

1/ Observational surface mixing ratio of ethane

We constructed a time series of annually averaged ethane mixing ratios using historical data from the Oregon Graduate Institute of Science & Technology (OGI), University of California – Irvine (UCI), and the National Oceanic and Atmospheric Administration Earth System Research Laboratory Global Monitoring Division (NOAA).

The OGI ethane mixing ratio data were measured at 6 sites ([Fig. xx \(show map\)](#)) distributed from the Northern Hemisphere to the Southern Hemisphere. Each site is sampled several times every month; although, full-year records are only available from 1985-1986 for sites in the Northern Hemisphere and 1983-1986 for sites in the Southern Hemisphere. The samples were analyzed at the Oregon Graduate Institute laboratory using gas chromatography (Khalil et al. 1983).

The UCI mixing ratio data were collected biweekly in the Pacific Basin from remote surface locations and analyzed at the UCI laboratory using gas chromatography within one month after the collection. The observational measurements are only available in March, June, September, and December, which correspond to the maximum, minimum and the inflections of the ethane seasonal cycle. The mixing ratio record spans from 1985 to 2008, although only complete full-year records are available in the following years: 1985, 1990, 1994, 1996-2008 (Simpson et al. 2012)

The NOAA mixing ratio data have 39 sites spanning from 2006 to 2014. The NOAA sites are distributed across the globe on all 7 continents. The samples were collected several times weekly and analyzed at the Institute of Arctic and Alpine Research, Atmospheric Research Laboratory (ARL), Boulder, Colorado, USA using gas chromatography (Helmig et al. 2017).

The ethane mixing ratio has a large seasonal cycle with a maximum occurs in March and a minimum occurs in September ([show a fig of a notable site](#)). For consistency with the UCI sampling, we defined the months March, June, September, and December each as a season and only these months are examined from NOAA and OGI. The UCI data is distributed from latitude 50° S to 75° N, so we constrained the analyses to these latitudes.

2/ Modeling of ethane mixing ratios

3/ Long-term history of Interhemispheric Ratio

We hypothesize that the interhemispheric differences in ethane mixing ratios are sensitive to changes in ethane's source emissions history. Therefore, we use the Interhemispheric Ratio (IHR) to infer the trend of interhemispheric differences in the ethane mixing ratio from 1983 to 2014. We also expect that the IHR will eliminate the absolute calibration differences between datasets.

a. Method 1

Because sampling locations vary with time, we cannot construct a long-term history of ethane mixing ratios from single stations. Instead, in our first method, we divided the atmosphere into 5 latitudinal bands: 50°S - 30°S, 30°S - 0°, 0° - 30°N, 30°N - 50°N, 50°N - 75°N. We combine all measurements in each band to create a time-series of observational ethane. The measurements that are greater than 3σ of the deseasonalized data in each band were removed. The annual latitudinal band average, \bar{x}_l , is calculated as the average of all four seasonal means in one year. We exclude years with less than 4 seasons.

The annual hemispheric means are calculated as weighted means of \bar{x}_l . For the northern hemisphere (NH)

$$\bar{x}_{NH} = \bar{x}_{0-30}w_{0-30} + \bar{x}_{30-50}w_{30-50} + \bar{x}_{50-75}w_{50-75} \quad (E1)$$

and the normalized weights are determined using the sine of the band latitudes

$$w_l = \frac{\sin(\theta_{lh}) - \sin(\theta_{ll})}{\sin(75)} \quad (E2)$$

where θ_{lh} is the higher latitude of a latitudinal band, and θ_{ll} is the lower latitude of a latitudinal band.

The procedure to calculate the southern semispheric (SH) means is similar using the latitudinal bands in the southern hemisphere.

