



Sensitivity of Upper Tropospheric Ozone to Its Precursors During the Asian Summer Monsoon

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Key Messages

- Ozone (O_3) in the upper troposphere (UT) increased by $\sim 70\%$ in 2014 compared to 1850 during the Asian summer monsoon (ASM), driven by increased emissions of NO_x , CO, and NMVOCs.
- Ozone production in the UT during the ASM is primarily NO_x -limited and favors the CO - NO_x - O_3 photochemical pathway.
- Excess ozone production in the UT led to a positive radiative effect of 0.9 Wm^{-2} at the top of the atmosphere (TOA).

Introduction

- Role of Tropospheric Ozone:**
 - Surface:** Acts as a harmful pollutant to human health and ecosystems.
 - UT:** Functions as a potent greenhouse gas governing Earth's radiative balance.
- Why Focus on the UT?**
 - Maximum Efficiency:** Longer lifetime and high per-molecule greenhouse efficiency.
 - Disproportionate Impact:** Small UT ozone changes trigger large shifts in radiative forcing.
- Emissions**
 - Pre-Industrial vs. Present-Day:** Anthropogenic precursor emissions (NO_x , CO, NMVOCs) have risen dramatically since 1850 due to industrialization, energy production, and transport (Fig. 1).
 - Pollution Pump:** Deep convection during the ASM (JJAS) uplifts surface pollutants into the ASM Anticyclone (ASMA).

