. C., Severinghaus, J., and Keeling, R. F. 2020. Gravitational separation of Ar/N₂ and age of air in the lowermost stratosphere in airborne observations and a chemical transport model, *Atmos. Chem. Phys. Discuss.* <https://doi.org/10.5194/acp-2020-95>

Related Datasets:

ATom: L2 Measurements from Medusa Whole Air Sampler (Medusa). ORNL DAAC, Oak Ridge, Tennessee, USA. <https://doi.org/10.3334/ORNLDAAC/1729>

ATom: Merged Atmospheric Chemistry, Trace Gases, and Aerosols. Data from all ATom instruments and all four flight campaigns, including aircraft location and navigation data, merged to several different time bases: <https://doi.org/10.3334/ORNLDAAC/1581>

ATom Flight Track and Navigational Data. Flight path (location and altitude) data for each of the four campaigns provided in KML and csv format: <https://doi.org/10.3334/ORNLDAAC/1613>

HIAPER Pole-to-Pole Observations (HIPPO) of Carbon Cycle and Greenhouse Gases Study. Data available at: https://www.eol.ucar.edu/field_projects/hippo

The O₂/N₂ Ratio and CO₂ Airborne Southern Ocean (ORCAS) project data available at: https://www.eol.ucar.edu/field_projects/orcas

Acknowledgement:

This study was funded under the ATom project, grant number NNX15AJ23G.

2. Data Characteristics

Spatial Coverage: Global

Spatial Resolution: Multiple points

Temporal Coverage: 2009-01-10 to 2018-05-21

Temporal Resolution: One-time estimates compiled from multiple campaigns (HIPPO, 2009-2011; ORCAS, 2016; ATom, 2016-2018)

Study Area: All latitude and longitude are given in decimal degrees.

Site	Northernmost Latitude	Southernmost Latitude	Easternmost Longitude	Westernmost Longitude
Global	84.53	-84.51	178.68	-179.58

Data file information

There is one data file with this dataset in comma-separated (.csv) format.

Table 1. Variables names and descriptions for **ArN2_AirAge_Trace_Gases.csv**.

Variable	Units	Description
date	YYYY-MM-DD	Sample date (UTC)
time	HH:MM:SS	Sample time in HH:MM:SS (UTC)
decimal_year	YYYY.YYY	Date as decimal year (UTC)
latitude	Decimal degrees	Latitude in decimal degrees
longitude	Decimal degrees	Longitude in decimal degrees
pressure	hPa	Ambient pressure
altitude	m	Altitude
theta	K	Potential temperature
AoA	year	Age of Air: The time elapsed since entry of an air parcel into the stratosphere
AoA_lowerBound	year	Age of Air, lower bound: 95% confidence interval
AoA_upperBound	year	Age of Air, upper bound: 95% confidence interval

CO2	ppm	Carbon Dioxide measurements compiled from the HIPPO, ORCAS and ATom project campaigns
ArN2	per meg	Argon to Nitrogen ratio (Ar/N2)
N2O	ppb	Nitrous Oxide measurements compiled from the HIPPO, ORCAS and ATom project campaigns
CO	ppb	Carbon Monoxide measurements compiled from the HIPPO, ORCAS and ATom project campaigns
O3	ppb	Ozone measurements from HIPPO and ATom
H2O	>ppm	Water measurements compiled from the HIPPO, ORCAS and ATom project campaigns
CH4	ppb	Methane measurements compiled from the HIPPO, ORCAS and ATom project campaigns

3. Application and Derivation

Transport in the middle atmosphere is driven by a combination of advection by the Brewer-Dobson circulation (BDC) and quasi-horizontal, two-way mixing by breaking waves. Models consistently predict an acceleration of the BDC due to climate change (Butchart, 2014) but subgrid-scale mixing processes and momentum transfer by unresolved buoyancy waves limit the ability to accurately simulate circulation in the stratosphere. An acceleration of the BDC has important repercussions for stratosphere-troposphere exchange (STE), and thus recovery of the ozone layer and the greenhouse effect of stratospheric water vapor.

