Simulating Marine Boundary Layer Clouds over the Eastern Pacific in a Regional Climate Model with Double-Moment Cloud Microphysics

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Abstract

A double-moment cloud microphysics scheme with a prognostic treatment of aerosols inside clouds has been implemented into the International Pacific Research Center (IPRC) Regional Atmospheric Model (iRAM) to simulate marine boundary layer clouds over the eastern Pacific and to study aerosol-cloud interactions including the aerosol indirect effect. This paper describes the new model system and presents a comparison of model results with observations. The results show that iRAM with the double-moment cloud microphysics scheme is able to reproduce the major features, including the geographical patterns and vertical distribution of the basic cloud parameters such as cloud droplet number, liquid water content or droplet effective radii over the eastern Pacific reasonably well. However, the model tends to underestimate cloud droplet number concentrations near the coastal regions strongly influenced by advection of continental aerosols and precursor gases. In addition, the average location of the stratocumulus deck off South America is shifted to the northwest compared with the satellite observations.

We apply the new model system to assess the indirect aerosol effect over the eastern Pacific by comparing a simulation with pre-industrial aerosol to an otherwise identical simulation with present-day aerosol. Resulting changes in the cloud droplet number concentration are particularly pronounced in Gulf of Mexico and along the Pacific coast lines with local changes up to 70 cm\(^{-3}\) (50% the present-day value). The modeled domain averaged 3-month (August-October) mean change in top of the atmosphere net cloud forcing over the ocean due to changes in the aerosol burden by anthropogenic activities is -1.6 W m\(^{-2}\).

1. Introduction
Cold sea surface temperatures (SSTs) in combination with warm, dry air aloft caused by large-scale subsidence in the subtropics lead to the formation of persistent marine stratocumulus decks over the eastern Pacific off the west coasts of North and South America [e.g. Albrecht et al., 1988]. These subtropical clouds impact the Earth's radiation budget and climate by reflecting incoming solar radiation [e.g. Randall et al., 1984]. Atmospheric aerosol plays a key role providing condensation nuclei required for cloud formation. Changes in atmospheric aerosol due to anthropogenic activities such as biomass burning, fossil fuel combustion or changes in land use are known to alter potentially cloud properties such as reflectivity or precipitation formation efficiency [e.g. Lohmann and Feichter, 2005]. Quantifying the indirect effects of aerosol is highly uncertain [Penner et al., 2006], and remains one of the largest uncertainties in our efforts to calculate radiative forcing [IPCC, 2007].

Tight coupling between atmosphere, ocean and land surfaces makes it difficult for numerical climate models to simulate marine stratocumulus decks over the eastern Pacific [Bretherton et al., 2004; Wang, 2004a,b]. Efforts in recent years have been devoted to understanding this system with the focus on realistically simulating the marine stratocumulus decks over the subtropical eastern Pacific. Several field experiments have been conducted to document the major features of the cloud decks and their links to the underlying ocean and larger-scale atmospheric circulation. The latest international regional experiment in the southeastern Pacific during October and November 2008 (VOCALS-REx, Wood and Mechoso [2008], http://www.eol.ucar.edu/projects/vocals/) was planned to measure and understand the coupled cloud-aerosol-ocean-land interaction processes. One of its hypotheses is that the polluted air is an important source of cloud condensation nuclei (CCN) and thus variability of the physico-chemical properties of anthropogenic aerosols could have a measurable impact upon the
formation of drizzle in stratocumulus clouds over the southeastern Pacific.

This modeling study aims at improving our knowledge of clouds, aerosols, and their interactions over the eastern Pacific in the context of a regional climate model. Specifically, we have implemented a double-moment cloud microphysics module [Phillips et al., 2007, 2008, 2009] into the Regional Atmospheric Model of the International Pacific Research Center (IPRC) iRAM [Wang et al., 2003; Wang et al., 2004a]. This allows us to calculate cloud droplet and ice crystal number concentrations from the predicted aerosol properties in addition to the mass mixing ratios of cloud and precipitation particles. iRAM has already been successfully applied to the study of eastern Pacific climate. In particular, with a single-moment cloud microphysics scheme [Wang, 2001], the model simulated reasonably well the stratocumulus deck and the cloud regime transition as well as the vertical cloud structure over the southeastern Pacific [Wang et al., 2004a,b]. However, because of the use of the single-moment bulk cloud microphysics scheme, the number concentrations of cloud droplets and ice crystals are both specified as constants. Knowledge of the cloud droplet and ice crystal number concentrations is essential to assess the indirect aerosol effect and study aerosol-cloud interactions. With the implementation of a double-moment cloud microphysics scheme into iRAM, we present an analysis of average microphysical cloud properties such as liquid water content and cloud droplet number concentrations from the results of a present-day model run (year 2006). Furthermore, we conducted a sensitivity experiment to assess the indirect aerosol effect over the eastern Pacific with special focus on the marine stratocumulus decks.

Section 2 describes the regional climate model iRAM, the double-moment cloud microphysics scheme, and the model setup. Modifications of the double-moment cloud scheme adopted for implementation into iRAM are also discussed. This is followed by a brief discussion
of the average microphysical cloud properties of the marine stratocumulus decks over the eastern Pacific in section 3. A comparison of model results from the control experiment with observations is presented in section 4. Section 5 shows results of the sensitivity of clouds over the eastern Pacific to changes in the atmospheric aerosol burden. Section 6 ends with a summary and conclusions.

2. Model and Model Simulations

2.1. The Regional Atmospheric Model – iRAM

We use the International Pacific Research Center (IPRC) Regional Atmospheric Model (iRAM) version 1.2 [Wang et al., 2004a] to study the marine boundary layer (MBL) clouds over the eastern Pacific. iRAM is based on the hydrostatic primitive equations in a vertical σ-coordinate. All simulations presented here were conducted at a horizontal resolution of 0.5° x 0.5° with 28 non-equidistant vertical layers from the surface up to about 10 hPa (~30 km), with 10 levels below 800 hPa. The model domain covers the geographical region 150°W-30°W and 40°S-40°N. The initial and lateral boundary conditions for the model integration are obtained from the National Centers for Environmental Prediction (NCEP) final analysis (FNL). The FNL data with a horizontal resolution of 1° x 1° and 26 vertical pressure levels at 6-h time intervals [Kalnay et al., 1996] are interpolated linearly to the model grid and time. Sea surface temperatures (SSTs) are prescribed according to daily mean satellite observations from the Advanced Very High Resolution Radiometer (AVHRR) and the Advanced Microwave Scanning Radiometer (AMSR) provided by the National Oceanic and Atmospheric Administration.
(NOAA) [Reynolds et al., 2007]. The prognostic model variables are nudged to the NCEP FNL analysis data within a 10° buffer zone along the lateral boundaries. This buffer zone is not taken into account when analyzing the model results. We calculated an ensemble of 10 model runs initializing the model at 0000 UTC on 18 to 27 July 2006 and continuously integrating through 31 October 2006. The 10 model runs are then averaged to reduce statistical noise introduced by, for instance, differences in convection due to slightly different initial conditions. The initial 5 to 14-day period is used to spin up the model physics and excluded from our analysis, which focuses on August-September-October (ASO) when the stratocumulus deck off the west coast of South America is most persistent. Concurrent with the minimum SSTs in this region, the marine stratocumulus clouds reach their maximum coverage [Klein and Hartmann, 1993]. We chose the year 2006 to comply with the pilot modeling study of PreVOCa within the CLIVAR project VAMOS Ocean-Cloud-Atmosphere-Land-Study (VOCALS). The VOCALS project focuses on the southeast Pacific climate system. An extensive survey of clouds, aerosols and meteorological parameters among many other parameters in October and November 2008 collected data well suited for upcoming model evaluations of the stratocumulus deck off the west coast of South America.