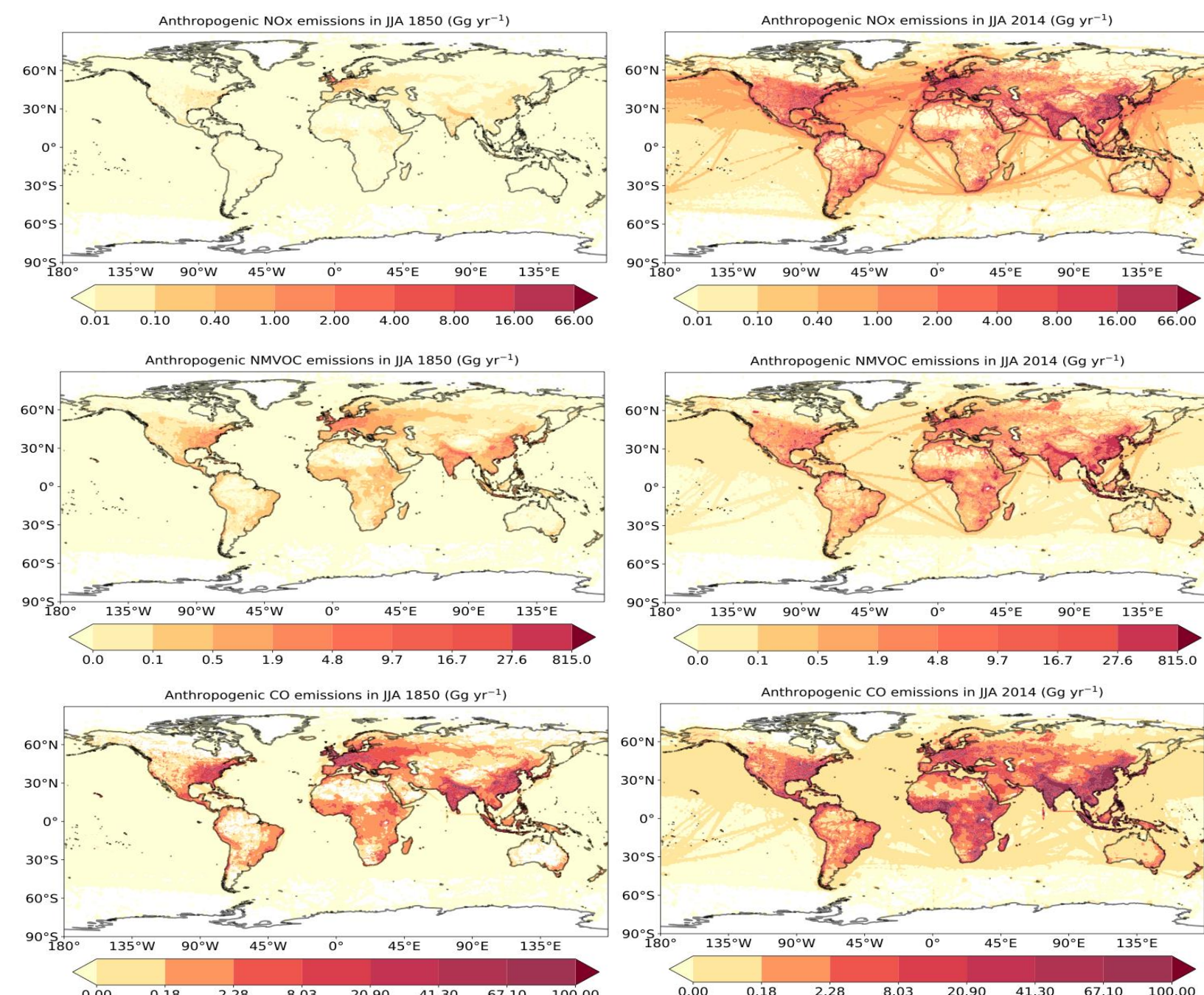


Fig. 1: NO_x , NMVOCs, CO emissions during 1850 and 2014

- NO_x vs. VOC Sensitivity Regimes**
 - NO_x - vs. VOC-sensitive:** Regions where ozone is strongly sensitive to NO_x changes are NO_x -limited, whereas regions sensitive to VOC changes are VOC-limited.
 - HCHO/ NO_2 ratio:** This ratio serves as a widely accepted metric to distinguish between NO_x -limited, VOC-limited regimes.
- Climate Relevance:** Enhanced human-driven precursors in the UT and inside the ASMA alter local ozone production and TOA radiation.
- Understanding ozone sensitivity to NO_x or VOCs in UT is essential for designing effective emission control policies that address both surface-level sources and their upper-atmospheric impacts.**

Objectives

- Quantify UT Ozone changes & precursor impacts:** Evaluate how UT ozone and its precursors (NO_x , CO, and NMVOCs) changed over the Asian Summer Monsoon Anticyclone from 1850 to 2014.
- Determine chemical regimes & radiative effects:** Identify dominant photochemical pathways in the UT using indicator ratios and calculate the climate impact in terms of radiative forcing changes at TOA.

Methodology

Experiments	Period of simulation	O_3 precursors
piClim-control	30 years	1850
piClim- O_3	30 years	2014 (NO_x + NMVOCs + CO)
piClim- NO_x	30 years	2014 (NO_x)
piClim-VOC	30 years	2014 (NMVOCs + CO)

- Models:** BCC-ESM1, GFDL-ESM4, and GISS-E2-1-G
- Study season:** ASM months of June-Jul-Aug (JJA)
- Changes in O_3 , NO_x , VOCs, their ratios, and RF due to increasing emissions = piClim- O_3 (or NO_x or VOC) – piClim-control**

