1/ Observational surface mixing ratio of ethane

The annually observational averaged ethane mixing ratio is calculated 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 are composed of 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). Respectively, 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/ Simulated modeling of ethane

3/ Calculations of Interhemispheric Ratio

The annually averaged ethane mixing ratio is sensitive to small differences in the instrumental calibration of each laboratory. Therefore, we use the Interhemispheric Ratio (IHR) to infer the trend of ethane mixing ratio from 1983 to 2014. We expect that the IHR will eliminate the absolute calibration differences between datasets.

a. Global IHR calculations ("Method 1")

i. IHR

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

$$\bar{x}_l = \frac{1}{4}\sum_{i=1}^{4}(\bar{x}_{season})_i \tag{E1}$$

$\bar{x}_{season}$ is the mean of a season.

The annual hemispheric means are calculated as a weighted means of $\bar{x}_l$ as following for the northern hemisphere (NH)

$$\bar{x}_{NH} = \bar{x}_{0-30} w_{0-30} + \bar{x}_{30-50} w_{30-50} + \bar{x}_{50-75} w_{50-75} \tag{E4}$$

and the weights are determined using the sine value of the latitudes

$$w_l = \frac{\sin(higher\_lat) - \sin(lower\_lat)}{\sin(highest\_lat)} \tag{E5}$$

The procedure to calculate the Southern Hemispheric (SH) means is similar.

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

ii. Uncertainty Calculations

The uncertainty, Δx_l, of $\bar{x}_l$ is the propagation of error from the standard error of each season resulted in the following

$$\Delta x_l = \frac{1}{4}\sqrt{\sum_{i=1}^{4} (SE_{season})_i^2} \tag{E2}$$

where SE_{season} is the standard error of each season, which is calculated as

$$(SE_{season})_i = \frac{(\sigma_{season})_i}{\sqrt{n}} \tag{E3}$$

n is the number of samples in a season, and σ_{season} is the standard deviation of a season. (How to talk about sites with only 1 measurement in a season?)

The uncertainty of the annual hemispheric mean, Δx_{NH} and Δx_{SH}, is calculated as the propagation of uncertainties from each latitudinal band Δx_l,

$$\Delta x_{NH} = \frac{1}{w_{total}} \sqrt{\sum_{i=1}^{N} (\Delta x_l^2 w_l^2)_i} \quad \text{(E7)}$$

where N is the number of bands of a hemisphere, and w_{total} is the sum of the weights in a hemisphere.

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

$$\Delta x_{IHR} = \sqrt{\left(\frac{\Delta x_{NH}}{\bar{x}_{SH}}\right)^2 + \left(\frac{\bar{x}_{NH} \Delta x_{SH}}{\bar{x}_{SH}^2}\right)^2} \quad \text{(E8)}$$

iii. 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 calculated the simulated IHR using the same method as the observational IHR except for the uncertainty calculations.

b. Method 2

What is the motivation for calculating IHR using Barrow and Cape Grim?

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 UCI network does not have data for the

Cape Grim site, so in order to obtain a continuous record, we used UCI ethane mixing ratios between latitudes 38°S to 46°S to represent Cape Grim for the UCI network.