Finally, we defined the annual Interhemispheric Ratio (IHR) as the quotient of the annual Northern Hemispheric mean over the annual Southern Hemispheric mean.

i. Uncertainty Calculations

The uncertainty, $\Delta\bar{x}_l$, of \bar{x}_l is determined by propagating the error from the standard error

$$\Delta\bar{x}_l = \frac{1}{4} \sqrt{\sum_{s=1}^4 SE_s^2} \quad (E3)$$

where SE_s is the standard error of seasons, which is calculated as

$$SE_s = \frac{\sigma_s}{\sqrt{n}} \quad (E4)$$

n is the number of observations in a season, and σ_{season} is the standard deviation of the observations.

The uncertainty of the annual hemispheric mean, $\Delta\bar{x}_{NH}$ and $\Delta\bar{x}_{SH}$, is calculated as the propagation of uncertainties from each latitudinal band $\Delta\bar{x}_l$,

$$\Delta\bar{x}_{NH} = \sqrt{\sum_{l=1}^N (\Delta\bar{x}_l^2 w_l^2)} \quad (E5)$$

where N is the number of latitudinal bands of a hemisphere.

The uncertainty of the IHR, Δx_{IHR} , is the propagation of uncertainties from the hemispheric means' uncertainties; the result is as follow

$$\Delta x_{IHR} = \sqrt{\left(\frac{\Delta x_{NH}}{\bar{x}_{SH}}\right)^2 + \left(\frac{\bar{x}_{NH}\Delta x_{SH}}{\bar{x}_{SH}^2}\right)^2} \quad (E6)$$

ii. Simulated mixing ratio

The simulated mixing ratio fields from each emission scenario were sampled at the spatial and temporal coordinates of the observed data from each network; consequently, the simulated data has the same location and time span as the observational data. We then calculated the simulated IHR using the same procedure as the observational IHR.

b. Method 2

In method 2, we constructed a history of ethane at the same site to eliminate the variation due to stations changing over time as in method 1.

i. Observational IHR

We used the observational ethane data from Barrow, Alaska, USA (71.3°N, 156.6°W) and Cape Grim, Tasmania, Australia (40.7°S, 144.7°E) to represent the ethane mixing ratio in the Northern Hemisphere and the Southern Hemisphere. The Barrow measurements are the longest ethane record that we have available from all 3 networks. The UCI network does not have data for the Cape Grim site, so in order to obtain a continuous record, we used UCI stations between latitudes 38°S to 46°S to represent Cape Grim for the UCI network. From our sensitivity study,

the UCI annual means are not sensitive to the latitudinal bounds that are used to represent Cape Grim in method 2 (see Sensitivity in Supplemental Section).

The IHR of method 2 is calculated using the same method as method 1 without applying the latitudinal weights. The Northern Hemisphere is replaced with Barrow, Alaska and Southern Hemisphere with Cape Grim, Tasmania. Figure xx shows the normalized IHR of method 1 and method 2.

ii. Simulated IHR

We used the GEOS-Chem output data at 71.3°N, 156.6°W for Barrow and 40.7°S, 144.7°E for Cape Grim to construct the simulated data set for this analysis. The simulated data set is analyzed using the same methods as the observational mixing ratio of method 2.

