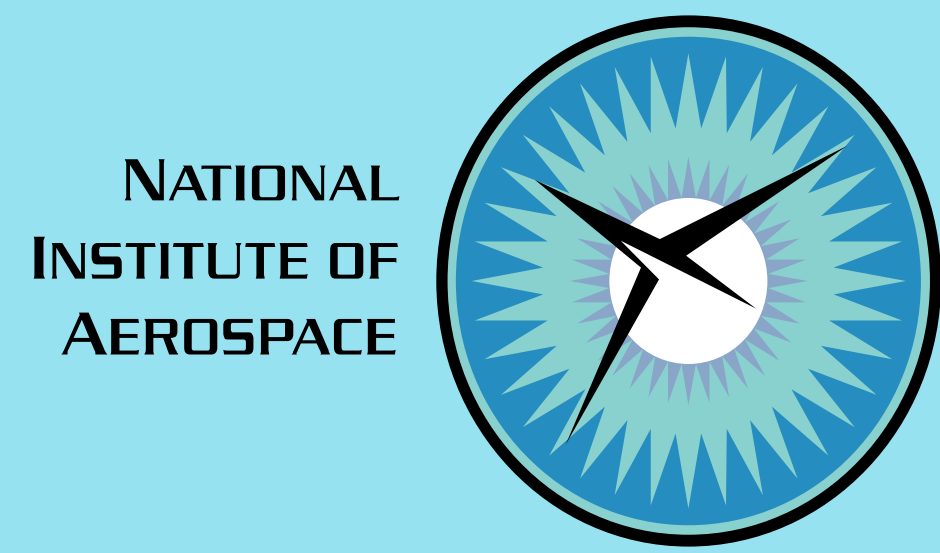
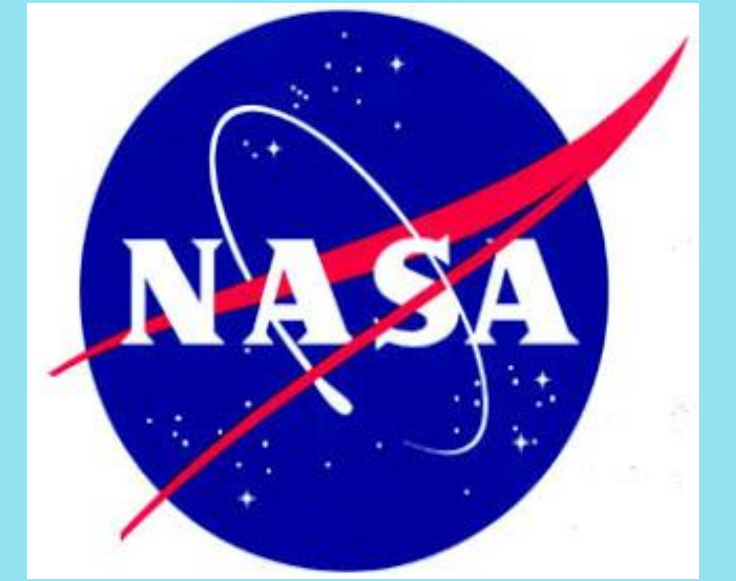




Radiative Effects of Aerosols Versus Clouds on Key Tropospheric Oxidants in Global Models

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Introduction

➤ Aerosols and clouds affect tropospheric photochemistry through absorbing and/or scattering solar radiation that determines photolysis frequencies (J-values). There is synergy between the effects of clouds and absorbing aerosols when they are located in the same layer or clouds are located below the aerosols.

➤ Since the distribution and optical properties of clouds in current climate models have large uncertainties, understanding this synergistic impact is important for improving our understanding of the effect of aerosols in these models.

➤ In this study, we compare and contrast the radiative effects of aerosols [Martin et al., JGR 2003] and clouds [Liu et al., JGR 2006, 2009], and quantify their synergistic impact on key tropospheric oxidants (OH, ozone) in two global 3-D chemistry and transport models (GMI and GEOS-Chem CTMs). Preliminary results are presented.

Global Models

➤ Global Modeling Initiative (GMI) CTM (tropospheric full-chemistry version) driven by NASA's DAO (a.k.a GEOS1-DAS), GISS II', fvGCM, and GEOS4-DAS meteorological fields (4° × 5°); GEOS-Chem, a global 3-D model of tropospheric ozone-NOx-hydrocarbon chemistry coupled with aerosol chemistry, driven by GEOS5-DAS assimilated meteorological fields (2° × 2.5°).

➤ GMI: uses global 3-D monthly mean fields for 1996-1997 of dry aerosol mass (offline) from the GOCART model [Chin et al., 2000, 2002; Ginoux et al., 2001]. The fields include sulfate, size-resolved mineral dust and sea salt, hydrophobic and hydrophilic black carbon, as well as hydrophobic and hydrophilic organic carbon. Nitrate aerosol is not included.

➤ GEOS-Chem: coupled aerosol (including sulfate-nitrate-ammonium, carbonaceous aerosols, sea salt, and mineral dust) chemistry with ozone-NOx-hydrocarbon chemistry.

➤ Aerosol optical depth (AOD) calculated from the mass concentration, extinction efficiency, and particle mass density [Martin et al., JGR 2003]. An external mixture of the different aerosol types is assumed.

➤ Cloud optical depth (COD) & cloud fraction from met. fields.

➤ Photolysis rate calculation: Fast-J [Wild et al., 2000].

