Improvement of Inorganic Aerosol Treatments in CESM/CAM5

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Introduction

Aerosol can affect the Earth's climate through aerosol-cloud-climate interactions. Aerosol size distribution, chemical composition, and number concentration are affected by various atmospheric processes such as nucleation, condensation, coagulation, thermodynamic equilibrium, and deposition. This study focuses on the improvement of condensation, new particle formation and inorganic aerosol thermodynamic equilibrium in the 5.1 version of Community Atmosphere Model (CAM5) of the Community Earth System Model (CESM). The aerosol module used in CAM5 is the 7-mode prognostic Modal Aerosol Model (MAM7). MAM7 uses the same accommodation coefficient (α=0.65) based on Adams and Seinfeld (2002) (AS02) for H_2SO_4, NH_3, and SOA (g) and does not include condensation for HNO_3. These treatments are improved in this work by adding condensation of HNO_3 and using different α values for different species following Zhang et al. (1998) (ZH98). There are three default nucleation parameterizations in CAM5/MAM7, the empirical power law of Wang et al. (2009) (WA09) for nucleation in the planetary boundary layer (PBL), the binary H_2SO_4-H_2O homogeneous nucleation of Vehkamaki et al. (2002) (as VE02), and ternary H_2SO_4-NH_3-H_2O homogeneous nucleation of Merikanto et al. (2007) (ME07) above PBL. Ions have been demonstrated as a significant source of new particles, especially in the upper troposphere and stratosphere. Therefore, an ion-mediated nucleation mechanism based on Yu (2010) (YU10) is incorporated into MAM7 to simulate new particle formation. MAM7 does not include aerosol thermodynamics. ISORROPIA II, a thermodynamic equilibrium model (Fountoukis and Nenes, 2007) (FN07) is thus incorporated into MAM7 to compute gas-aerosol equilibrium with a metastable assumption. As an initial test, only sulfate, ammonium, and nitrate are calculated in ISORROPIA II.

Model Configurations

Simulation period
- June, July, August (JJA), 2001

Grid resolution
- Horizontal: 0.9° × 1.25°   Vertical: 30 layers

Emissions
- Compiled based on several public available emission datasets for 2001 (Zhang et al., 2012)
- BVOC: MEGAN; Dust: Zender (2003); Seasalt: Martensson et al. (2003)

Chemistry options
- Gas-phase mechanism: 2005 version of the Carbon Bond Mechanism for Global Extension (CB05_GE) (Karamchandani et al., 2011)
- Aerosol-phase mechanism: 7-mode prognostic Modal Aerosol Model (MAM7) (Liu et al., 2012)
- Aqueous-phase chemistry: Barth et al. (2000)

Simulation design for inorganic aerosol improvements
- MVW: default condensation and nucleation parameterizations (ME07 and VE02 above PBL and WA09 in the PBL)
- MVW_con: same as MVW but with updated condensation module
- MYW: same as MVW_con but with ME07 and YU10 above PBL and WA09 in the PBL
- MYW_ISO: same as MYW but with ISORROPIA II for aerosol thermodynamics

Model Evaluation and Datasets

- Surface measurements of PM and precursors: CONUS (CASTNET, IMPROVE, STN), Europe (EMEP, AirBase, BDQA), and East Asia (MEP of China)
- New particle formation rates (J): land-, ship-, and aircraft-based measurements compiled by Yu et al. (2008)
- Satellite Data: Cloud Condensation Nuclei (CCN) and Cloud Fraction (CF) from MODIS; Cloud Droplet Number Concentration (CDNC) from MODIS (Bennartz, 2007)

