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# Ratios of Greenhouse Gas Emissions Observed over the Yellow Sea and the East China Sea Y. Liu<sup>1</sup>, L. Zhou<sup>1</sup>, P.P. Tans<sup>2</sup>, K. Zang<sup>1</sup> and S. Cheng<sup>1</sup>

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During a cruise of the survey vessel Dongfanghong II on the Yellow Sea and the East China Sea in the spring of 2017 we performed accurate measurements of the mole fractions of carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), carbon monoxide (CO), and nitrous oxide (N<sub>2</sub>O) using two types of Cavity Ring-Down Spectrometers (CRDS). The spatial variations of the mole fraction of the four trace gases were very similar. The emission sources of these gases were divided into several regions by using the NOAA HYSPLIT model. Then we analyzed the variations of the ratios of the mole fraction enhancements between every pair of trace gases downwind of these source areas. The ratios showed that the distributions of these trace gases over the Yellow Sea and the East China Sea in the spring were mainly caused by the emissions from Eastern China. The much higher enhancement ratio of  $\Delta CO/\Delta CO_2$  and the lower ratio of  $\Delta CH_4/\Delta CO$  observed in the air parcels from big cities like Beijing and Shanghai indicated high CO emission from the cities during our time of observation. Compared with the values of NOAA's Marine Boundary Layer (MBL), the ratios of the averages in the air coming from the northern sector (Russia) were on average closer to the MBL, and the air that stayed over the Yellow Sea and the East China Sea was a mixture of emissions from wide regional areas. The highly variable N<sub>2</sub>O data of the air from Qingdao and Shanghai showed much more fluctuation.





The R/V DongFangHong2 is a specially designed marine survey vessel, equipped with varies observation and sampling instruments.



**Measurement system in the atmospheric Lab**  $N_2O$  mixing ratio was measured every 4-10 s by a CRDS 5101i analyzer.  $CO_2/CH_4/CO$  mixing ratios was measured every 5 s by a CRDS 2401 analyzer.









### Calibrations

#### **Stability of successive calibrations (CO<sub>2</sub>)**

### Stability of successive calibrations $(CH_4)$

#### **Stability of successive calibrations (CO)**

#### Stability of successive calibrations (N<sub>2</sub>O)

CB09388 N<sub>2</sub>O

- Response of the analyzers were calibrated 35 times during the whole voyage. Every standard gas ran for ten minutes each time.
- All four standards were used during a calibration were to generate a response curve for each instrument.
- During sample air measurements, in between the calibrations, we used the average of the bracketing calibrated response curves to correct the observation data.



#### Air trajectories

to clarify the mole fraction distribution, we calculated the hourly average data and used the NOAA HYSPLIT Model to generate 72-hour backward air trajectories, with the starting point at 300 m altitude.



Wind vector over the China sea-shelf boundary layer



**Preliminary results** (GHG mole fraction during the voyage)



CO

# **Enhancement ratios**

• we use two different methods to calculate the enhancement ratios and the uncertainties of every pair of the four gases, and compared with the MBL values of CO<sub>2</sub>, CO, CH<sub>4</sub>, N<sub>2</sub>O during the same time.



Average values for the Philippine Sea observed during the cruise are <u>410.7ppm, 113ppb, 1896ppb, 332.5ppb</u> NOAA's MBL values, representative of large ocean areas, at the same latitude zone and during the cruise, were <u>410.7ppm, 128ppb, 1911ppb, 330.2ppb</u>

Time

#### This indicated that our Philippine Sea data was good reference values for this study.

We looked at the difference of the ratios among different source regions.





#### **Ratios pairwise between the observed enhancements** relative to the Philippine Sea, $\Delta CO_2$ , $\Delta CO_2$



bars gave the number of hourly averages used for each region, n1 for low-noise data (blue and

gray bars), n2 for high-noise data (orange and yellow bars). We did not plot bars when they

were based on 5 or fewer hourly averages.

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# Conclusions

- The two Picarro analyzers G2401 for CO<sub>2</sub>, CH<sub>4</sub>, CO and G5101i for N<sub>2</sub>O show good stability during the voyage on the ship.
- The characteristics of the mole fraction distributions of the four gas species are very similar to each other.
- According to the NOAA HYSPLIT Model, all the data are divided into different source regions, which are mainly north of China and coastal cities of East China.