➤ GMI & GEOS-Chem model experiments:

- ✓ With aerosols, with clouds
- ✓ Without aerosols, with clouds
- ✓ With aerosols, without clouds
- ✓ Without aerosols, without clouds

Model Representations of the Vertical Coherence of Clouds

➤ LIN: Linear Assumption

$$\tau_c' = \tau_c \cdot f$$

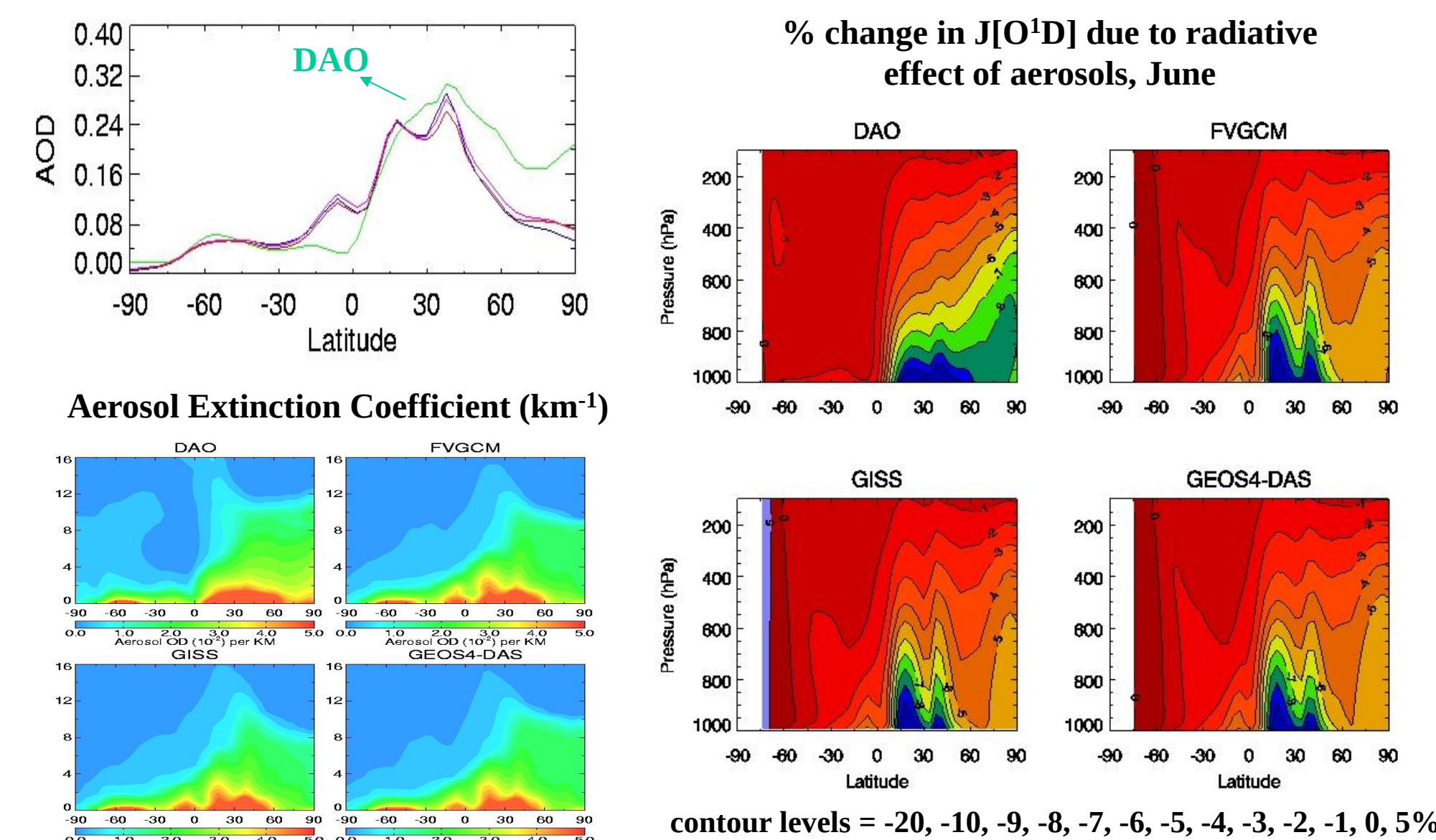
grid-scale OD in-cloud OD cloud fraction