References

Figures

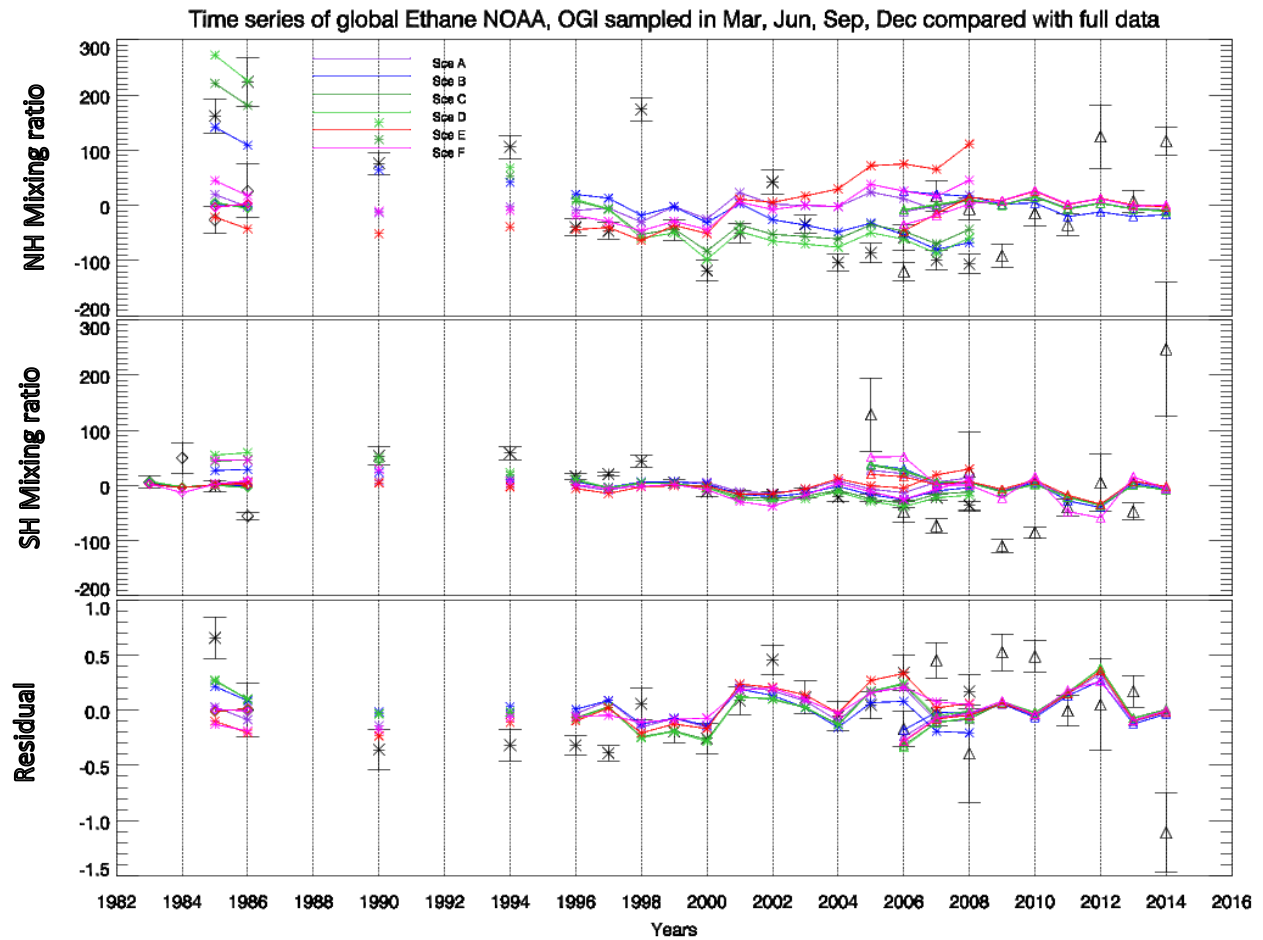


Figure 1: Normalized time series of global ethane. The symbols represent different networks: diamond – OGI, asterisk – UCI, triangle – NOAA. The color black is observational data; other colors are simulated emission scenarios.