Results

UT ozone & precursor impacts

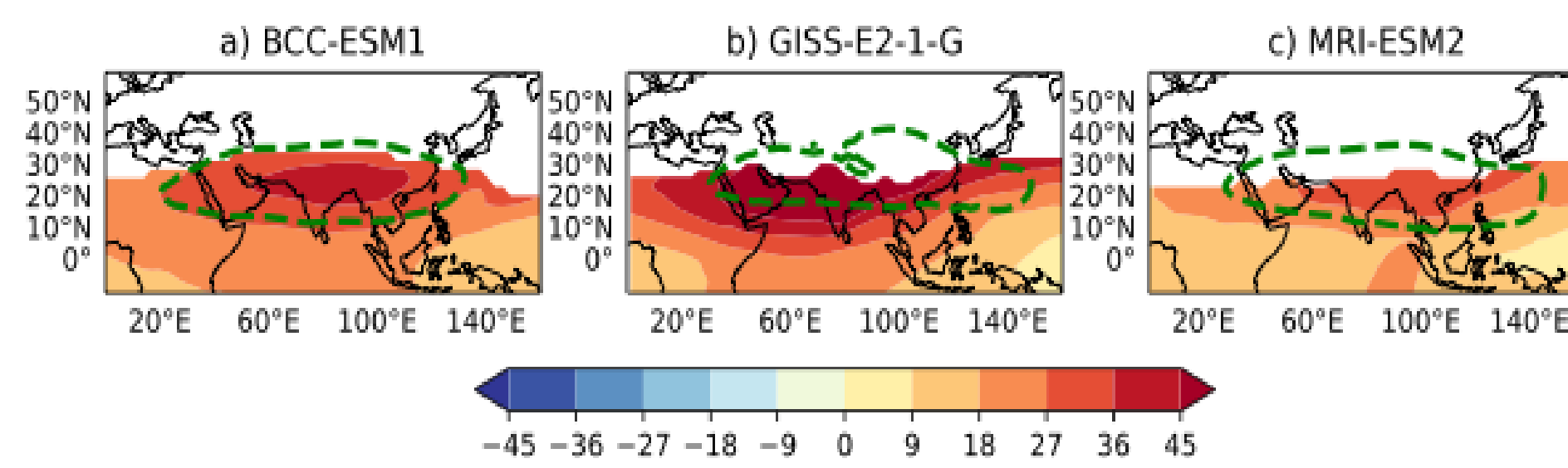


Fig. 2: Ozone changes between 1850 and 2014

- Present-day anthropogenic emissions increased UT ozone by $\sim 70\%$ (~ 35 ppb) inside the ASMA, with peak increases over the Indo-Gangetic Basin (Fig. 2).