➤ RAN: Approximate Random Overlap

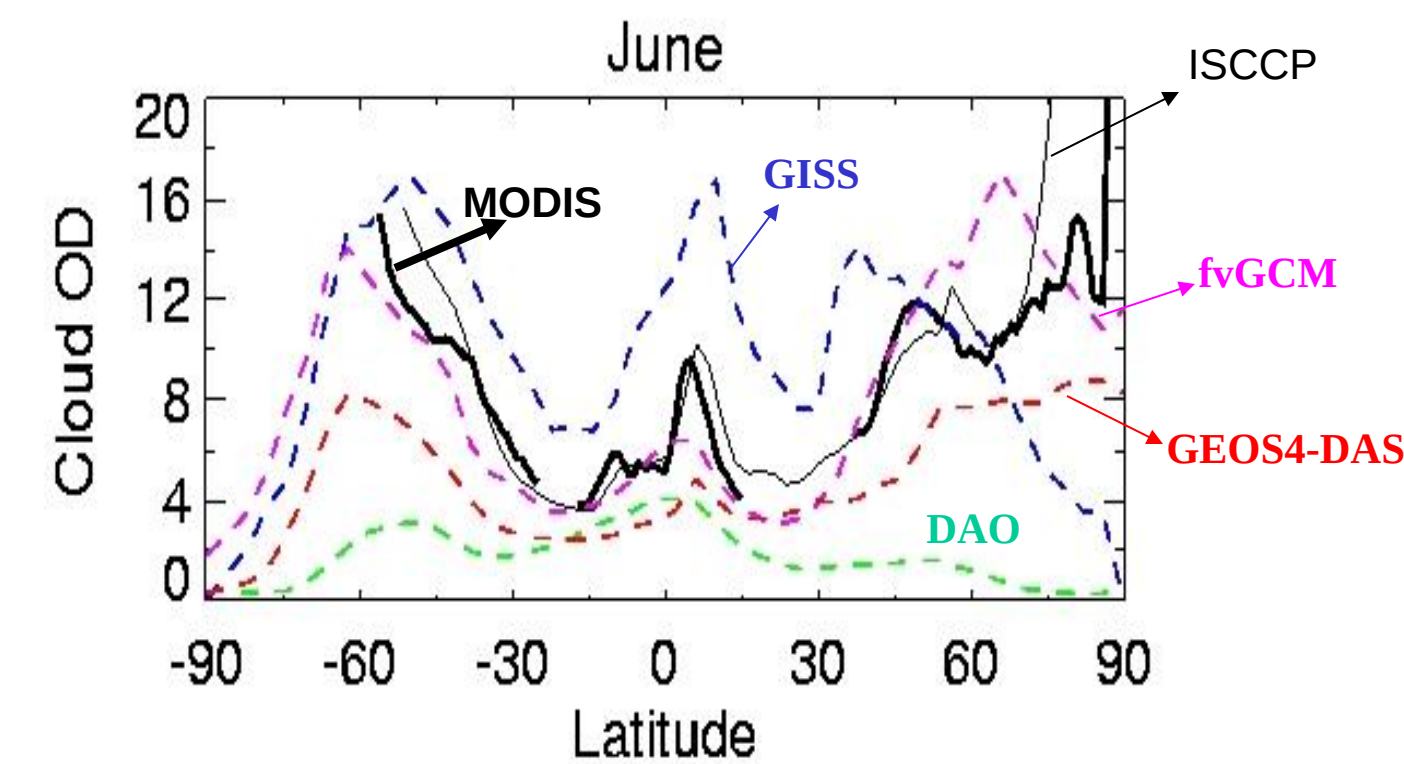
$$\tau_c' = \tau_c \cdot f^{3/2}$$

In terms of cloud effects on J[O¹D] and J[NO₂], on average, RAN and Maximum-Random Overlap differ by only ~2% throughout the troposphere [Liu et al., JGR 2006].

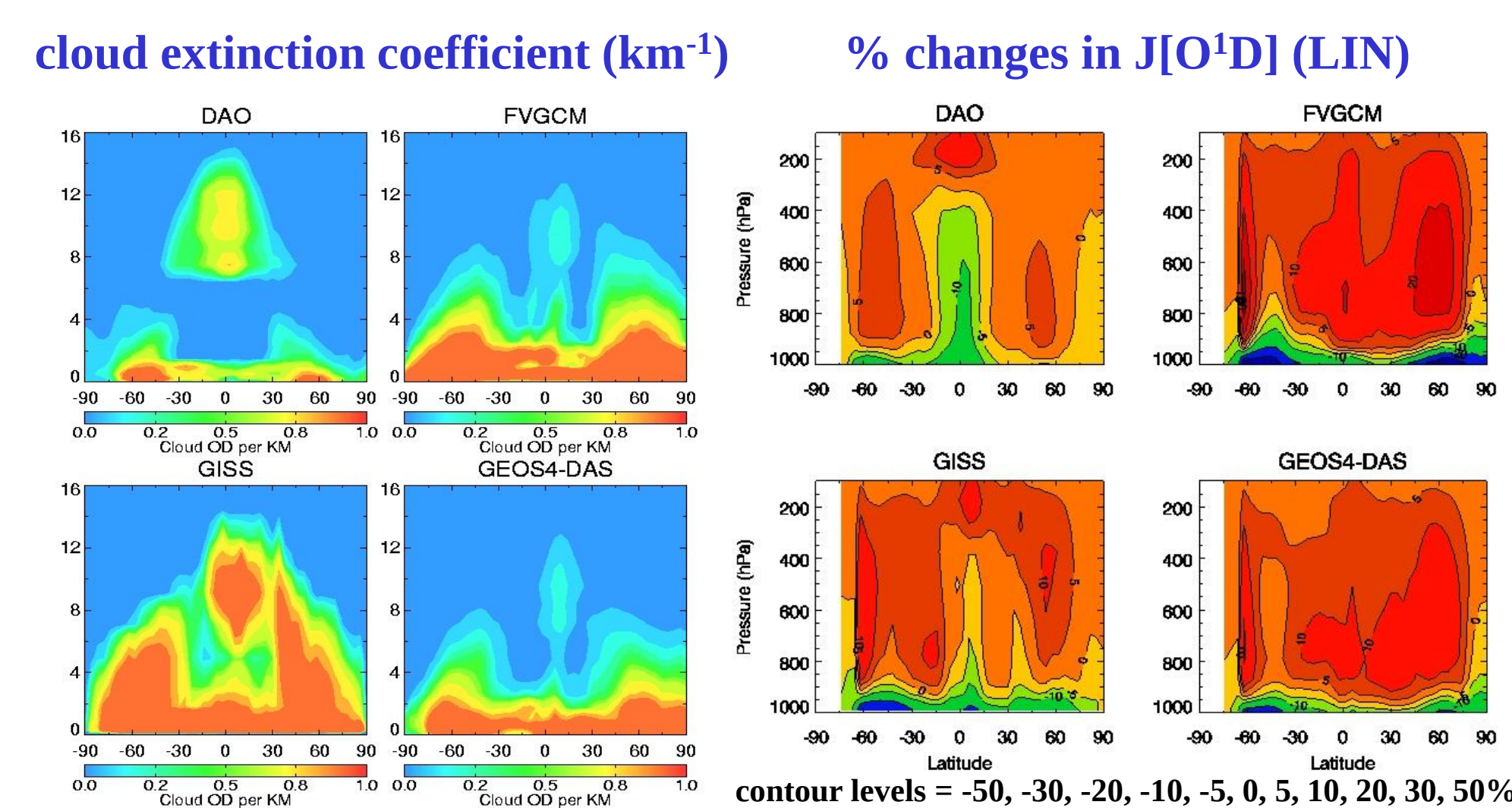
GMI: AOD and Effect on J[O¹D] (June)