Cloud microphysics are calculated by a double-moment cloud microphysics scheme with a prognostic treatment of 6 aerosols species inside clouds [Phillips et al., 2007, 2008, 2009; see section 2.2], which replaces the original single-moment cloud microphysics module of iRAM [Wang, 2001]. The cloud microphysics module is coupled to the radiation scheme and provides effective radii of cloud droplets and ice crystals as well as the liquid water and ice content as input for the radiative transfer calculations. The radiation scheme is based on the radiation package of Edwards and Slingo [1996] with improvements by Sun and Rikus [1999]. It considers
four spectral bands in the solar spectral range and seven spectral bands in the thermal spectral range. Cloud cover (CC) is diagnosed from cloud liquid water/ice content and ambient relative humidity following Xu and Randall [1996]:

\[
CC = RH^{0.25} \cdot \left(1 - \exp\left(-\frac{100 \cdot (q_l + q_i)}{(1 - RH) \cdot q_s^{0.89}}\right)\right)
\]

(1)

where RH denotes relative humidity, \(q_l\) and \(q_i\) the cloud liquid water and cloud ice mass mixing ratios, respectively, and \(q_s\) the saturation vapor mixing ratio.

Sub-grid scale convection including shallow, mid, and deep convection is parameterized following Tiedtke [1989] with modifications by Gregory et al. [2000]. Cloud water and cloud ice detrained at the cloud tops are considered as an additional source of cloud water/ice used by the grid-scale cloud microphysics [Wang et al., 2003]. As the convection scheme provides no information on the numbers of detrained cloud particles, we assume that detrained cloud water and cloud ice increase the size of pre-existing cloud particles but do not increase particle numbers.

For additional details on iRAM, we refer to Wang [2001], Wang et al. [2004a] and the literature cited therein.

2.2. Double-Moment Cloud Microphysics

We replaced the original single-moment cloud microphysics module [Wang, 2001] used in iRAM by a double-moment cloud microphysics scheme with a prognostic aerosol component inside clouds [Phillips et al., 2007, 2008, 2009]. In addition to the mass mixing ratios of water
vapor, cloud liquid water, cloud ice, rain, snow, and graupel already calculated by the single-moment scheme, the new double-moment cloud microphysics scheme also considers the number mixing ratios of cloud droplets and ice crystals. The sizes of cloud and precipitation particles are assumed to obey gamma distributions. The scheme predicts diffusional growth of cloud particles and precipitation explicitly with a linearized supersaturation scheme from the modeled updraft and properties of cloud liquid and water vapor. The predicted supersaturation is also applied to calculate the activation of aerosol particles at cloud base or inside the cloud when supersaturation becomes high enough. The double-moment cloud microphysics scheme includes primary and secondary ice nucleation [Hallet and Mossop, 1974] as well as homogeneous freezing of aerosols and cloud droplets. Heterogeneous ice nucleation by insoluble aerosol species (mineral dust, black carbon, hydrophobic particulate organic matter) is calculated by an empirical parameterization relating the ice nuclei activity of an insoluble aerosol species to the total surface area that is interstitial and immersed in cloud liquid. Ice nuclei are treated prognostically inside the clouds accounting for scavenging of unactivated ice nucleating particles [Phillips et al., 2008]. The heterogeneous ice nucleation scheme is based on field measurements [Phillips et al., 2008] and has been evaluated with laboratory and field observations by Phillips et al. [2008] and by Eidhammer et al. [2009]. Homogeneous freezing of super-cooled cloud droplets occurs at temperatures of -36°C using a parameterization of the fraction of supercooled droplets that evaporate during freezing, as described by Phillips et al. [2007]. Activation of droplets is treated with a scheme by Ming et al. [2006] at cloud-base and with the predicted supersaturation in-cloud, assuming κ-Köhler theory [Petters and Kreidenweis, 2007], as outlined by Phillips et al. [2009]. In the present paper for the first time, the assumption of internal mixing has been extended to include all soluble aerosol species, as detailed below.
Some processes in the double-moment cloud microphysics scheme have been modified for application in iRAM. We refer to Phillips et al. [2007, 2008, 2009] for more information on all other cloud microphysical processes calculated by the double-moment cloud microphysics scheme and on its semi-prognostic (in-cloud) aerosol component, with six chemical species of aerosol.

**Aerosols**

Following Phillips et al. [2008, 2009] we consider the six aerosol species: sulfate (SO₄), sea salt, soluble and insoluble particulate organic matter (POM), mineral dust, and black carbon (BC) with the main modification being a more complete representation of internal aerosol mixtures. Mineral dust and sea salt are assumed to be externally mixed, whereas POM, BC and SO₄ are assumed to be internally mixed particles. In addition, we consider a pure SO₄ mode over the oceans, in which sulfate is assumed to be fully neutralized by ammonium forming ammonium sulfate. The aerosol size distributions are represented by a sectional discretization with 20 size bins covering the size range from 0.01 to 1 µm for sulfate and carbonaceous particles and from 0.05 to 10 µm for sea salt and mineral dust particles. The option of the feedback from clouds onto aerosol fields in the cloud-free environment described by Phillips et al. [2009] was not invoked, but their prognostic treatment of aerosol inside clouds was included, as well as size distributions of aerosol. With extra prognostic variables for each aerosol species described by Phillips et al. [2009], the model keeps track of aerosol particles in each species that are activated, immersed in-cloud particles or interstitial, treating the most relevant sources and sinks. These processes are activation of interstitial aerosol, accretion of cloud particles by
precipitation, evaporation or sublimation of cloud particles or any hydrometeors, and nucleation of cloud liquid or cloud ice (deposition, conventional contact-freezing). This simplified approach represents physical processes that modify aerosol fields on time-scales of less than about a day but omits slower processes such as aerosol microphysics changing the aerosol size-distribution on the timescale of days to weeks or aerosol dry deposition. This assumes that these slow processes are already implicitly included in the aerosol climatology [Phillips et al., 2009].