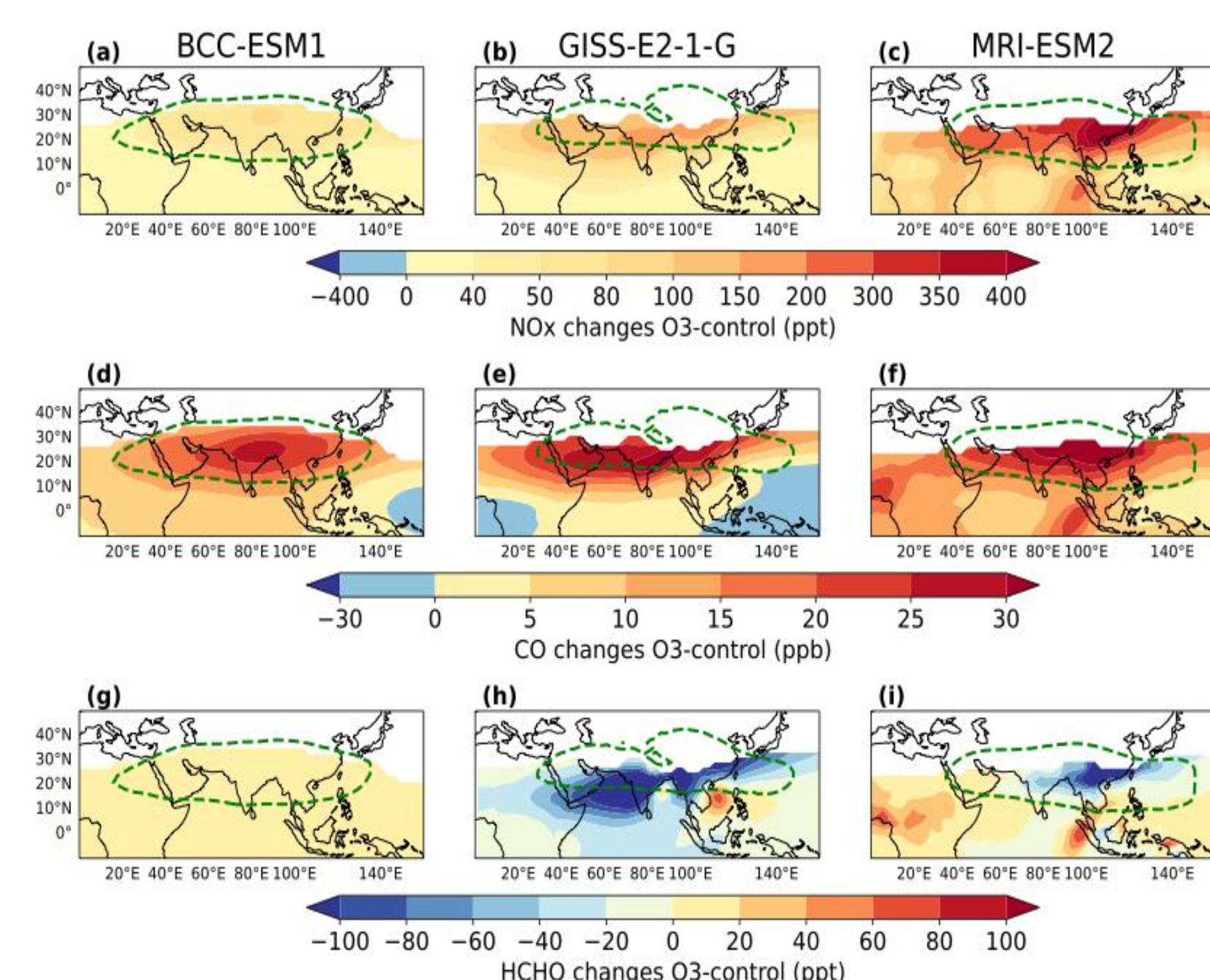


Fig. 3: NO_x , CO, HCHO changes between 1850 and 2014

- NO_x increased substantially across all models (80-300 ppt) due to transport and local production.
- CO increased by 20-40 ppb.
- HCHO changes in the UT remained predominantly negative or marginally positive, driven by changes in OH concentrations (Fig. 3).

Results

Chemical regime in the UT – NO_x vs. VOC limited

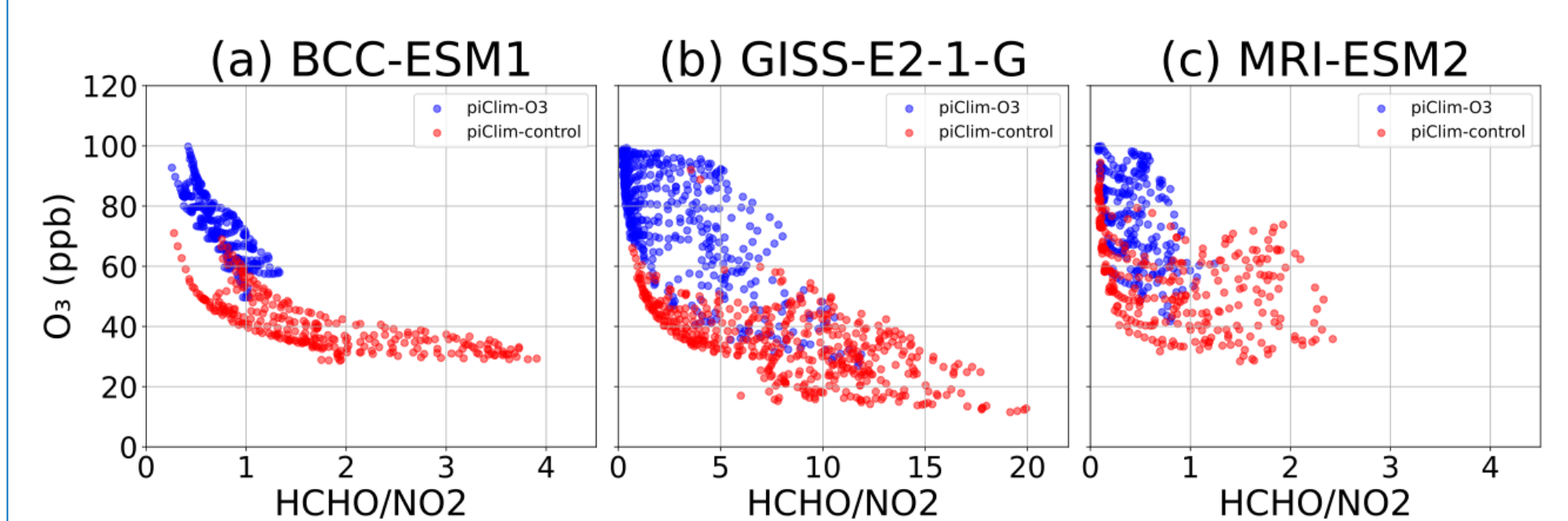


Fig. 4: Ozone vs. HCHO/ NO_2 during 1850 and 2014

- HCHO/ NO_2 ratio decreased by 0.8-5 ppt/ppt**, driven by a rapid increase in anthropogenic NO_x emissions compared to VOCs.
- Downward shift indicates a **transition from a more NO_x -sensitive regime toward a less NO_x -limited regime**.
- Higher ozone mixing ratios (50-100 ppb) consistently cluster at lower HCHO/ NO_2 values and vice-versa.
- Present-day simulations shift toward lower ratios while achieving higher ozone concentrations.