GMI: Monthly Zonal Mean Cloud Optical Depth (June)

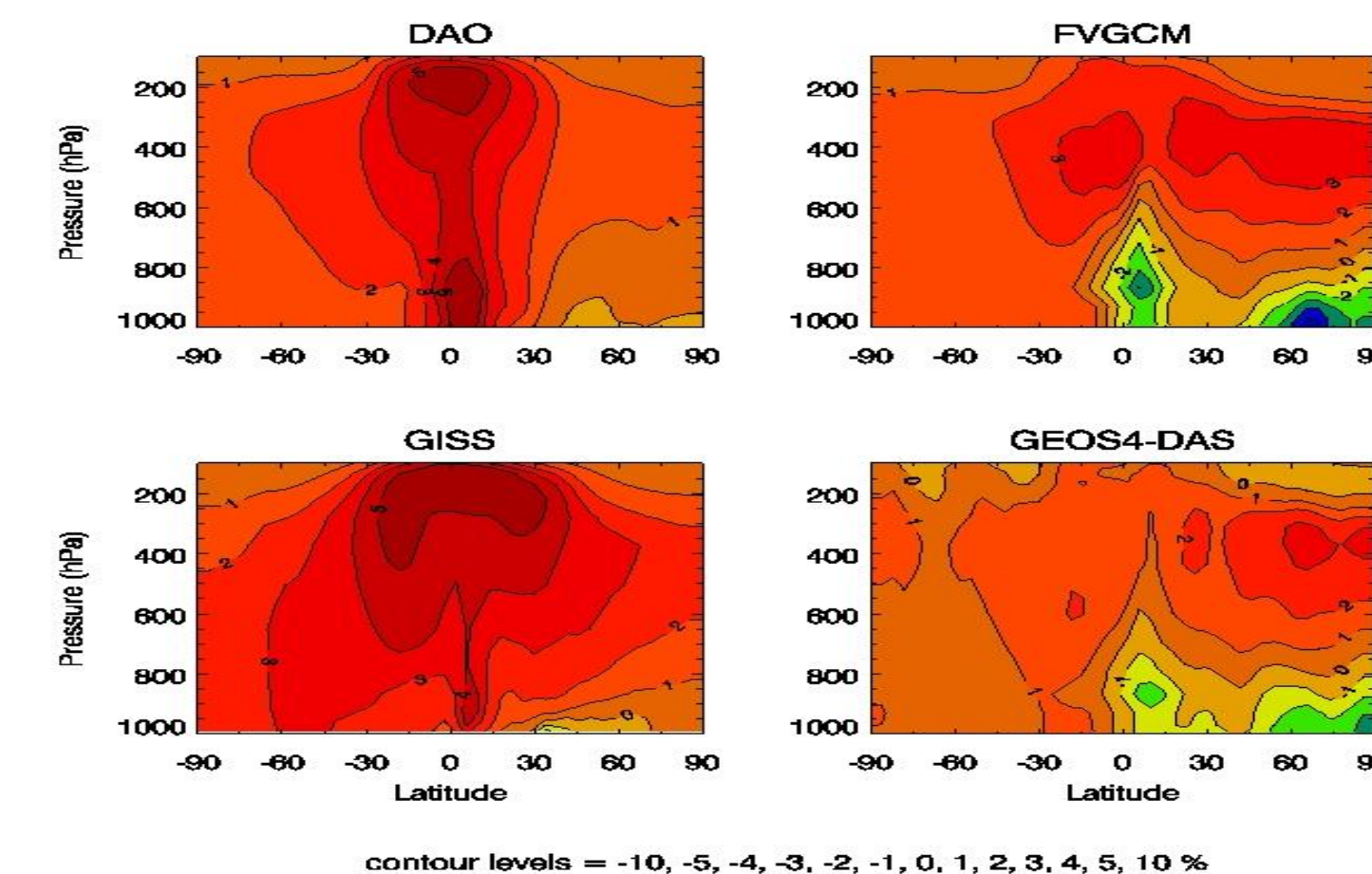


GMI: Cloud OD and Effect on J[O¹D] (June)



