**1/ Simulation modeling**

**2/ First Method to calculate IHR**

1. **Observational Data**
2. **Air Archives:**

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 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 a full-year sample is 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 data were collected weekly in the Pacific Basin from remote surface locations and analyzed at the UCI laboratory using gas chromatography within one month after the collection. The data 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 record is available in the following years: 1985, 1990, 1994, 1996-2008 (Simpson et al. 2012)

The NOAA data has 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 the maximum ratio occurs in March and the minimum ratio occurs in September (show a fig of a notable site). The UCI data are only available in March, June, September, and December, which correspond to the maximum, minimum and the inflections of the seasonal cycle. We defined each of the 4 months as a season and only the aforementioned 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 those latitudes.

1. **Calculations:**

We divide the atmosphere into 5 latitudinal bands: 50°S - 30°S, 30°S - 0°, 0° - 30°N, 30°N - 50°N, 50°N - 75°N. The data in each band is deseasonalized by removing the average seasonal cycle calculated over an averaging window of multiple years at monthly resolution (Fig?). A Gaussian fit is applied to the deseasonalized data to obtain the standard deviation (σ) of the latitudinal band (Fig. xx). We removed data that is 3σ away from the mean.

The annual latitudinal band average, , is calculated as the average of all four seasonal means in one year.

(E1)

The uncertainty, of each latitudinal band is the propagation of error from the standard error of each season resulted in the following

(E2)

where SEseason is the standard error of each season, which is calculated as

(E3)

n is the number of samples in a season.

The annual hemispheric means, is calculated from the weighted mean of the latitudinal bands as following for the northern hemisphere

(E4)

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

(E5)

For the southern hemisphere, the mean is calculated as

(E6)

and for the weights

(E6)

The uncertainty of the annual hemispheric mean, is calculated as the propagation of uncertainty from each latitudinal band :

(E7)

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

The mixing ratio in the northern hemisphere is 3-4 times higher than the mixing ratio in the southern hemisphere (need to find a source to cite? or say from observation?), thus global mixing ratio gradient is sensitive to the changes in the emissions of the northern hemisphere. To compare the ethane mixing ratio gradient with different emission scenarios without the influence of the northern hemisphere, we defined the Interhemispheric Ratio (IHR) as the quotient of the Northern Hemispheric Mean (NHM) over the Southern Hemispheric Mean (SHM). We expect that the IHR can also eliminate the bias in the absolute calibration differences that might exist between different sampling networks. The uncertainty of the IHR, , is the propagation of uncertainty from the hemispheric means’ uncertainties; the result is as follow

(E8)

**3.1.2/** **Simulated data**

The simulated mixing ratio to use for the global analysis is built from the GEOS-Chem data structure output using the spatial and temporal parameters of the observed data from each network; consequently, the simulated data will have the same location and time span as the observed data. There are 6 simulated data sets corresponding to 6 emission scenarios. Each simulated data set goes through the same calculations as the global observed data analysis except for the uncertainty calculations (need to justify not using uncertainty calculations for simulated data or should it be in the discussion section?)

Fig. xx shows the observed NHM, SHM and the IHR as discrete data points and the simulated data as continuous lines. To compare the trends of the observed ethane mixing ratio and the simulated mixing ratio, we normalized the data over the time period within a sampling network; Fig. xx shows the normalized data.

**3.2/ 2-site analysis**

**3.2.1/ Observed Data**

We used the observed 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 compare with the time series of the Barrow site, we used UCI data between latitudes 38°S to 46°S to represent Cape Grim for the UCI network. (How to justify the use of the latitude bounds? or refer back to it in the sensitivity study section)

The 2 sites are deseasoned using the same technique as the global analysis. The same data filtering algorithm is also applied to each site where data larger than 3σ are removed.

Similar to the global analysis, the annual mean of each site, , is also calculated as the average of the mean of each season.

(E9)

The uncertainty of the annual mean, , is the propagation of error from the standard error of each season.

(E10)

The Interhemispheric Ratio (IHR) of the 2-site analysis is defined as the quotient of the annual means of the Barrow site over the Cape Grim site. The uncertainty of the IHR, , is the propagation of uncertainty from the annual uncertainties of each site. The result is similar to the equation E8 where Barrow represents the Northern Hemisphere and Cape Grim represents the Southern Hemisphere.

(E11)

**3.2.2/ Simulated Data**

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 observed data set of the 2-site analysis. The annual mean of the ethane mixing ratio is a simple average of all 4 seasons in one year; the IHR is the quotient of the annual mean of Barrow over Cape Grim of the same year. Fig. xx shows the time series of the annual ethane IHR resulted from the 2-site analysis and the annual means of the Barrow site and the Cape Grim site; the simulated emission scenarios are shown as continuous lines. Fig. xx shows the same time series but normalized over individual data network.