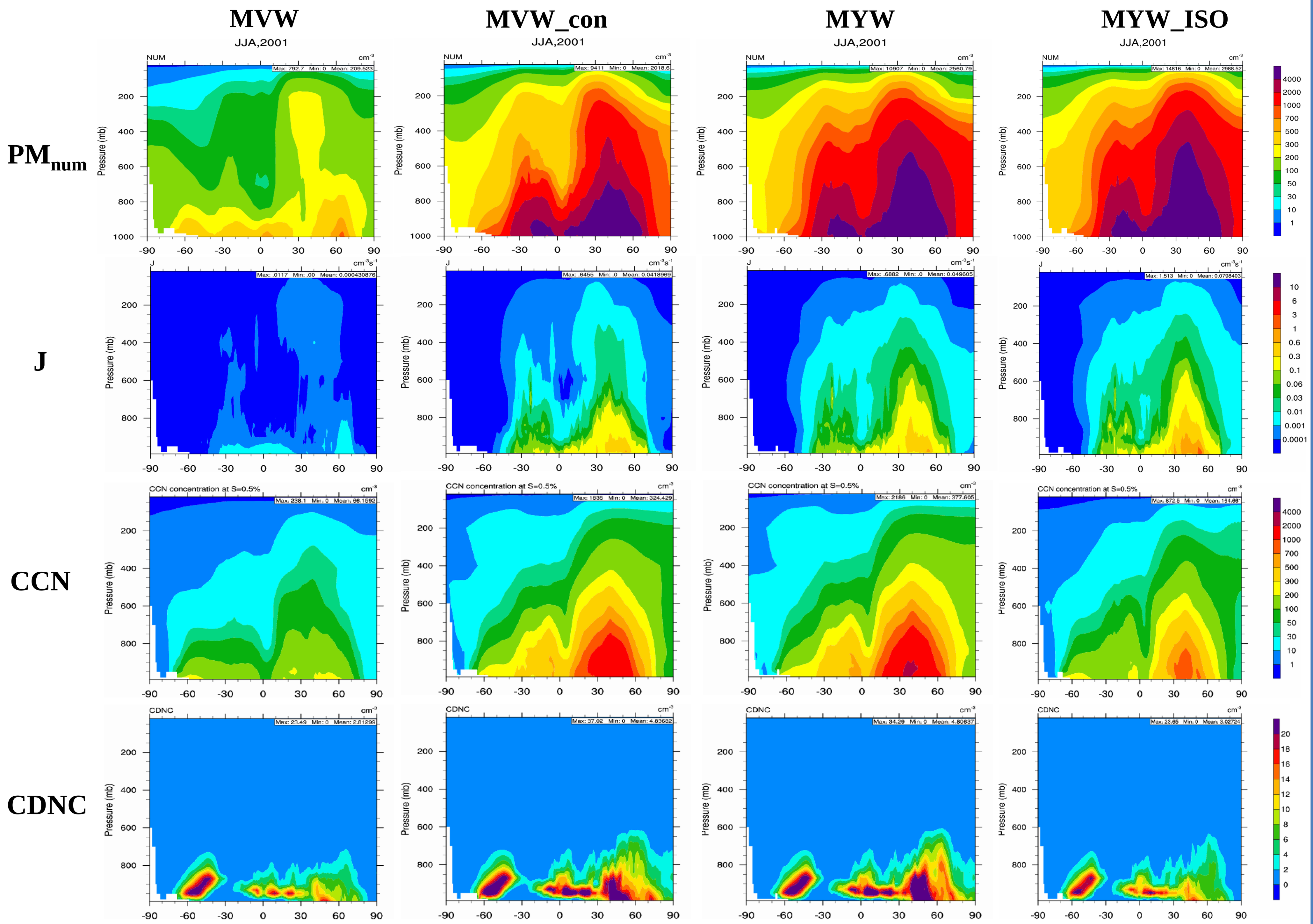
CASTNET: the Clean Air Status and Trends Network; IMPROVE: Interagency Monitoring of Protected Visual Environments; STN: Speciation Trends Network; EMEP: the European Monitoring and Evaluation Program; BDQA: Base de Données sur la Qualité de l'Air; AirBase: the European air quality database