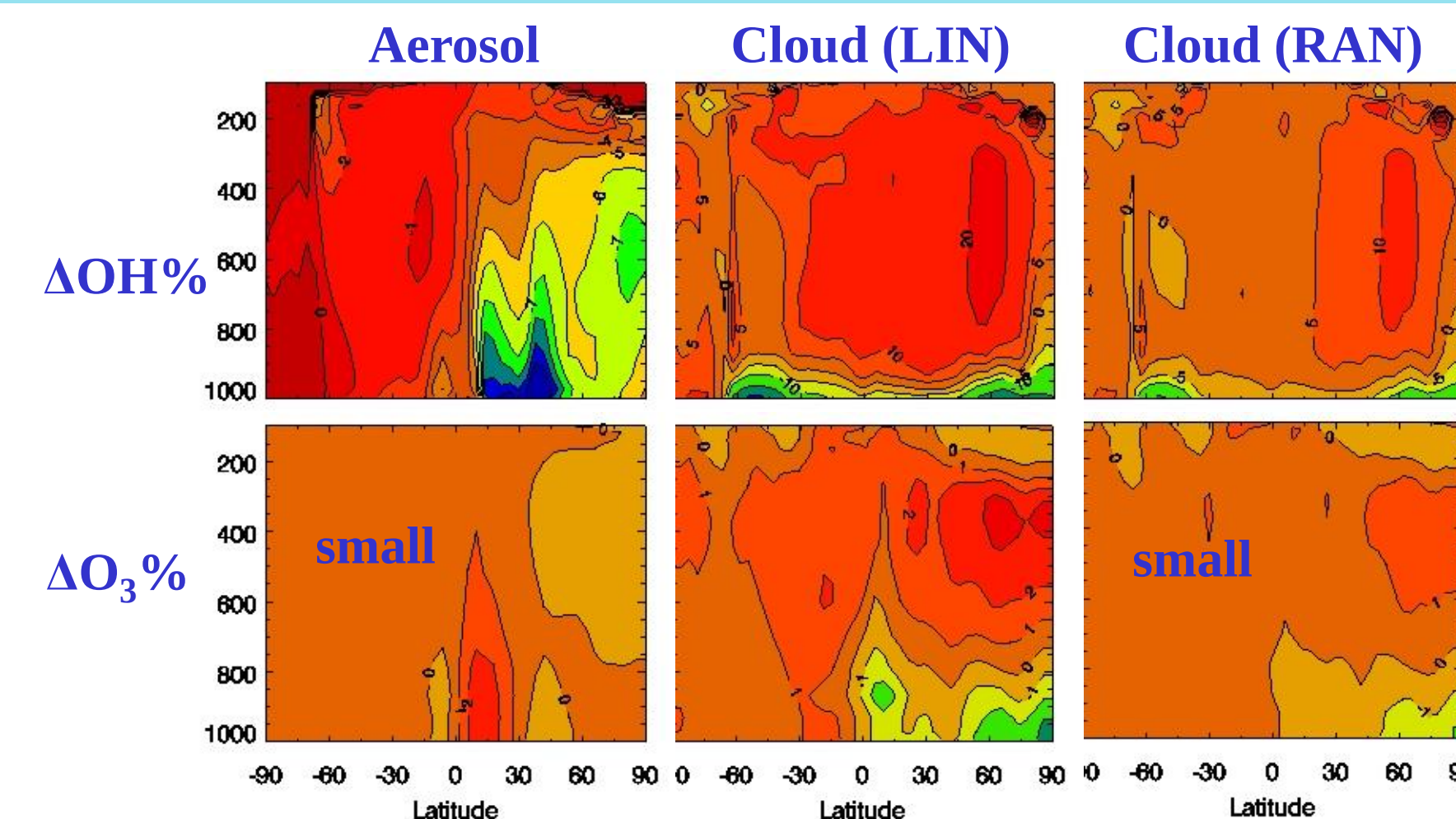
Tropical UT: optically much thinner in fvGCM & GEOS4-DAS

GMI: Radiative Effects of Clouds on Ozone (% changes in ozone, LIN, June)