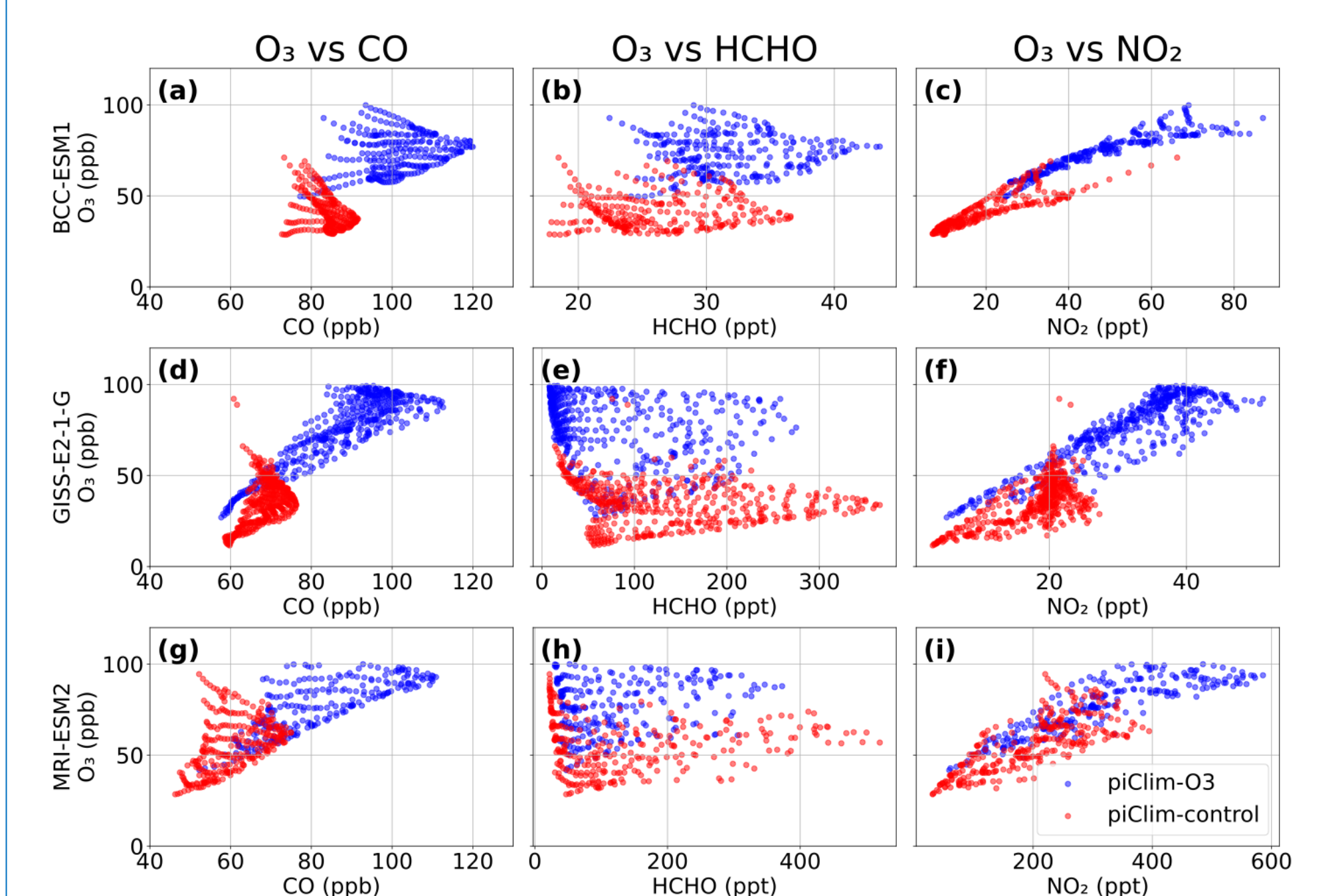


Fig. 5: Ozone vs. CO, HCHO, NO_x during 1850 and 2014

- Strong NO_2 Response:** Increasing NO_2 consistently enhances UT ozone, confirming strong NO_x limitation.
- Inverse HCHO Trend:** Higher HCHO aligns with lower ozone, reinforcing that UT chemistry is NO_x -limited rather than VOC-limited.
- CO Pathway Dominance:** Rising CO substantially increases ozone, proving the dominance of the CO - NO_x - O_3 pathway.