The mass mixing ratios of the six aerosol compounds are prescribed by 3-dimensional climatological monthly mean fields. Aerosols can be modified by clouds and advected in the model but mass mixing ratios are restored to the prescribed climatology once the cloud evaporates completely. A cloud is considered to be evaporated completely if the sum of cloud liquid and cloud ice is zero or (in case of warm clouds) the number of droplets decreases below 0.1 cm$^{-3}$. The mass mixing ratios of sulfate particles are taken from simulations with a global tropospheric aerosol chemical transport model with a horizontal resolution of 2.8° x 2.8° with 34 vertical levels below 10 hPa. The chemical transport model is coupled with a general circulation model (GCM), which is nudged using reanalysis data [Tanaka et al., 2003]. For the mass mixing ratios of carbonaceous particles and mineral dust, geographical distributions are taken from the GCM results obtained by Takemura et al. [2000]. The horizontal resolution of these data is 1.125° x 1.125° with 18 vertical levels between the surface and 10 hPa. The total mass of carbonaceous particles given by Takemura et al. [2000] has been split into black carbon, soluble and insoluble organics according to the average atmospheric burdens of these aerosol compounds calculated by global model simulation with detailed aerosol microphysics [Lauer et al., 2007]. The geographical distribution of sea salt is also taken from global model simulations by Lauer et al. [2007]. These average sea salt mass concentrations are scaled to match observations which
typically find about 10-20 µg m$^{-3}$ over the tropical and subtropical eastern Pacific near the surface [McNaughton, 2008]. Furthermore, the vertical sea salt profiles have been adjusted to the constant mixing ratio frequently observed within the marine boundary layer by averaging the sea salt concentrations in the lowermost 2 km. Sea salt mass concentrations are assumed to decrease exponentially with height above the MBL.

We use measured log-normal size-distributions from McNaughton et al. [2008] to calculate particle number concentrations corresponding to the prescribed aerosol mass profiles. The number concentrations are then scaled to match the total mass given by the prescribed 3-dimensional aerosol climatology. Table 1 summarizes the parameters for the size-distributions used in our model experiments. More details about aerosol parameters are given by Phillips et al. [2009].

Figure 1 shows a comparison of vertical mean profiles for sulfate, mineral dust and sea salt from the aerosol model climatology used in this study with measurements from the Intercontinental Chemical Transport Experiment (INTEX) [Molina et al., 2009; Singh et al., 2009]. The INTEX-B measurements cover the geographical regions 88°W-98°W, 20°N-30°N taken in April and May 2006 over the Gulf of Mexico and 130°W-140°W, 20°N-30°N taken in March 2006 over the Pacific Ocean near Hawaii [McNaughton et al., 2009]. Sulfate mass concentrations from the model climatology are in good agreement with the observations over the eastern North Pacific and within the MBL over the Gulf of Mexico. Sulfate aerosol is underestimated in the middle and upper troposphere over the Gulf of Mexico. Enhanced sulfate and especially dust observed in the 2-6 km altitude range is pollution outflow from Mexico City (2000 meters above sea level), which is not captured by the model aerosol climatology. This might result in an underestimation of cloud droplets in the middle troposphere over the Gulf of
Mexico. Average sea salt concentrations are in the range of observations in both cases but measurements of dry sea salt mass are about a factor of two higher on average. Because of the large size of sea salt particles, this underestimation of sea salt mass translates only in a small underestimation of the number of potential cloud condensation nuclei suggesting that the scaling of the original model sea salt profiles (see previous paragraph) provides a reasonable estimate of sea salt aerosols for the purpose of this study. Mineral dust concentrations in the middle and upper troposphere are overestimated by one order of magnitude by the model climatology, which might result in a too high ice crystal number concentration. We do not expect mineral dust to play an important role in the lower troposphere, as no ice clouds exist in the tropical and adjacent subtropical regions over the ocean simulated in this study. The vertical profiles for mineral dust mass concentration from the model climatology show less decrease with altitude than the observations, particularly over the Gulf of Mexico. This suggests that removal of mineral dust particles, presumably by gravitational settling and wet deposition, is too weak in the GCM that calculated the mineral dust climatology. Figure 1 also shows total number of particles > 10 nm calculated from the aerosol mass concentrations given by the model climatology and the prescribed size-distributions (Tab. 1) in comparison with INTEX-B measurements. Total particle numbers are in good agreement over the eastern North Pacific in the middle and lower troposphere up to about 6 km but overestimated above. The high number concentrations from the model climatology in the upper troposphere are mainly related to small sulfate particles. As sulfate mass from the model climatology is in good agreement, this overestimation might be caused by assuming too small particles when calculating particle number from sulfate mass compared with the INTEX-B data. We do not expect this to be a problem for the calculation of cloud droplet numbers because these very small particles (10-30 nm) tend to be activated only in
deep convective updrafts with speeds of several m/s. Such deep convective clouds do not play a major role in most of the tropical and adjacent subtropical regions over the ocean simulated in this study.

Over the Gulf of Mexico we find good agreement between total particle number in the upper troposphere above 6 km, but an underestimation of particle number in the lower and middle troposphere compared with observations. This underestimation is particularly pronounced in the altitude range 2-6 km, which is influenced by pollution outflow from Mexico City. This finding is consistent with the underestimation of sulfate mass in this altitude range and seems to confirm that cloud droplets in the middle troposphere over the Gulf of Mexico could be underestimated in the model.

Alternatively to the model climatology, the mass concentration of sea salt can be calculated prognostically using emission fluxes calculated from the modeled 10 m wind speed [Monahan, 1986]. Dry deposition of sea salt particles at the surface and gravitational settling of sea salt particles are then calculated following Binkowski and Shankar [1995] using the formulation of the aerodynamic resistance by Kerkweg et al. [2006]. The average sea salt mass concentrations calculated by the prognostic scheme are within the range of observations over the tropical and subtropical eastern Pacific near the surface [McNaughton, 2008]. When calculated prognostically, sea salt introduces additional noise to our sensitivity studies. As a result, sea salt has been prescribed from the 3-dimensional aerosol climatology in all model runs discussed in this study.

*Aerosol Activation*

The activation of aerosol particles in warm clouds uses the scheme by Ming et al. [2006]
and that by Phillips et al. [2008] for ice particles, as described in detail by Phillips et al. [2009]. There are several prognostic variables for each aerosol species (e.g. the number mixing ratio of aerosols lost by being activated as droplets) so that the components of each aerosol species that are interstitial and immersed in cloud-liquid are predicted. Another key modification described by Phillips et al. [2009] is that an algorithm distinguishes between in-cloud and cloud-base grid-points, allowing more accurate treatment of cloud-base droplet activation with the scheme by Ming et al. [2006] and in-cloud activation using (κ-)Köhler theory from the resolved supersaturation predicted with the linearized scheme of Phillips et al. [2007]. The look-up table for cloud-base droplet nucleation relating aerosol mass concentration and updraft to the number of CCN from Phillips et al. [2009] has been replaced by this new treatment of aerosol activation. It allows more accurate calculation of cloud-base droplet activation of all of our assumed internal aerosol mixtures. Internal mixtures of soluble aerosol species are treated as follows and is described in the present paper for the first time. Critical droplet diameters and supersaturations as well as the equilibrium supersaturations of the droplets are obtained from the κ-Köhler theory, which describes the relationship between particle dry diameter and the number of cloud condensation nuclei (CCN) using the hygroscopicity parameter κ [Petters and Kreidenweis, 2007]. The multicomponent hygroscopicity parameter for internally mixed aerosols is calculated by averaging the hygroscopicity parameters of all components, with values from Petters and Kreidenweis [2007], weighted by their volume fractions in the mixture.