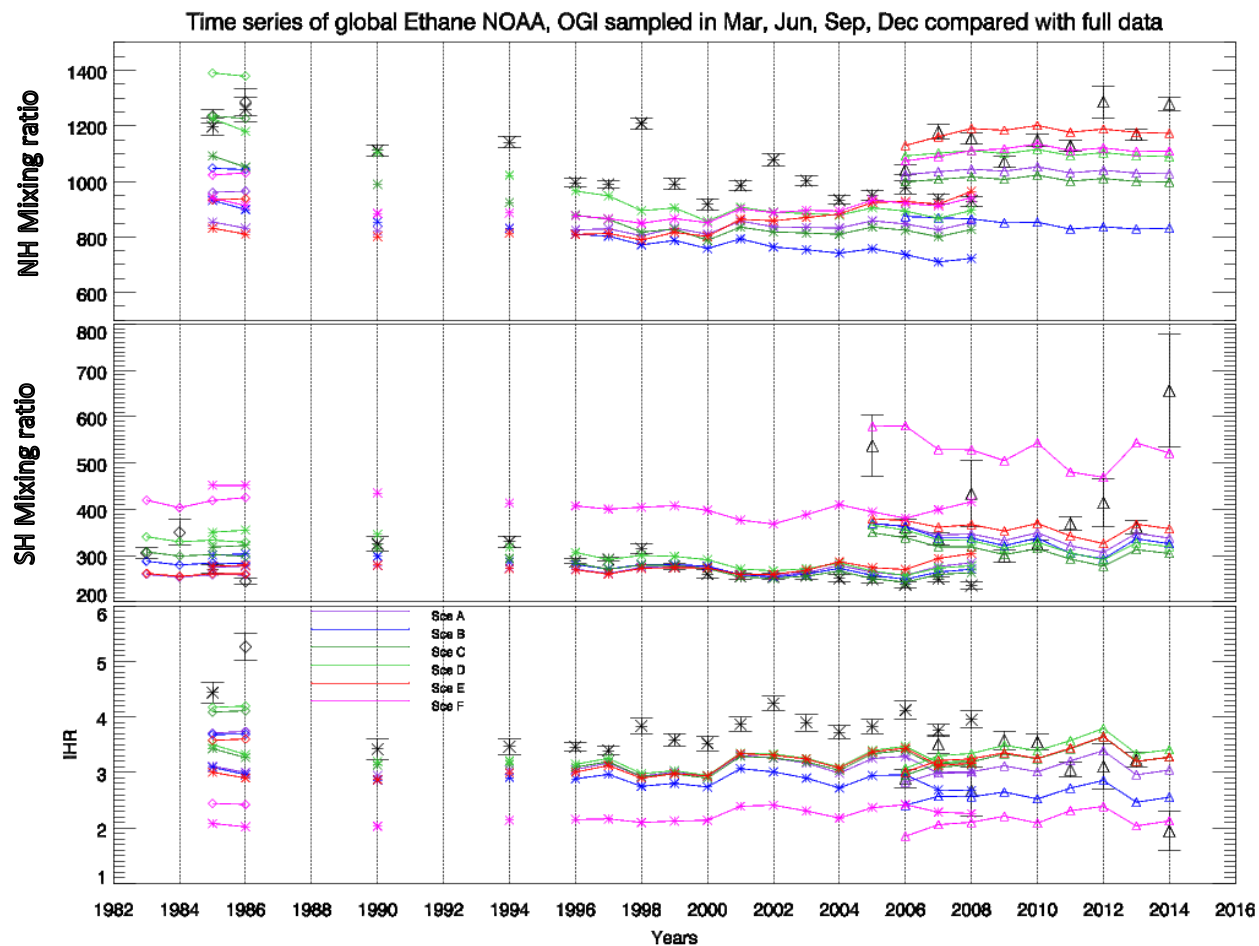


Figure 2: Time series of global ethane. The symbols represent different networks: diamond – OGI, asterisk – UCI, triangle – NOAA. The color black is observational data; other colors are simulated emission scenarios.