Radiative effects

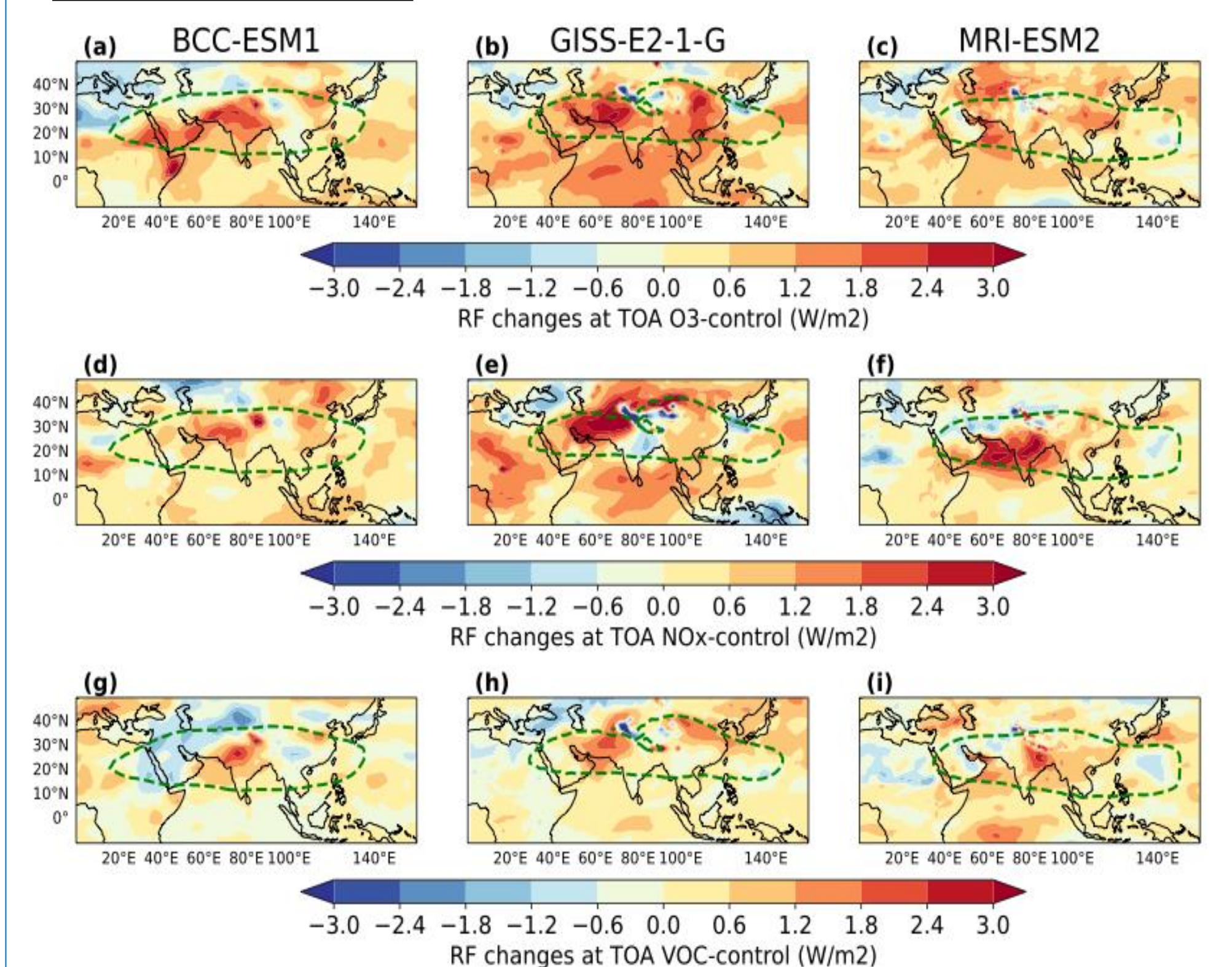


Fig. 6: RF changes due to NO_x , CO, HCHO

- Total precursors together drive a radiative effect of **$+0.90 \text{ W/m}^2$** over the ASMA (Fig. 6).
- NO_x -driven changes = **$+0.67 \text{ W/m}^2$**
- NMVOCs and CO- driven changes = **$+0.36 \text{ W/m}^2$**

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