The number of activated aerosols depends nonlinearly on the maximum supersaturation. The grid spacing and the time step of large scale models do not allow processes that drive local cooling (e.g., vertical updraft) and thus determine maximum supersaturation to be resolved. Using the large-scale average vertical motion to obtain local cooling rates, which determine
aerosol activation, would result in a strong underestimation of activated cloud droplets [Ghan et al., 1997]. Thus, a parameterization for the sub-grid scale vertical velocity is needed to calculate the number of activated aerosols in large-scale models. Following Lohmann et al. [1999], we calculate the vertical velocity $w$ used in the nucleation parameterization as the sum of the grid mean vertical velocity $\bar{w}$ and its sub-grid scale variance $w'$ multiplied by a scaling factor $c$. The subgrid-scale variance of the vertical velocity $w'$ is calculated from the root-mean-square of the turbulent kinetic energy ($TKE$), the scaling factor $c$ is set to 0.7 [Lohmann et al., 1999]:

$$w = \bar{w} + w' = \bar{w} + c \cdot \sqrt{TKE} \quad \text{(2)}$$

The new aerosol activation scheme allows us to switch between internally mixed aerosols and the widely used assumption of external mixtures. Furthermore, additional aerosol compounds can be included easily.

**Homogeneous Aerosol Freezing**

In case of temperatures below 235 K and exceeding a size and temperature dependent critical supersaturation with respect to ice, homogeneous aerosol freezing (freezing of liquid aerosols) is assumed to occur instantaneously [Phillips et al., 2007]. A look-up table for the critical supersaturation is calculated off-line for different particle sizes and hygroscopicity parameters $\kappa$ following Koop et al. [2000] using the formulation of water activity by Petters and Kreidenweis [2007]. This look-up table, similar to that described by Phillips et al. [2009], replaces the one previously used in Phillips et al. [2007] and covers wider size ranges of aerosol particles as well as additional chemical species. The curvature/surface tension (Kelvin) effect
that becomes important at the small aerosol sizes < 0.05 μm is now treated. In addition, the new look-up table can be used for both internally and externally mixed aerosol particles.

**Autoconversion of Cloud Droplets to Precipitation**

The autoconversion of cloud droplets to rain water is parameterized after Liu et al. [2007]. This parameterization does not make the common assumption that the number autoconversion rate is linearly proportional to the mass autoconversion rate, suggesting more accurate results when assessing the indirect aerosol effect [Liu et al., 2007]. Liu et al. [2007] analytically relate the number autoconversion rate to the liquid water content, droplet number concentration, and relative dispersion of the cloud droplet size distribution. This formulation replaces the autoconversion rate described by Phillips et al. [2007], who incorporated a dependency on predicted droplet mean size and spectral width, into an earlier parameterization by Lin et al. [1983]. Alternatively, we also implemented the autoconversion parameterization of Khairoutdinov and Kogan [2000], which is used by Phillips et al. [2009]. Unless otherwise noted, all results shown in this study have been calculated using the autoconversion scheme by Liu et al. [2007] resulting in an improved agreement for our regional model with a 50-km resolution between modeled and observed cloud liquid water path and cloud cover (section 4.1 and 4.2).

3. Modeled Average Microphysical Cloud Properties

Figure 2 shows a zonal cross section along 10°S through the center of the modeled
stratocumulus deck. Shown are the August-October 2006 averages of cloud liquid water content and cloud droplet number concentration. The quantities depicted are conditionally averaged over cloudy grid cells only using a threshold for minimum cloud liquid water content of 0.025 g kg\(^{-1}\).

The marine stratocumulus clouds off the west coast of South America are trapped in the MBL below a strong temperature inversion. The boundary layer height increases westward from the west coast of South America as SST increases. West of about 100°W, the increasing SST causes a transition from stratus/stratocumulus clouds to trade cumulus clouds with cloud liquid water spread throughout a vertical domain ~2 km thick. The predicted mixing ratio of cloud liquid is about 0.2 to 0.3 g kg\(^{-1}\) for both stratocumulus and trade cumulus clouds. The average number concentration of cloud droplets is between 50 and 80 cm\(^{-3}\) for stratocumulus clouds and between 20 and 40 cm\(^{-3}\) for trade cumulus clouds, respectively. High number concentrations of cloud droplet exceeding 100 cm\(^{-3}\) are found within about 5° (longitude) off the west coast of South America. These high droplet number concentrations are caused by high loadings of natural and anthropogenic sulfate (volcanoes and copper smelters) and carbonaceous particles (anthropogenic pollution and biomass burning) advected from continental sources. The use of prescribed monthly mean aerosol fields in the current simulation prevents an evaluation of individual transport events. Over land, the average number concentration is between 200 and 300 cm\(^{-3}\) as a result of aerosol emissions from biomass burning and fossil fuel combustion in South America. Droplet concentrations in this range were also predicted in cloud ensembles over Oklahoma during summer by the high-resolution study using a cloud-system resolving model by Phillips et al. [2009]. In the present simulation, the high and steep Andes act as a barrier for aerosols emitted in central South America, preventing most of the continental emissions from being advected over the eastern Pacific.
4. Comparison to Observations

The implementation of the new cloud microphysics scheme into iRAM requires a comparison of the results from the new model version against observations in order to evaluate the ability of the model to reproduce observed cloud properties. Certain difficulties arise when comparing regional climate model results with observations. First, because of the coarse spatial resolution of the model, individual clouds cannot be resolved. Highly variable species such as cloud droplet number concentration measured, for instance, by individual flights can hardly be compared with simulated concentrations representing average values for the whole grid box. The basic strategy followed here to circumvent this problem is to average all measurement data available within a larger domain. Second, since iRAM uses a prescribed aerosol climatology to drive the cloud microphysics scheme (see section 2.2), the model is not fully capable of simulating real episodes suited for a one by one comparison. The aerosol climatology represents monthly mean concentrations representative for the late 1990’s to early 2000’s and does not capture specific events of aerosol formation or removal which might play an important role locally. Thus, measurements taken during a specific period of time cannot be compared with the model results directly, but based on time averaged means only. Therefore, only observed data covering at least several days are used here for comparison with our model results.

4.1. Total Cloud Cover
Figure 3 shows a comparison of the average total cloud cover calculated by iRAM with International Cloud Climatology Project (ISCCP) satellite observations [Rossow et al., 1996, http://isccp.giss.nasa.gov/products/onlineData.html] for the time period August-October 2006. The satellite data show three regions of high cloud cover with average values exceeding 75% near the Intertropical Convergence Zone (ITCZ) between the equator and about 10°N and the two stratocumulus decks off the west coasts of North and South America. These regions are reproduced by the model, although the position of the main stratocumulus deck off South America is shifted about 9° farther northwest than the observed. This is related to a bias in the position of the simulated liquid water content (see also section 4.2) used to calculate cloud cover in the model (equation 2). It is a model deficiency already known from simulations with the original single-moment cloud microphysics scheme and might be caused by the coarse model resolution. The high and steep Andes and the land-sea contrast are not resolved well at the model resolution of 0.5° x 0.5° [Wang et al., 2004a].

A region with low average values of coverage by all clouds (less than 40%) is found from August to October 2006 between the equator and about 15°S and extending in the east-west direction from about 110°W to the western boundary of the model domain at 140°W. This region is reproduced by the model, but is larger and extends about 5° to 10° further to the south than in the satellite observations. An underestimation of the total cloud cover by the model is also found north of 15°N between 90°W and 140°W. This might be caused by the cloud cover parameterization which might not always be able to adequately indicate the presence of stratocumulus clouds and thus underestimates the coverage by low clouds. A prognostic calculation of the cloud cover might improve prediction of coverage by low clouds compared to diagnostic schemes such as the one used in this study. The overall cloud cover is slightly
underestimated in this region by the model. The domain averaged total cloud cover in iRAM (52\%) for the time period August-October 2006 is smaller than from the ISCCP satellite observations which show about 60\%. This bias is mostly due to coverage by low clouds cover being underestimated south of the equator between 110°W and 140°W.