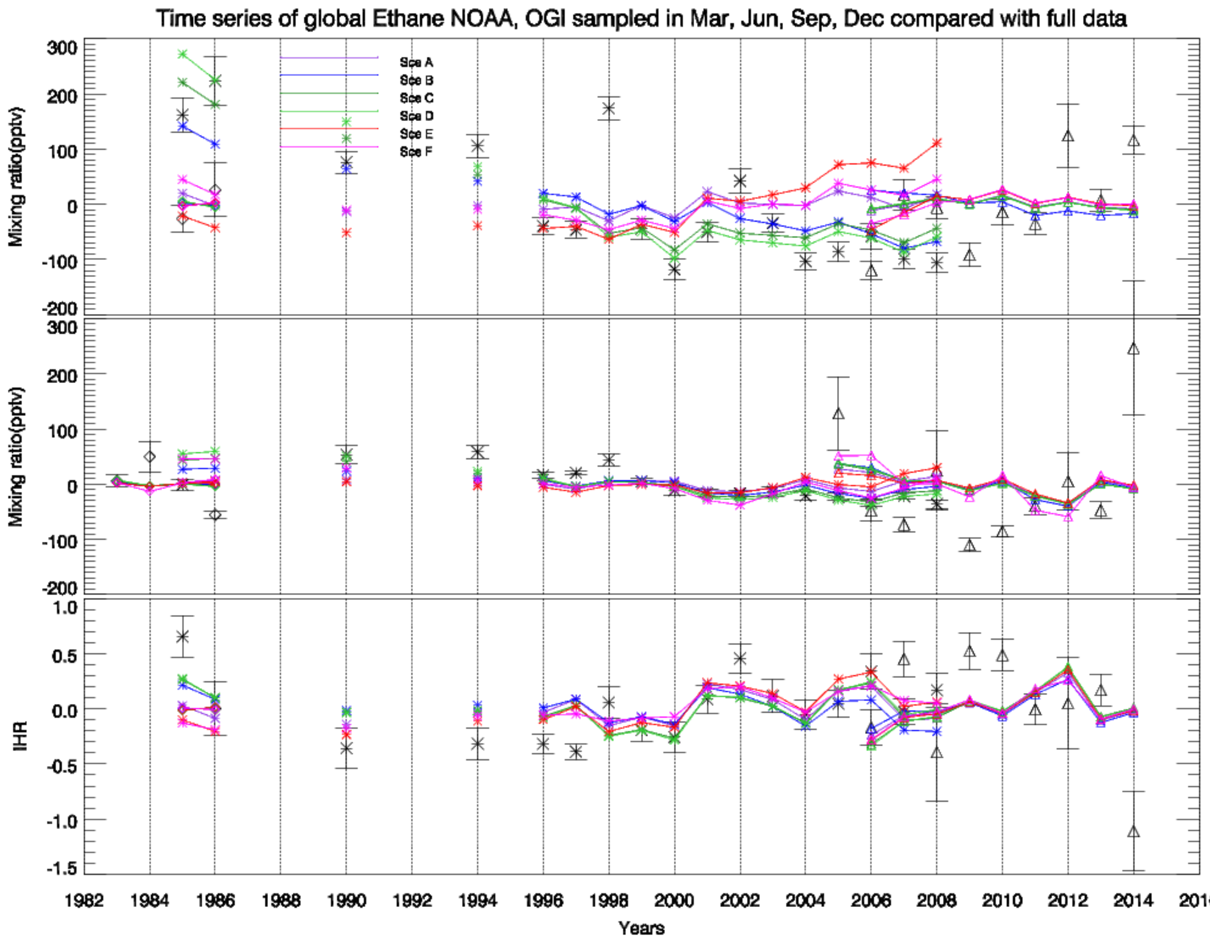
The IHR of method 2 is calculated using the same method as “method 1” without applying the latitudinal weights. The Northern Hemisphere is replaced with Barrow, Alaska and Southern Hemisphere with Cape Grim, Tasmania.