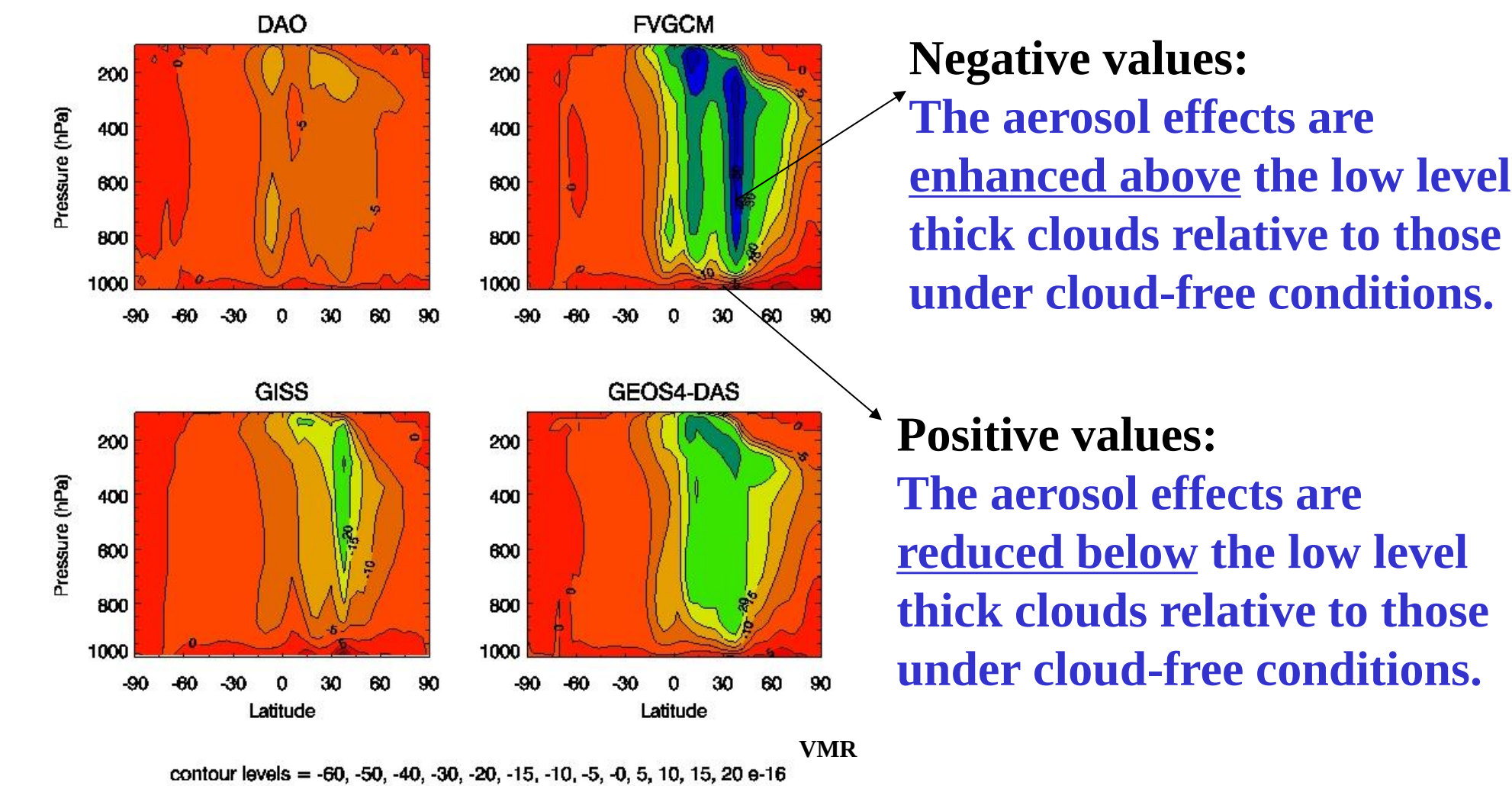
- Effects on tropical UT O₃: ~5% or less for all met./cloud fields; using RAN (vs LIN) for cloud overlap yields even lower effects.
- Sensitivity similar to results from GEOS-Chem driven by GEOS-3 [Liu et al., JGR 2006], but much less than those from MOZART-2 (~20-30%) [Tie et al., JGR 2003].

GMI / GEOS4-DAS: Radiative Effects of Aerosols vs Clouds on OH and O₃ (June)



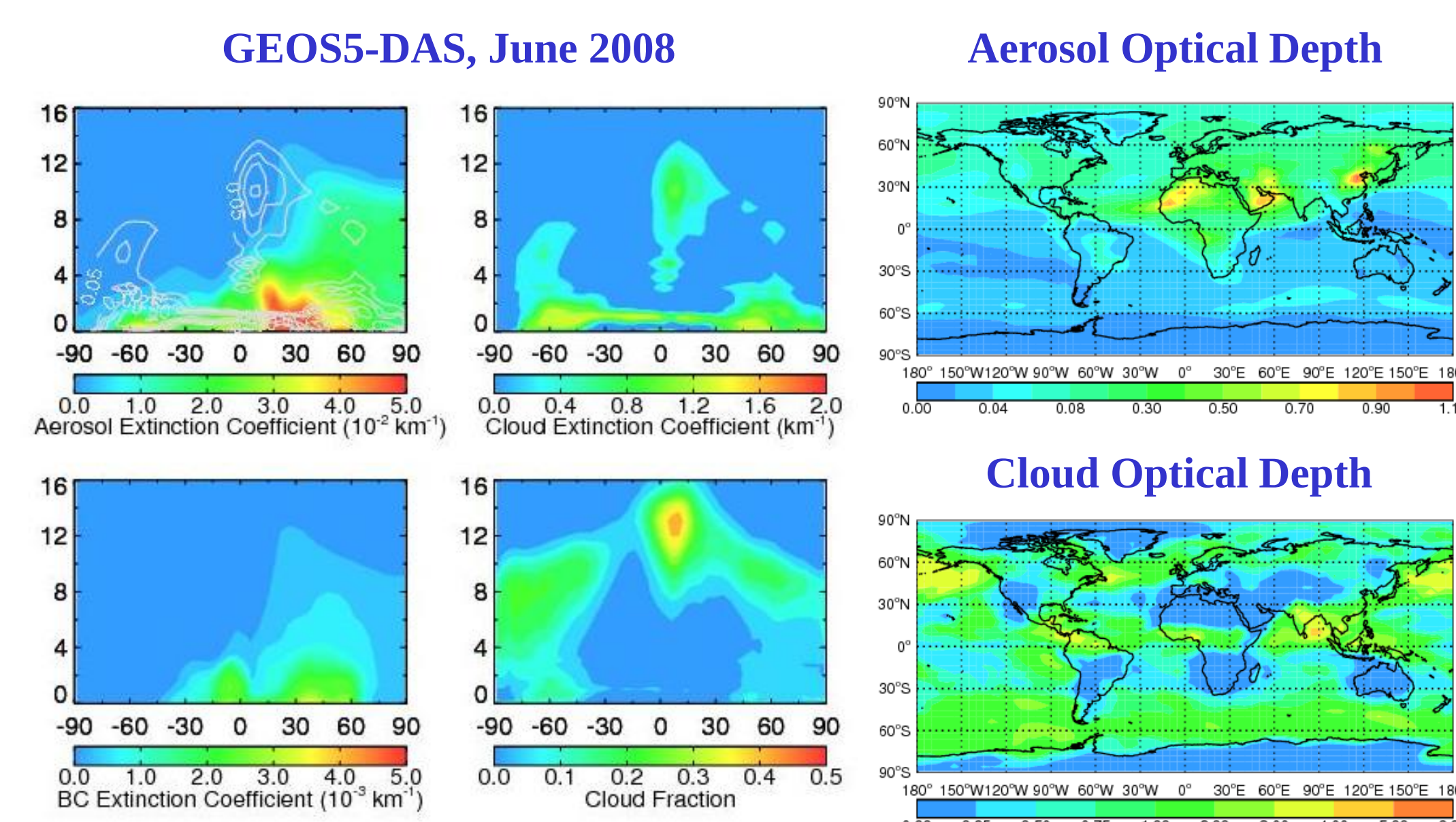
- Aerosol**
 - Decrease OH throughout troposphere;
 - Effects comparable to those of clouds near surface at NH mid-latitude;
 - Small effect on the global ozone burden.
- Cloud**
 - Enhance (reduce) OH above (below) low level thick clouds;
 - Vertical distributions (high clouds) are highly uncertain among models;
 - Small effect on the global ozone burden.