4.2. Liquid Water Path

The geographical pattern of the vertically integrated liquid water content from Special Sensor Microwave Imager (SSM/I) satellite observations [O’Dell et al., 2008] shows high values in the ITCZ exceeding 200 g m⁻² as well as for the two stratocumulus decks off the west coasts of North and South America with average values around 100 g m⁻². Regions with low liquid water path (LWP) values (< 30 g m⁻²) are found over the eastern Pacific close to the west coast of California and in a region south of the ITCZ extending from the western boundary of the model domain at 140°W to about 100°W and in the meridional direction from about 5°N to 10°S (Figure 4). These features of the geographical distribution are reproduced by iRAM reasonably well. As already seen from the total cloud cover in Figure 3, the location of the stratocumulus deck off the west coast of South America is shifted by about 9° to the northwest compared with the satellite observations. This shift might be caused by the horizontal model resolution, which is too coarse to resolve the steep topography of the Andes well resulting in difficulties representing the proper land-sea contrast and the impact of the mountains on the model dynamics. While the observed liquid water path is reasonably well reproduced in the ITCZ and the stratocumulus decks, the model tends to underestimate the average liquid water path over other parts of the ocean by about 30\%. The low liquid water path region off the west coast of the Baja California Peninsula and that south of the ITCZ around 120°W (less than 40 g m⁻²) simulated by iRAM are
both more extensive than that from the satellite observations. Averaged over the whole domain, iRAM simulates a liquid water path of 65 g m\(^{-2}\) over the oceans, the corresponding SSM/I observation gives 83 g m\(^{-2}\) averaged over the whole domain. The linear spatial correlation coefficient between the iRAM simulation and the SSM/I observation over the oceans is 0.77, indicating a reasonable skill of the model reproducing the spatial pattern of the cloud liquid water path over the oceans.

\section*{4.3. Daily Rainfall}

Figure 5 shows a comparison of daily mean rainfall from the model simulation with TRMM Microwave Imager (TMI) satellite estimations \cite[Wentz, 1983, \url{http://apdrc.soest.hawaii.edu/w_data/atm3.htm}]. The model captures the location and intensity of the 3-month mean precipitation over the oceans reasonably well. However, the narrow band of maximum precipitation in the ITCZ is overestimated by the model in both meridional extent and intensity. The overestimation of precipitation by the model in the ITCZ could be caused by an overestimation of droplet sizes. The overestimation of droplet radii is caused particularly by an underestimation of droplet numbers and/or overestimation of cloud liquid water (see also section 4.6 and fig. 8). Larger droplets result in increased autoconversion efficiency and thus enhance precipitation formation. The model simulates drizzle under the stratocumulus deck off the coast of South America, which is not present in the satellite observations. However, satellite measurements of precipitation are known to have difficulties in detecting such light drizzle, which has been reported in several in-situ observational studies \cite[e.g. Garreaud et al., 2001].

\section*{4.4. Cloud Droplet Number Concentration}
A comparison of cloud droplet number concentration of marine boundary layer clouds calculated by the model and obtained from Moderate Resolution Imaging Spectrometer (MODIS) satellite measurements of liquid water path and cloud optical thickness [Bennartz, 2007] is shown in Figure 6. As for the satellite data, only cases in which the liquid water path exceeds 25 g m\(^{-2}\) are taken into account to calculate the mean values from the model data. The model data have been averaged over clouds within all vertical model layers below \(\sigma = 0.75\). For the geographic region studied, this approximately resembles the averages for low marine clouds represented by the satellite data in which a threshold for the minimum liquid cloud fraction of 0.8 has been used [Bennartz, 2007]. We averaged satellite data over the months August-October for the years 2003 to 2007. We chose the multi-year average instead of the year 2006 for comparison with the model data for two reasons: First, the multi-year data cover a wider geographical area and the noise introduced by grid cells with only very few observations in August to October 2006 is reduced. Second, cloud droplet number concentration depends on the aerosol loading, which is not exactly representative for the year 2006 in the model but for average present-day conditions only. The prescribed aerosol climatology used in the model cannot represent individual events such as a specific storm system which might affect the aerosol loading significantly on local to regional scales. The model underestimates the 3-month average cloud droplet number concentration particularly over the coastal waters of the Baja California Peninsula and northern Chile. Compared with the satellite data, iRAM underestimates cloud droplet number concentrations close to the west coast of South America by about a factor of 3. The predicted number concentrations of cloud droplets decrease too quickly from the coastlines westward to the open ocean. The current model version considers aerosols in the form of a prescribed monthly mean climatology only. This does neither allow the model to capture
individual advection events nor to represent changes in aerosol sizes or chemical composition during advection of continental aerosols to the ocean. The mixing of continental aerosol particles with marine background aerosols is expected to be most important near the coastlines. This suggests that errors introduced by the current aerosol representation are an important factor for the large error in cloud droplet number concentrations in the model near the coastlines. Over most of the remote ocean, the model simulation and satellite observation agree reasonably well with number concentrations of cloud droplets ranging between 30 and 50 cm$^{-3}$ west of South America. However, the simulated droplet numbers over the remote Pacific south of 15°S are underestimated by about a factor of 2 compared with the satellite data. A tongue of elevated droplet number concentrations extending from the northern tip of Peru far out to the Pacific Ocean to about 130°W is also found in the model data. Droplet number concentrations in this tongue reach about 80-100 cm$^{-3}$ in both model data and satellite estimation, compared with droplet numbers less than 50 cm$^{-3}$ over the remote southeastern Pacific. The tongue might be formed by persistent advection (and thus captured by the aerosol climatology) of continental aerosols and precursor gases from South America over the Pacific, increasing the number of cloud condensation nuclei available for cloud formation. This is also suggested by our estimate for the anthropogenic aerosol fraction, which shows a higher anthropogenic contribution to the aerosol loading within this tongue than over the adjacent Pacific regions (see Figure 10). The linear spatial correlation coefficient between iRAM and MODIS over the oceans is 0.27, reflecting considerable differences between the model simulation and satellite observation, particularly along the Chilean Coast, over the coastal waters of the Baja California Peninsula, and in the Atlantic Ocean. Good agreement between the modeled and observed number concentrations of cloud droplets is found particularly in the remote central eastern Pacific.
4.5. Diurnal Cycle of Extent and Properties of Marine Stratocumulus Clouds