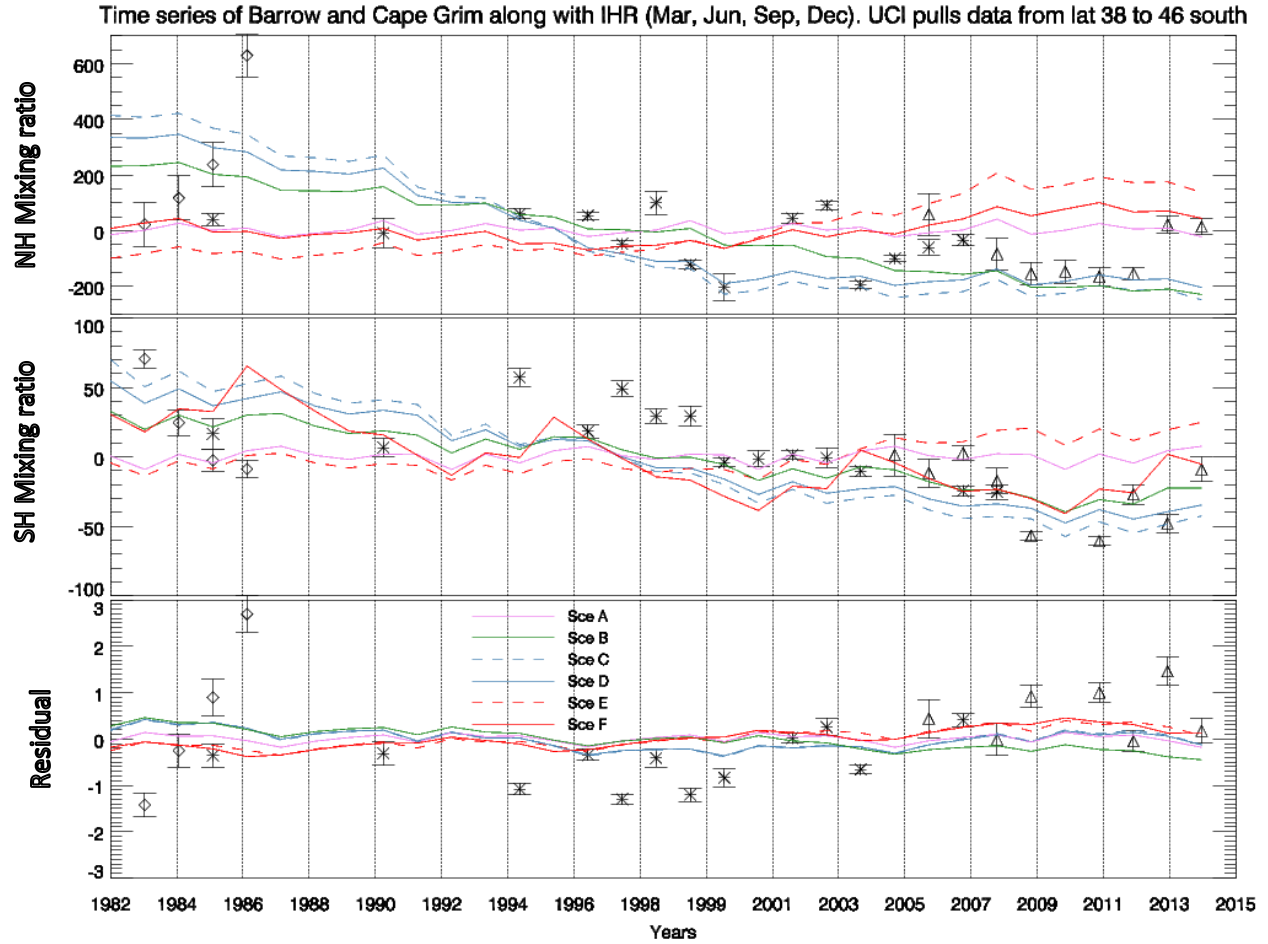


Figure 3: Normalized time series of ethane calculated using Method 2. The symbols represent different networks: diamond – OGI, asterisk – UCI, triangle – NOAA. The color black is observational data; other colors are simulated emission scenarios.