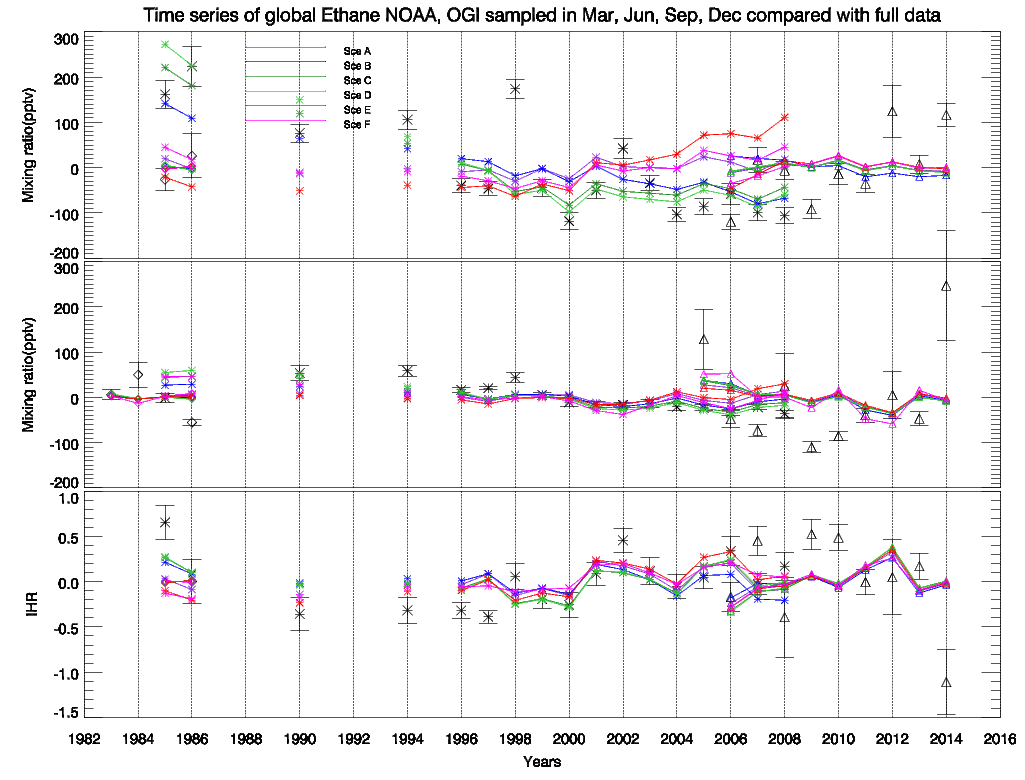
**4/ Sensitivity**

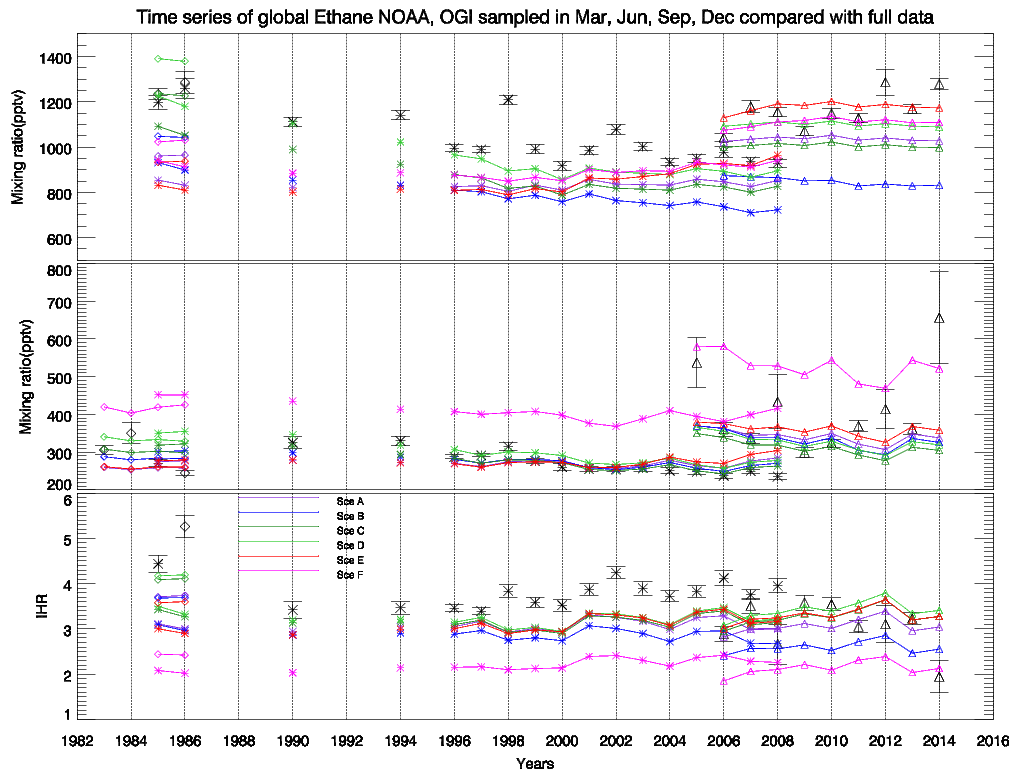
**4.1/ Observed data sensitivity**

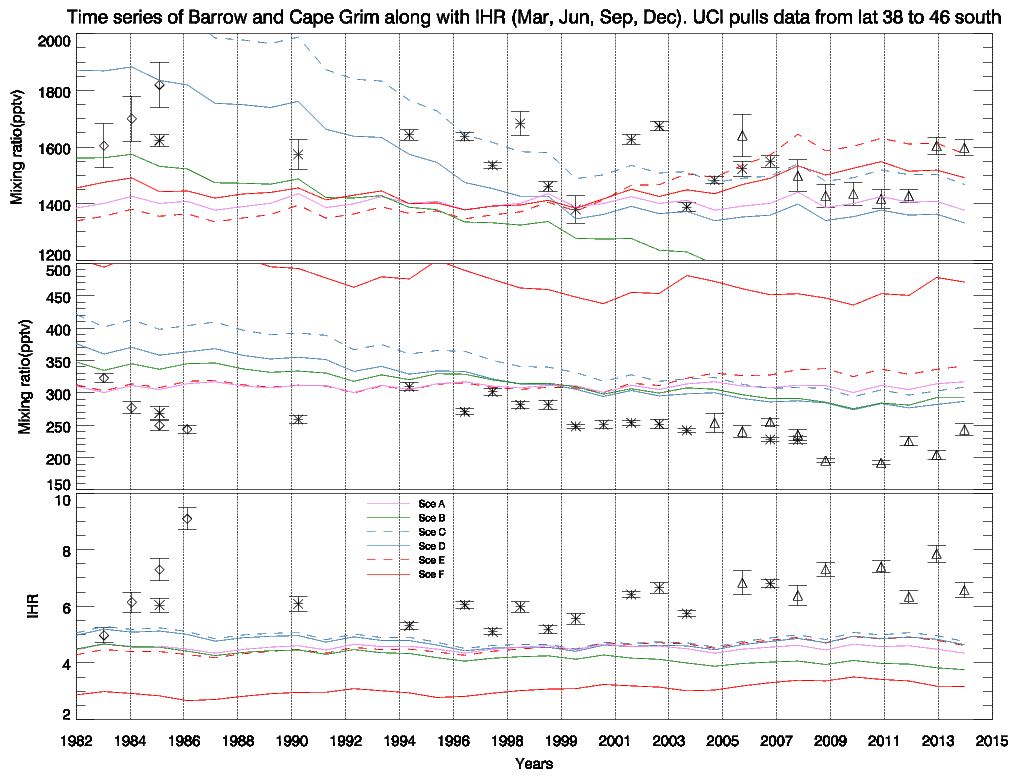