Measurements of liquid water path, cloud cover and cloud optical depth measured during the Stratus Ocean Reference Station (20°S, 85°W) mooring recovery and deployment cruises have been used to calculate an average diurnal cycle of cloud properties including cloud droplet number concentrations of marine low clouds [de Szoëke et al., 2009]. The cruises are performed annually between October and December under the stratus clouds west of northern Chile. We compare data obtained in October 2006 (STRATUS) with results from the iRAM simulation for October 2006 (Figure 7). In order to compare the diurnal cycle of clouds in the stratocumulus deck, we averaged over a 5°x5° domain in the center of the modeled stratocumulus deck off South America which is shifted to the northwest compared with observations (see sections 4.1 and 4.2) rather than the exact geographical region of the ship measurements. Figure 7 shows a distinctive diurnal cycle of the liquid water path with minimum values in the afternoon (12 to 18h local time) and maximum values during the night hours, particularly around sunrise. The diurnal cycle of cloud liquid water path including amplitude is well reproduced by the model. The ratio of maximum to minimum LWP is about 2 for both model simulation and ship observations. The observed diurnal cycle of the cloud cover shows a minimum around noon, but no distinctive variation in the morning and in the afternoon. In contrast, the modeled diurnal cycle of cloud cover shows a minimum in the early afternoon (14h local time), the amplitude of the diurnal cycle is small. iRAM systematically overestimates cloud cover by about 10%. For the STRATUS data, cloud droplet number concentrations are estimated from observed cloud optical depth limiting measurements to daytime hours. These cloud droplet numbers represent averages over the whole vertical extent of the cloud. Measured droplet numbers range 40 and 80 cm$^{-3}$.
during most of the day. The predicted droplet numbers ranging between 50 and 80 cm\(^{-3}\) are well within the observed range. Observations show no significant diurnal cycle in cloud droplet number, whereas the model calculates elevated droplet number from around noon to 18h LT. Except for the systematic bias in cloud cover, the diurnal cycle of marine cloud extent and water path in the stratocumulus deck off South America observed during the STRATUS cruise 2006 are reproduced reasonably well by the model. This suggests that the model physics capture the basic interaction of radiation and meteorological fields driving the diurnal cycle of cloud properties and extent in this region.

4.6. Vertical Structure of Cloud Properties

Vertical profiles of several cloud microphysical properties were measured from aircraft during the East Pacific Investigation of Climate Processes in the Coupled Ocean-Atmosphere System (EPIC) 2001 [Raymond et al., 2004, http://www.eol.ucar.edu/raf/Projects/EPIC2001/]. We averaged measured cloud droplet number concentration, cloud liquid water content, and effective droplet radius available from 19 flights during EPIC in September and October 2001 to obtain domain averages for comparison with our model simulation. The measurement domain covers the topical Pacific between 93°W and 99°W and 1°S and 16°N with particular focus on the ITCZ north of about 7°N. In order to improve comparability of the model simulation and the observations, we binned the measurements to the horizontal model grid (0.5° x 0.5°) and 50 hPa layers in the vertical. Following Rauber et al. [2007] we used a threshold of 0.025 g kg\(^{-1}\) for the minimum total cloud water content (sum of liquid and ice) to distinguish between in-cloud and cloud-free cases when calculating the conditional averages shown in Figure 8. It should be noted that this is not a one-by-one comparison as model and observational data represent different
years. The temporal (2-month mean) and spatial averaging smoothes differences caused by short lived synoptical weather patterns between the two different years. This should provide an idea about typical average conditions in this region as September and October neither in the year 2001 nor in 2006 are within the period of a strong El Niño or La Niña event.

iRAM is capable of reproducing the basic features of the vertical profile of cloud droplet number concentration from EPIC measurements, showing mean number concentration of about 100 cm$^{-3}$ throughout the lower troposphere between about 900 and 700 hPa (iRAM about 90 cm$^{-3}$ between 900 hPa and 800 hPa, decreasing to 50 cm$^{-3}$ at 700 hPa). This is followed by a drop in number concentration down to about 40-70 cm$^{-3}$ between 700 and 500 hPa (iRAM: 20-50 cm$^{-3}$ between 700 and 500 hPa, Figure 8a). The clouds observed above 550 hPa are not reproduced by the model, which shows decreasing droplet number concentrations above this altitude. The normalized mean error (NME) defined as

$$\sum_{i=1}^{N} \left| \frac{\text{model}_i - \text{observation}_i}{\text{observation}_i} \right| \cdot 100\%$$

(3)

of the mean cloud droplet numbers is 31%, indicating that iRAM captures the observed vertical mean profile reasonably well, but tends to underestimate droplet number particularly in the middle and upper troposphere.

Both vertical profiles of the modeled and observed cloud liquid water content show mean values increasing from about 0.1 g kg$^{-1}$ to 0.25 g kg$^{-1}$ from the near surface layer to about 800 hPa. The cloud water content stays between 0.2 and 0.3 g kg$^{-1}$ above 800 hPa (Figure 8b). While the modeled vertical profile of cloud water content is in good agreement with the aircraft
measurements in the lower troposphere up to about 800 hPa, iRAM overestimates the mean cloud water content by about 30-40% between 700 and 550 hPa. Liquid water clouds observed above 550 hPa are not reproduced by the model showing rapidly decreasing cloud liquid water in this altitude range. NME for mean cloud liquid water content is 38%.

The droplet effective radius defined as the ratio of 3rd and 2nd moments of the droplet size is shown in Figure 8c. The iRAM simulation and the EPIC aircraft measurements are in good agreement in the lower troposphere up to about 850 hPa, but overestimated by about 2 µm between 800 and 550 hPa. This overestimation is mainly caused by the overestimation of the corresponding cloud liquid water content and to some extent also by the underestimation of droplet numbers in this altitude range. Above this altitude, no significant water clouds are simulated in iRAM and the effective radius drops down to a lower threshold value applied to ensure numerical stability in case of negligible cloud droplets or liquid water content. As noted in the discussion of droplet number concentrations, the model does not reproduce these clouds above 600 hPa. NME for cloud droplet effective radius is 24%.

4.7. Cloud Radiative Forcing

The shortwave (longwave) cloud radiative forcing quantifies the impact of clouds on the Earth's radiation budget in the solar (thermal) spectral range. We calculate the shortwave (longwave) cloud forcing SCF (LCF) at the top of the atmosphere (TOA) as the difference between the all-sky shortwave (longwave) radiation and the clear-sky shortwave (longwave) radiation at the TOA. Figure 9 shows the August-October 2006 average TOA SCF and LCF in iRAM in comparison to Clouds and the Earth's Radiant Energy System (CERES) satellite observations for the same period. Here we use the CERES FM1+FM3 Edition 2 ES4 data set for
The model captures the basic geographic pattern of the SCF with highest absolute values over the eastern Pacific along the ITCZ and stratocumulus decks and lowest absolute values south of the ITCZ from 100°W to the western boundary of the model domain. Differences in the geographical distribution of the SCF are found particularly in the region with the stratocumulus deck off South America, which is too far northwest in the model due to the shift of the simulated cloud deck (see sections 4.1 and 4.2). Compared with CERES satellite data, the model overestimates the SCF in the ITCZ by about 30%, but reproduces measured SCF of about -100 W m$^{-2}$ in the stratocumulus decks reasonably well. The domain averaged SCF calculated for the year 2006 by iRAM (-59 W m$^{-2}$) is about 30% higher than the 2006 average from CERES measurements (-47 W m$^{-2}$), but compares reasonably well to the 3-month domain average of -52 W m$^{-2}$ from the CERES Energy Balanced and Filled (EBAF) climatology for the years 2000-2005 (CERES EBAF TOA Terra Edition 1a data set, Loeb et al., 2009). The low interannual variability of average SCF from the CERES EBAF climatology (-52 W m$^{-2}$) for the years 2000-2005 (standard deviation < 1 W m$^{-2}$) suggests that the SCF estimate from the unbalanced CERES data (-47 W m$^{-2}$) for the year 2006 has an uncertainty of at least 5 W m$^{-2}$. The overestimation of the modeled average SCF for the year 2006 might therefore be smaller than 30%. This is a significant improvement compared with results from the original single-moment cloud scheme (-85 W m$^{-2}$) [Wang et al., 2004a] with the modeled SCF being now much closer to the satellite observations. A reduction of the SCF overestimation is found particularly in the ITCZ and the
stratocumulus deck off South America. This improvement in SCF is particularly related to the
now physically based calculation of cloud droplet numbers instead of assuming a constant
droplet number as done in single-moment cloud schemes. Droplet numbers and thus particle size
are crucial not only for the cloud microphysics, but also for the calculation of droplet effective
radii determining the clouds’ reflectivity.