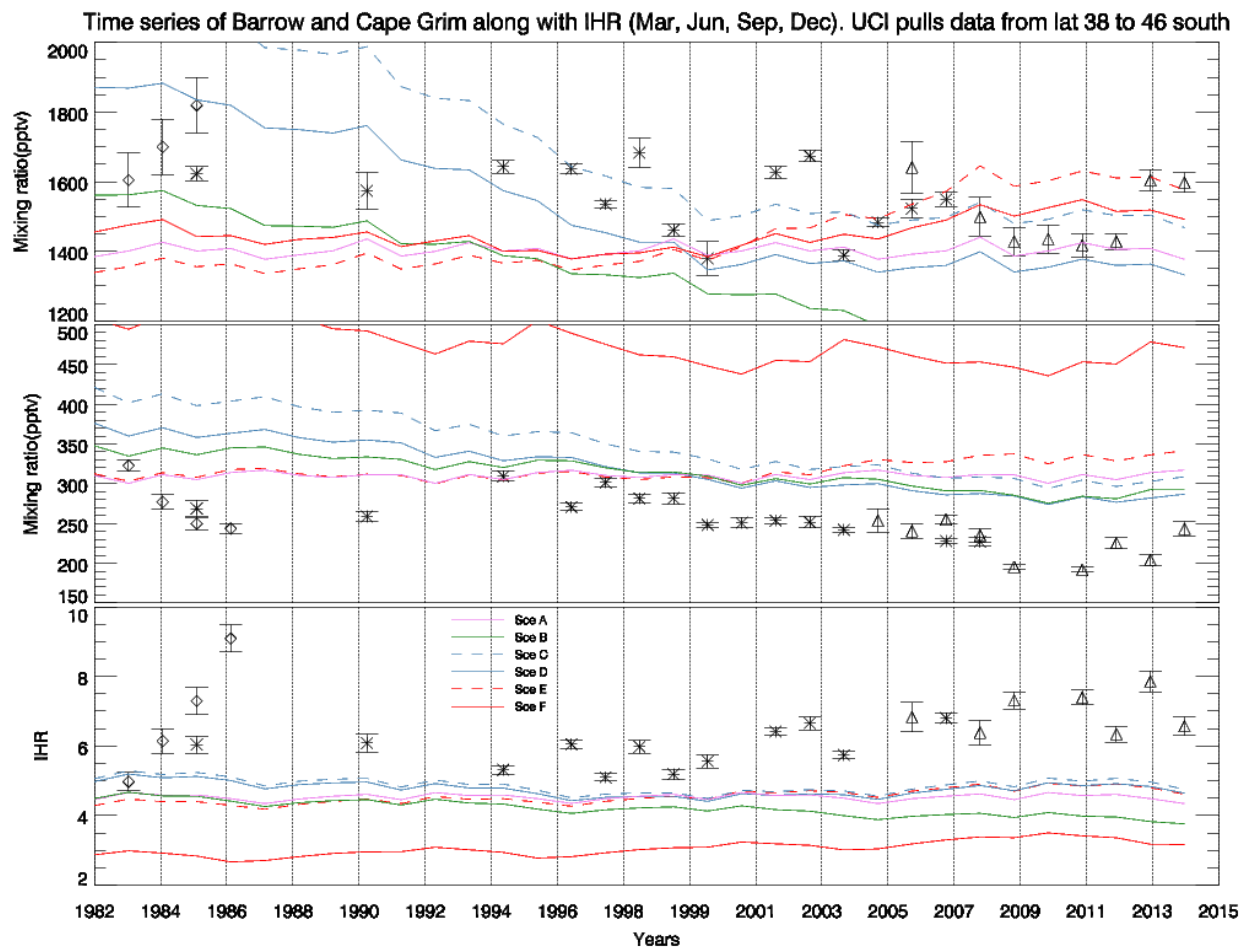


Figure 4: Time series of ethane calculated using Method 2. The symbols represent different networks: diamond – OGI, asterisk – UCI, triangle – NOAA. The color black is observational data; other colors are simulated emission scenarios.