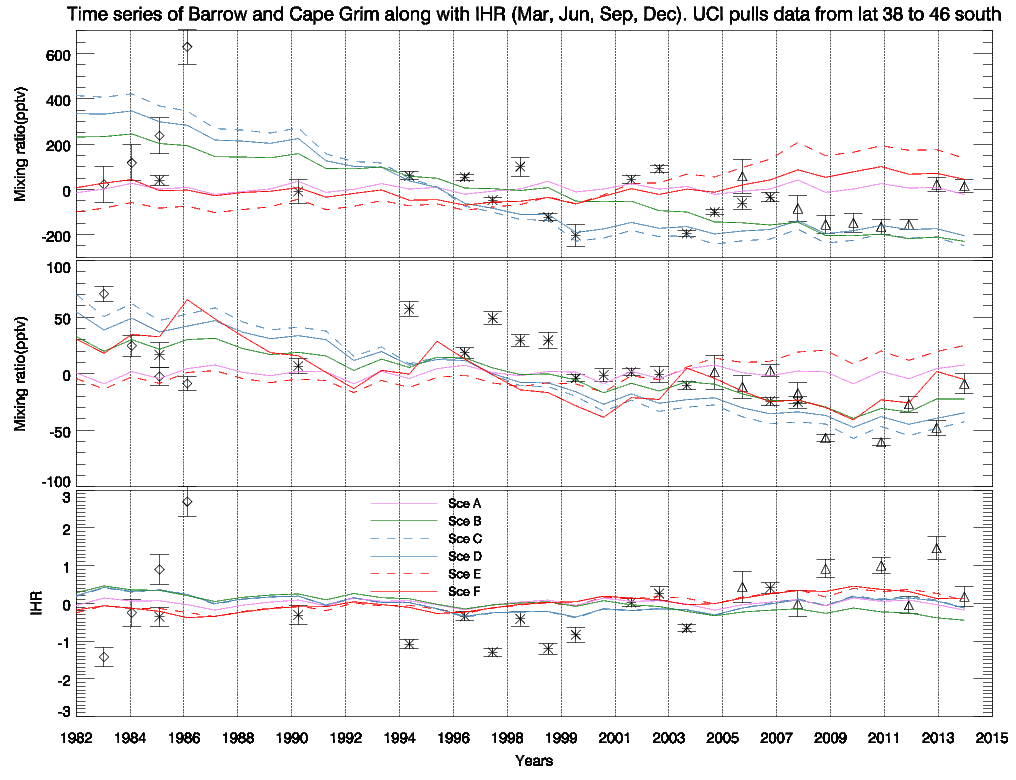
**4.2/ Simulated data sensitivity**

**References**

**Figures**

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