The geographical pattern of the LCF as well as its 3-month domain average calculated from
iRAM and obtained from CERES measurements are in reasonable good agreement particularly
over the ocean. However, the model underestimates LCF over central South America by a factor
of 2 and overestimates LCF along the Andes. This overestimate in LCF is mainly caused by an
overestimate of the high cloud cover in this region (see also fig. 3). A possible reason for this
overestimate could be excessive wave activity along the steep topography of the Andes, which is
not resolved well at a horizontal resolution of 0.5°x0.5° (see also section 4.1). The August to
October 2006 LCF domain average amounts to 26 W m⁻² for both iRAM and the CERES data.

It should be noted that besides cloud microphysical properties also the cloud cover plays a
crucial role in calculating cloud forcing. The cloud cover parameterization is therefore also a
possible source of errors in modeled cloud forcing. In addition to more detailed cloud
microphysics, further model improvements might also consider improving the cloud cover
parameterization or the use of prognostic cloud cover schemes.

5. Sensitivity to Changes in the Aerosol Burden
Altering the concentration of aerosol number in the ambient atmosphere can potentially alter the number of cloud condensation nuclei (CCN) activating to form clouds and thereby alter cloud optical properties and their radiative effects [Albrecht, 1989; Twomey, 1974]. Depending on local meteorological conditions, altering the number and the size of cloud droplets may also modify cloud liquid water content, cloud lifetime and precipitation patterns [Ackerman et al., 2004; Rosenfeld, 2000]. AeroCom designed and carried out a preliminary model comparison designed to test model diversity with respect to simulating the indirect effect of aerosols on climate [Penner et al., 2006]. One of the conclusions reached during this assessment was that prediction of aerosol concentrations leads to large uncertainties in the indirect aerosol effect. This finding supports the consensus reached in the 2007 IPCC report which lists aerosol indirect effects as one of the largest uncertainties in calculating radiative forcing [IPCC, 2007].

The stratocumulus decks over the eastern Pacific off the subtropical coasts of North and South America have a major impact on the Earth's radiation budget and thus climate by effectively reflecting incoming solar radiation [e.g. Randall et al., 1984]. In order to assess the indirect aerosol effect of anthropogenic emissions of aerosols and precursor gases on clouds over the eastern Pacific, we conduct a sensitivity experiment using aerosol fields for pre-industrial conditions. We compare the results of this sensitivity experiment with an otherwise identical model run using present-day aerosols (see section 2.2). The sensitivity experiment using pre-industrial aerosols consists of an ensemble of 10 model runs which are averaged to reduce statistical noise. The pre-industrial aerosol climatology is calculated from our present-day aerosol fields by multiplying each grid cell with the ratio of aerosols from natural sources to total aerosol loading. The aerosol loading from natural sources is taken from a global model simulation using pre-industrial emissions of primary particles and precursor gases, the total aerosol loading from a second simu-
lation with the same model and identical model setup using present-day aerosol and precursor gas emissions. Anthropogenic increases in biomass burning are also considered by the emission datasets [Lauer et al., 2007].

We did not use the results of the pre-industrial simulation from Lauer et al. [2007] directly because differences in the two present-day aerosol climatologies suggest that in addition to differences in the global models also differences in the emissions of natural sources such as dimethyl sulfide (DMS) over the oceans or volcanic SO$_2$ exist. These differences result in different aerosol loadings which are not entirely due to anthropogenic activities alone when assessing the indirect aerosol effect. To minimize this effect, present-day and pre-industrial simulations must use identical emission data sets for all natural sources. As an example, Figure 10 shows the anthropogenic fraction of sulfate aerosols at 850 hPa obtained from the results of the present-day and pre-industrial model runs by Lauer et al. [2007]. These fractions are used to estimate pre-industrial aerosol loadings from our present-day aerosol climatology. In this study, we use identical SST and lateral boundary conditions (present-day) in both model runs to minimize noise generated by differences in boundary conditions. Changes in cloud properties can then be attributed to changes in the atmospheric aerosol burden due to anthropogenic activities. However, some noise is introduced as differences in cloud properties feed back on model dynamics via changes in the radiation fluxes as well as differences in convection during the model simulation. It should be noted that the use of fixed SSTs does not allow representing all feedback mechanisms between atmosphere and ocean. The changes in cloud properties and radiative fluxes due to anthropogenic aerosol emissions presented here are changes without response of the ocean and might look different after long integrations with an interactively coupled ocean.
Figure 11 shows the differences in mean cloud droplet number concentration (a) and effective droplet radius (b) of low clouds for the period August-October 2006 (conditional averages). The differences are considered to be statistically significant if the t-test applied to the mean values from all ensemble members gives significance at a confidence level of 99%. The most dominant changes in cloud droplet number concentration are found over the Gulf of Mexico. There the differences in the aerosol loading are large, particularly due to anthropogenic SO₂ emissions advected from continental sources, which result in increased sulfate burdens. Changes in average cloud droplet number concentrations of low clouds range from about 70 cm⁻³ near the US Atlantic coastlines and 10-30 cm⁻³ along the Pacific coastlines of North and South America to about 2-3 cm⁻³ over the remote southeastern Pacific. The average change in droplet number concentration over all ocean grid cells in the model domain is 12 cm⁻³ (20% of the present-day value). We also find that the average liquid water path over the oceans in the model run with present-day aerosols increases by about 3 g m⁻² (4% of the present-day value) compared with the run using pre-industrial aerosols. Changes in liquid water path amount up to 6-8 g m⁻² along the Pacific coastlines and up to 8 g m⁻² over the Gulf of Mexico but only 1-3 g m⁻² over the remote southeastern Pacific. These changes in droplet number concentration and liquid water path result in a decrease of average effective droplet radii in the present-day run compared with the pre-industrial model run.