ii. Simulated IHR

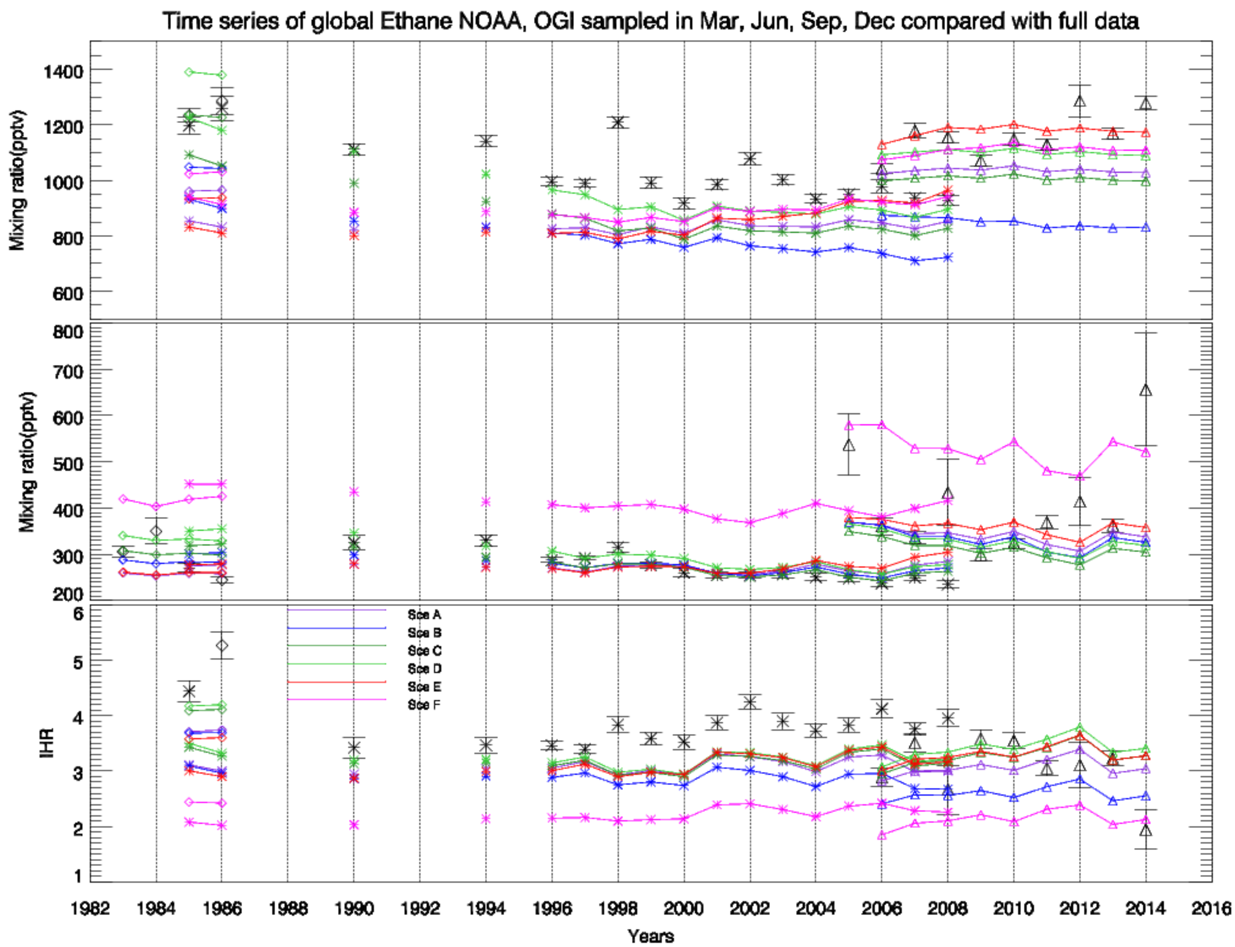
We used the GEOS-Chem output data at 71.3°N, 156.6°W for Barrow and 40.7°S, 144.7°E for Cape Grim to construct the simulated data set for this analysis. The simulated data set is analyzed using the same methods as the observational mixing ratio of method 2.