GMI: Effect of Clouds on Aerosol Radiative Impact on OH (June) (OH_{aero} - OH_{noaero})_{cloudy} - (OH_{aero} - OH_{noaero})_{clear}



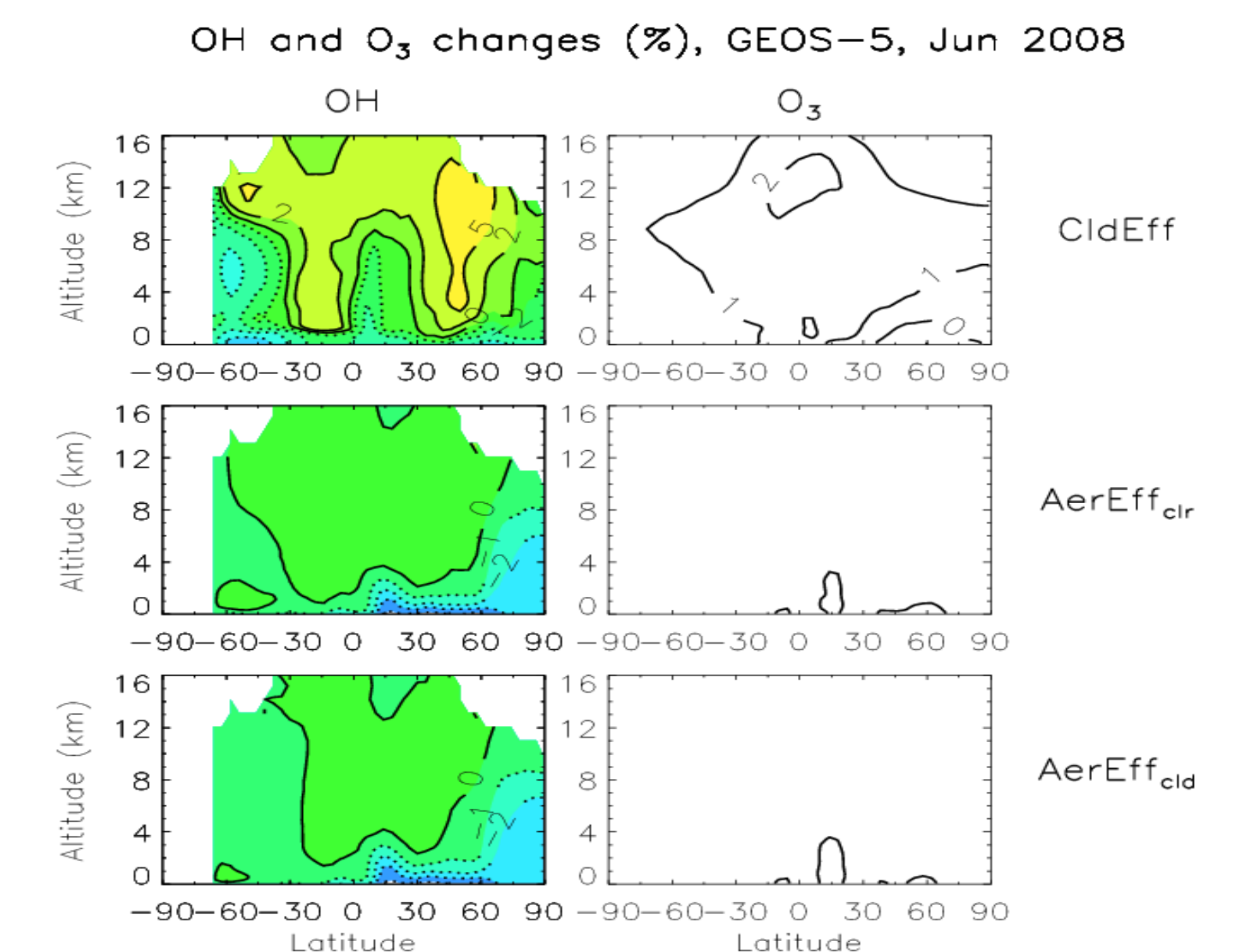
- Less synergy between cloud and (absorbing) aerosols in DAO/GISS is partly due to optically thicker clouds in UT.