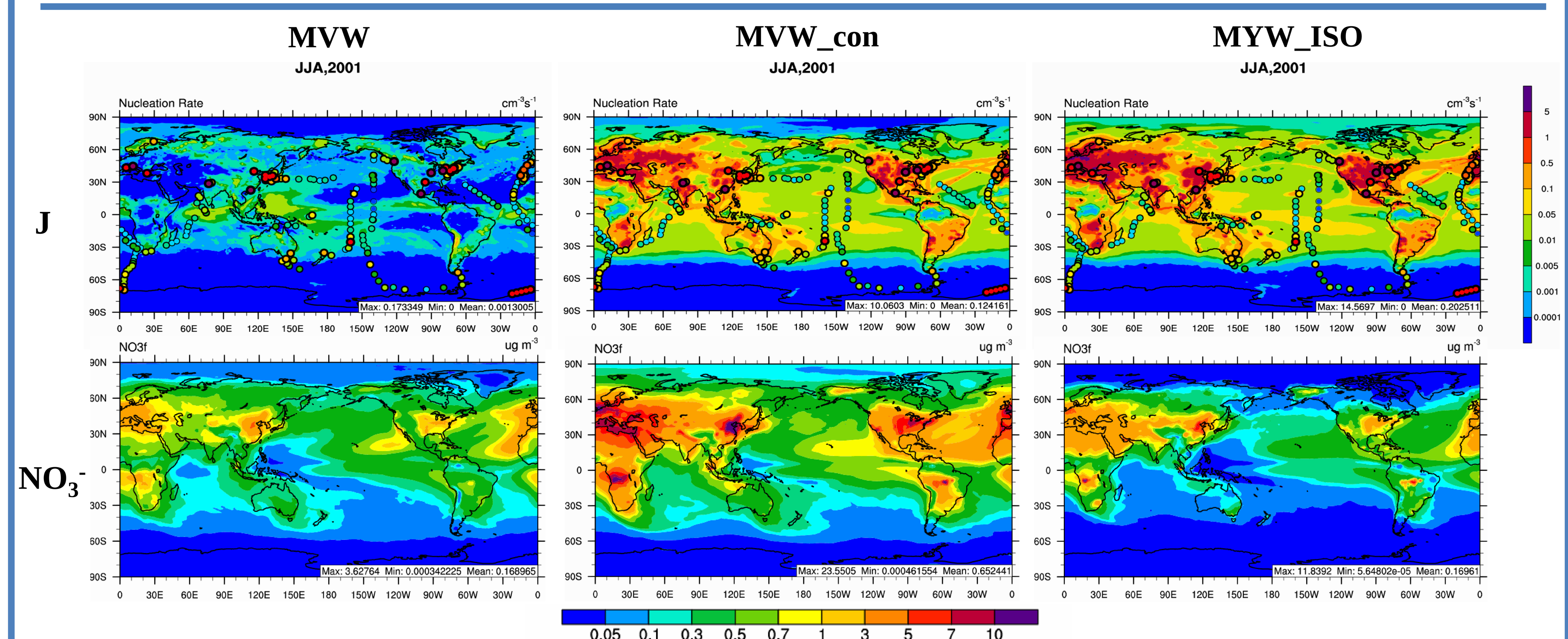
Parameterizations

	Condensation		Nucleation				Thermodynamics
	constant α	variable α	WA09	VE02	ME07	YU10	ISORROPIA II
Theory	Irreversible condensation	Irreversible condensation	Cluster activation	Classical binary homogeneous	Classical ternary homogeneous	Ion-mediated nucleation	Equilibrium partitioning
Input Variable	α = 0.65 for H_2SO_4, NH_3, SOA(g)	α_{H2SO4}=0.02 α_{NH3} = 0.097 α_{HNO3}=0.0024 $\alpha_{SOA(g)}$=0.65	C_{H2SO4}	C_{H2SO4}, T, RH	C_{H2SO4}, C_{NH3}, T, RH	C_{H2SO4}, T, RH, Q, S	TSO_4, TNH_4, TNO_3, P, T, RH

C_{H2SO4}, C_{NH3}: number concentration of sulfuric acid and NH_3; T: temperature; RH: relative humidity; Q: concentration of ion-pair; S: surface area of existing particles; TSO_4: total concentration of sulfate in Aitken and accumulation modes; TNH_4: total concentration of ammonium in Aitken and accumulation modes and NH_3 in the gas phase; TNO_3: total concentration of nitrate in Aitken and accumulation modes and HNO_3 in the gas phase; P: pressure; α: accommodation coefficient

Simulation comparison of different parameterizations





PM_{num}: total aerosol number concentration; J: Nucleation Rate; CCN: Cloud Condensation Nuclei; CDNC: Cloud Droplet Number Concentration