References

Figures



Time series of global Ethane NOAA, OGI sampled in Mar, Jun, Sep, Dec compared with full data
Sce A
Sce B
Sce C
Sce D
Sce E
Sce F
Mixing ratio(pptv)
Mixing ratio(pptv)
IHR
Years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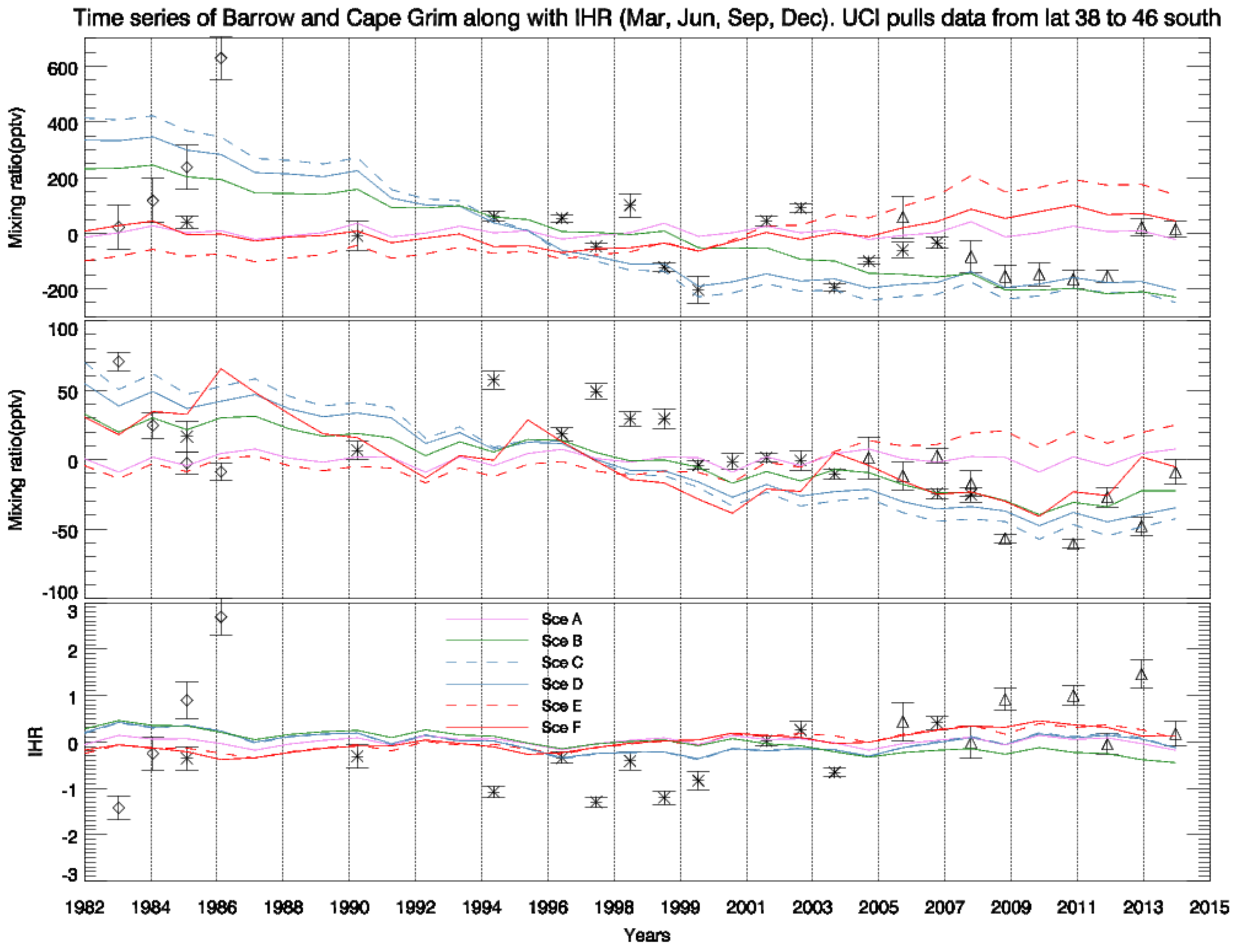