GEOS-Chem / GEOS5-DAS: Aerosol and Cloud Optical Depths

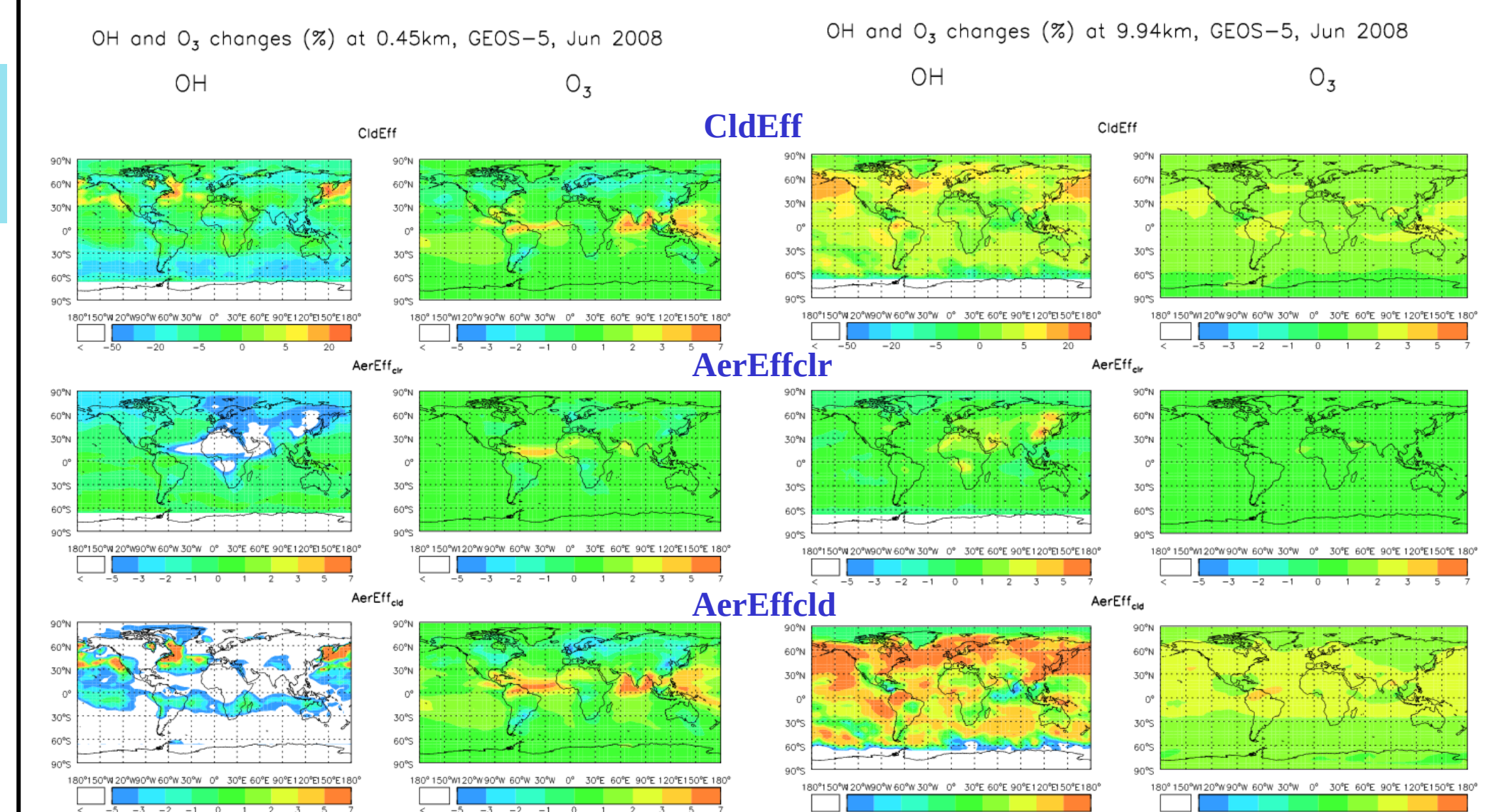


- Relative vertical & horizontal distributions of aerosols vs clouds. Left: Monthly zonal mean aerosol & cloud extinction coefficients, BC extinction coefficients, cloud fraction. Right: Monthly mean column aerosol & cloud ODs.

GEOS-Chem / GEOS5-DAS: Radiative Effects of Aerosols vs Clouds on OH and O₃ (June)



CldEff: effects of clouds
AerEffclr: effects of aerosols under clear sky conditions
AerEffcld: effects of aerosols under cloudy conditions



Summary and Conclusions

- Global 3-D model simulations indicate that both aerosols and clouds have modest radiative effects on the global ozone burden.
- While the radiative effects of aerosols on OH are much smaller than those of clouds in most of the free troposphere, their (negative) effects are comparable near the surface at the northern mid-latitudes.
- We showed with global 3-D model simulations that aerosol effects are enhanced (reduced) above (below) the low level thick clouds relative to those under cloud-free conditions.
- High clouds reduce cloud-aerosol synergistic effects.
- A regional analysis of cloud-aerosol synergistic effects is ongoing.

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