Performance Statistics

		Obs.	MVW NMB (%)	MVW NME (%)	MVW_con NMB (%)	MVW_con NME (%)	MYW NMB (%)	MYW NME (%)	MYW_ISO NMB (%)	MYW_ISO NME (%)
SO_2	CONUS	2.6	329.0	332.1	303.8	307.1	300.3	303.1	313.6	316.8
	Europe	5.5	129.8	183.8	133.7	190.1	105.4	166.8	136.5	189.3
	East Asia	20.5	-76.6	76.6	-76.5	76.5	-77.4	77.4	-75.1	75.1
NH_3	Europe	10.9	-82.5	83.3	-87.2	87.3	-89.1	89.1	-85.7	85.7
NO_2	East Asia	12.3	-89.3	89.3	-87.8	87.8	-88.2	88.2	-88.9	88.9
HNO_3	CONUS	1.9	12.7	43.3	-79.4	79.4	-80.7	80.7	-5.9	37.0
	Europe	0.5	244.1	255.3	-51.9	54.3	-56.2	56.9	160.7	167.1
SO_4^{2-}	CONUS	3.8	-8.0	24.5	7.4	25.3	12.6	25.7	6.5	26.7
	Europe	2.6	111.6	115.2	119.5	121.3	108.5	111.5	138.5	142.1
NH_4^+	CONUS	1.7	-29.9	36.9	12.2	54.3	16.4	55.5	-9.0	41.1
	Europe	1.1	37.9	39.9	130.5	133.3	130.2	132.6	87.2	89.6
NO_3^-	CONUS	0.5	13.7	88.9	598.3	645.4	627.3	671.4	68.4	136.2
	Europe	1.6	-20.5	33.7	252.4	252.4	243.5	243.5	17.9	32.9
$PM_{2.5}$	CONUS	9.5	-12.5	23.1	30.9	37.4	31.8	39.1	-3.6	20.7
	Europe	13.4	23.6	29.1	59.2	62.0	31.2	36.5	46.6	48.5
PM_{10}	Europe	25.4	139.1	139.5	125.1	125.3	125.3	125.4	180.0	180.0
	East Asia	17.8	-47.2	54.2	-23.9	58.2	-29.5	50.5	-39.9	53.5
J	Globe	0.55	-99.8	99.9	-47.6	85.5	-43.7	86.8	2.1	105.5
CCN	Globe	2.39×10^8	-81.9	82.3	-27.6	44.7	-11.4	45.5	-60.4	62.1
CDNC	Globe	110.5	-48.5	74.1	-6.4	93.6	-6.0	89.9	-39.9	73.9
CF	Globe	69.5%	-4.3	16.1	-2.3	16.4	-2.0	16.6	-4.1	15.7

SO_2 and NO_2 concentrations in East Asia are in ppb. J is in cm^{-3} s^{-1}. CCN is in cm^{-2}. CDNC is in cm^{-3}. CF is in %. All other concentrations are in μg m^{-3}.

Summary of Preliminary Results

- Treating condensation of HNO_3 and using variable accommodation coefficients in MAM7 give better results than default treatments in terms of J, CCN, CDNC, and CF.
- YU10 predicts nucleation rates ~59% higher than VE02 in the middle/upper troposphere and lower stratosphere, and slightly improves J, CCN, CDNC, and CF.
- With ISORROPIA II, the concentrations of nitrate and ammonium decrease, resulting in decreased PM_{num} in the accumulation mode and an increased PM_{num} in the Aitken mode due to the dominancy of the effect of ternary homogeneous nucleation over the effect of thermodynamic partitioning on total nitrate and total ammonium. MVW_ISO improves predictions of J, NH_4^+, and NO_3^-, but degrades performance of HNO_3 and SO_4^{2-} over Europe. It also improves predictions of CDNC over the remote ocean, especially in the North Pacific Ocean and North Atlantic Ocean, despite underprediction over land for ~80%.
- Compared with MVW, MYW_ISO with improved inorganic aerosol treatments improves model performance for some variables, e.g., HNO_3 over CONUS and Europe, NH_4^+ and $PM_{2.5}$ over CONUS, and J, CCN, and CDNC over globe. Large biases in some variables remain. For example, the large biases in simulated SO_2 and NH_3 over Europe and NO_2 over East Asia are likely due to uncertainties in their emissions, which lead to large biases in simulated HNO_3, SO_4^{2-}, NH_4^+, and $PM_{2.5}$. The large bias in simulated SO_2 over CONUS is caused by much higher simulated SO_2 at several sites in the forest areas. The underpredictions of CCN and CDNC may be related to inaccurate PM predictions and uncertainties in other cloud variables. Other factors contributing to these biases may include the use of a coarse grid resolution and uncertainties in observational data.

References

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