Time series of Barrow and Cape Grim along with IHR (Mar, Jun, Sep, Dec). UCI pulls data from lat 38 to 46 south
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Mixing ratio(pptv)
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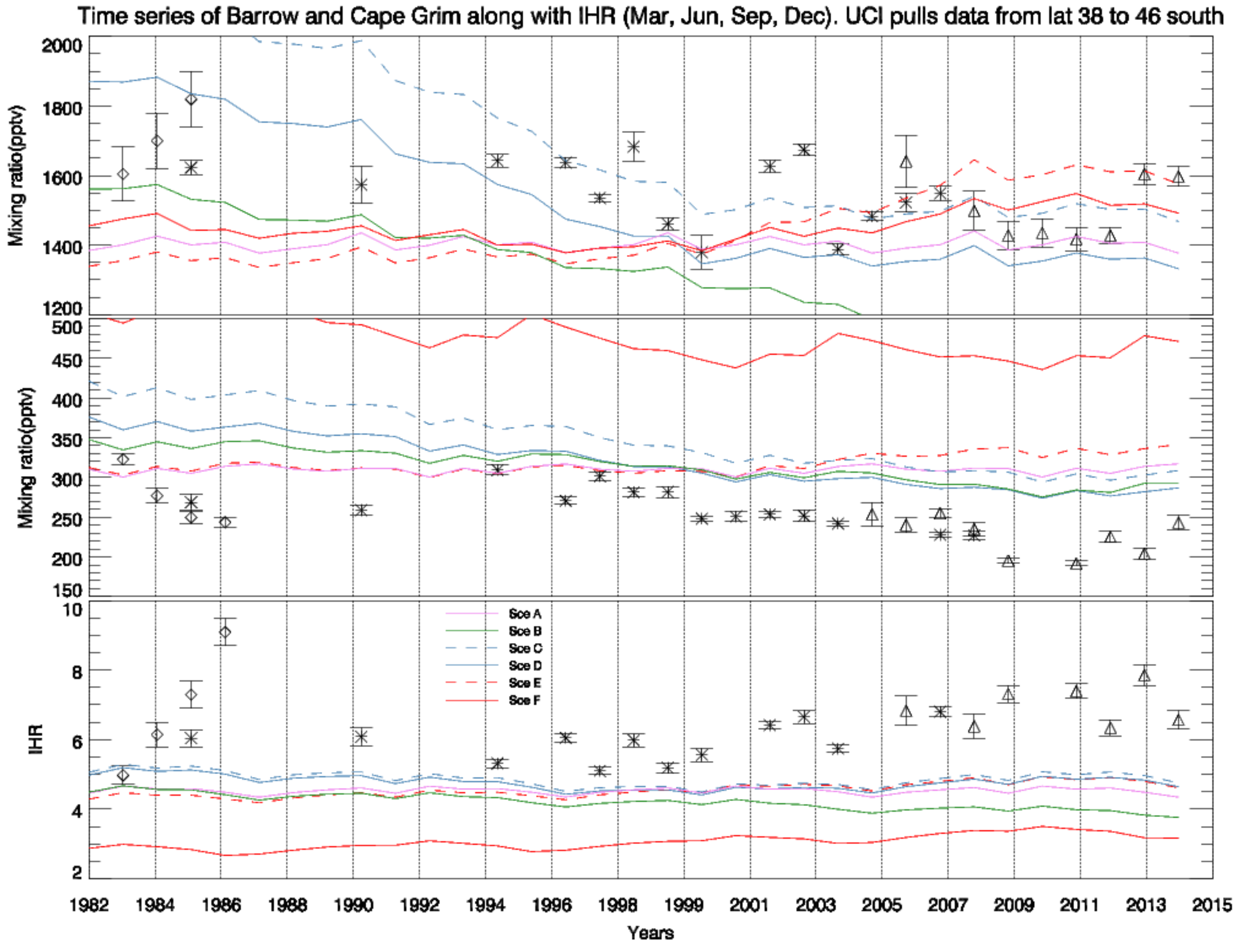
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