Stratospheric air is detectably fractionated by gravitational settling (GS), with the degree of fractionation strongly correlated to AoA. 2-D and 3-D models both show a pattern of GS which increases with altitude and latitude, similar to the patterns observed in tracers with a significant stratospheric sink such as N2O, and consistent with a positive correlation with AoA. This study investigated the calibration of GS of Ar/N2 as an AoA tracer. The data were used to calculate the mean age of air, a widely used indicator of stratospheric circulation (Birner et al., 2020).

4. Quality Assessment

Quality assessment procedures related to the measurement and analysis of the trace gases differ by instruments used in the projects.

5. Data Acquisition, Materials, and Methods

Following is a brief synopsis of the sampling, analyses, and data compilation and processing as described in Birner et al., 2020.

Discrete 1.5L flask samples were taken with the Medusa Whole Air Sampler and shipped to Scripps Institution of Oceanography (SIO) for analysis of Ar/N2 ratios on an IsoPrime Mass Spectrometer. Changes in Ar/N2 ratios are in delta notation

$$\delta = \left(\frac{R_{SA}}{R_{REF}} - 1 \right) \times 10^6 (\text{per meg})$$

where R_{SA} is the mixing or isotope ratio in the sample and R_{REF} the ratio in a reference mixture.

Available simultaneous, high-frequency, measurements were compiled for a range of other trace gases including N2O, CO2, O3, CH4, and CO. These were used to identify Medusa samples with stratospheric influence and to calculate AoA. The N2O, CH4, CO, CO2, and continuous H2O measurements were from the HIPPO, ORCAS, and ATom project campaigns. O3 measurements were from HIPPO and ATom. Air samples were used from all five HIPPO campaigns, ORCAS, and ATom campaigns 2–4. Samples from ATom 1 were not used because of apparent biases. An averaging kernel was applied to the continuous and semi-continuous aircraft data, such as N2O, O3, and H2O, to match it to Medusa samples.

Composition, rather than potential temperature or altitude, effectively selects samples with a clear stratospheric signature in the lowermost stratosphere where mixing with the upper troposphere could be substantial and minimizes the impact of synoptic scale variability. Samples were labeled as “stratospheric” if (i) water vapor levels were below 50 ppm and either (ii.a) O3 values exceeded 140 ppb or (ii.b) N2O (detrended to a reference year of 2009) was below 315 ppb. These criteria yielded 235 lower stratospheric samples with high-quality N2O and $\delta(\text{Ar/N2})$ data, spanning a wide range of latitudes poleward of 40° in both the Northern and Southern Hemisphere (Figure 1) (Birner et al., 2020).

AoA calculation

Stratospheric AoA was calculated from N2O using an updated hemisphere-specific N2O-AoA relationship. The method broadly follows Andrews et al. (2001) who assumed a bimodal age spectrum and use multiple observations of CO2 binned by N2O values to resolve the seasonal cycle of CO2. Properties of the age spectrum for each N2O bin, including AoA were constrained by optimizing the agreement between observed CO2 concentrations and concentration implied by randomly generated age spectra in each N2O bin. A Markov chain Monte Carlo (MCMC) method was used to obtain robust uncertainties for all estimated parameters of the age spectrum. The algorithm builds on a Metropolis-Hasting sampler (Metropolis et al., 1953; Hastings, 1970) as implemented by Malinverno (2002) to evaluate probability distributions for each age spectrum parameter and automatically chooses whether a unimodal or bimodal representation of the age spectrum is more appropriate in each N2O bin (Birner et al., 2020). The new AoA-N2O relationships for each hemisphere are given by NH AoA = $-0.0002361 (323.23 - \text{N2O})^2 + 0.05530 (323.23 - \text{N2O})$, and SH AoA = $-0.0001754 (323.23 - \text{N2O})^2 + 0.05359 (323.23 - \text{N2O})$ (Birner et al., 2020).

6. Data Access

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

[ATom: Age of Air, ArN2 Ratio, and Trace Gases in Stratospheric Samples, 2009-2018](#)

Contact for Data Center Access Information:

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7. References

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