Reductions in effective droplet radii are about 0.5 µm along the Pacific coastlines and up to about 1.5 µm near the US Atlantic coast over the Gulf of Mexico but negligible over the remote southeastern Pacific. These predicted changes in droplet size are significant, in view of the estimate by Liou and Ou [1989] with a 1D climate model that a reduction by only 0.5 µm in mean droplet radius globally would offset all equilibrium warming from CO₂-doubling. We
consider the changes in effective droplet radius along the Pacific coastlines of North and South America as a lower limit because the present-day simulation underestimates the number concentration of cloud droplets by a factor of 2-3 along the Pacific coastlines (see sections 4.4). In particular these regions are expected to experience the largest impact of anthropogenic emissions because of their vicinity to the continental aerosol and precursor sources. This suggests that the indirect aerosol effect along the Pacific coastlines might be underestimated by the model. The average decrease in effective droplet radii over the ocean is 0.36 µm (2.7% of the present-day value). For a given liquid water content and geometric thickness, cloud optical thickness is inversely proportional to the droplet effective radius as smaller droplets are more effective in reflecting solar radiation than larger cloud droplets. The results of the 3-month sensitivity experiment (10 ensemble members) do not show statistically significant changes in the TOA SCF over the remote ocean for the stratocumulus deck off South America. The changes in SCF calculated by iRAM are too small compared with the variability of the individual ensemble members to quantify the radiative forcing of the indirect aerosol for the Pacific stratocumulus decks. Thus, only domain averages can be given, which amount to -2.6 W m\(^{-2}\) for the change in the 3-month average TOA SCF and -1.6 W m\(^{-2}\) TOA net cloud forcing (\(\text{CF}_{\text{net}} = \text{TOA SCF} + \text{TOA LCF}\)) over the ocean. No statistically significant changes are found for total cloud cover and precipitation.

Sensitivity of the Calculated Indirect Aerosol Effect to the Autoconversion Scheme

In addition to the sensitivity study described above, we conducted a second pair of otherwise identical model experiments to assess the indirect aerosol effect applying the
alternative autoconversion scheme from Khairoutdinov and Kogan [2000] (KK2000, see section 2.2) instead of the parameterization from Liu et al. [2007] (L2007). Autoconversion is an important process controlling the amount of liquid water and number concentration of cloud droplets in warm clouds. Autoconversion rates predicted by the two schemes for droplet numbers and liquid water contents typically found in marine stratocumulus clouds differ up to a factor of five. Despite these differences, both autoconversion schemes predict similar changes in the domain averaged number of cloud droplets amounting 11 cm\(^{-3}\) for KK2000 and 12 cm\(^{-3}\) for L2007. Also predicted changes in TOA shortwave and longwave cloud forcing (\(\Delta SFC = -2.6 \text{ W m}^{-2}\) and \(-2.9 \text{ W m}^{-2}\), \(\Delta LCF = 1.0 \text{ W m}^{-2}\) and \(0.9 \text{ W m}^{-2}\) for KK2000 and L2007, respectively) agree within the variability of the individual ensemble members. However, differences between both autoconversion schemes are found for the liquid water path. Changes predicted by KK2000 (4.0 g m\(^{-2}\)) are about 50\% larger than those predicted by L2007 (2.7 g m\(^{-2}\)), which suggests a higher sensitivity of KK2000 to aerosol perturbations than of L2007. We also found that in particular the variabilities of liquid water path, droplet number concentration, cloud forcing and effective radius are about 50-100\% higher among the individual ensemble members for KK2000 than for L2007. Again, this suggests a higher sensitivity of KK2000 to changes in cloud microphysical properties than of L2007. Nevertheless, differences in predicted changes of domain averaged TOA cloud forcing are small.

Table 2 summarizes the changes in the domain averages over the ocean due to anthropogenic emissions of aerosols and precursor gases for both autoconversion schemes.

6. Summary and Conclusions
A double-moment cloud microphysics scheme with semi-prognostic (in-cloud) aerosol component with six aerosol chemical species has been implemented into the IPRC Regional Atmospheric Model iRAM. This allows for a more detailed representation of cloud processes and aerosol-cloud interactions. In order to evaluate the results from a first 3-month integration performed with the new model system, the model output of iRAM has been compared with various observations such as cloud droplet number concentration, effective droplet radii and cloud forcing at the top of the atmosphere. The results show that iRAM reproduces the geographical patterns of basic cloud parameters such as cloud cover, liquid water path, cloud droplet number concentration, effective droplet radius, and precipitation reasonably well.

Good agreement between model results and satellite observations is found particularly for the stratocumulus decks off South America although the modeled stratocumulus deck is shifted to the northwest compared with that observed. Very high number concentrations of droplets observed near the coastlines of North and South America are not captured by iRAM, which underestimates droplet number by a factor of 3 in these regions. This could be related to the use of the aerosol climatology as input for the cloud microphysics scheme in the current model version. Firstly, the fixed aerosol climatology does not allow us to represent individual advection events of continental aerosols and precursor gases over the ocean. Secondly, the pre-scribed aerosol size-distributions are currently limited to average continental and marine conditions. Thus, mixing of continental aerosols with marine background particles and the associated changes in particle sizes and chemical composition cannot be captured. This implies that errors in representing aerosols and thus the calculated cloud droplet number concentrations are the largest near the coastlines. Also, no feedback from clouds onto the environmental aerosol fields was represented here.
The comparison with observations also showed that the model simulated stratocumulus deck off South America is about 9° too far to the northwest. This model deficiency is already known from model experiments with the single-moment cloud microphysics scheme and thus might be because of the horizontal resolution of the model, which is too coarse to resolve the steep Andes and their impact on the atmosphere. The longwave cloud forcing is found to be in good agreement with satellite measurements both qualitatively and quantitatively. The geographical pattern of the shortwave cloud forcing is also in good agreement with satellite observations. The shortwave cloud forcing is significantly improved compared with the old single-moment cloud microphysics scheme, but still overestimated particularly in the ITCZ. This might be partly due to the radiation code, which does not have all the most modern features and partly due to the overestimated clouds/precipitation in the ITCZ.

We conducted a sensitivity experiment with pre-industrial aerosol fields to assess the indirect aerosol effect of anthropogenic emissions of particles and precursor gases on clouds over the eastern Pacific by comparing the results with a corresponding present-day simulation. This sensitivity experiment shows an increase of about 20% (12 cm$^{-3}$) in the average cloud droplet number concentration over the ocean, particularly along the Pacific coastlines and over the Gulf of Mexico and Atlantic Ocean with local changes up to 70 cm$^{-3}$. With only a small increase in cloud liquid water content of about 4% of its present-day value, the additional anthropogenic CCN result in a decrease in the 3-month mean effective droplet radii of low clouds by 0.36 µm over the ocean. Based on a comparison of the results from the present-day aerosol loading run with observations, we consider these changes along the Pacific coasts of North and South America as a lower limit. The modeled change in the domain averaged TOA net cloud forcing over the ocean is -1.6 W m$^{-2}$ and in TOA shortwave cloud forcing -2.6 W m$^{-2}$. No statistically
significant changes are found in this study for total cloud cover and precipitation, as the changes are small compared with the variability of the 10 ensemble members. An additional sensitivity study using the alternative autoconversion scheme from Khairoutdinov and Kogan [2000] replacing the parameterization from Liu et al. [2007] showed little effect on predicted changes in domain averaged droplet numbers and cloud forcing due to anthropogenic aerosols. This sensitivity study however showed larger changes in liquid water path (50%) and higher variability of predicted changes in cloud properties (50-100%) for KK2000 than for L2007. This suggests a higher sensitivity of KK2000 to perturbations of cloud microphysical properties than that of L2007.

The use of a prescribed aerosol climatology does not allow resolution of individual events such as storm systems which might affect the atmospheric aerosol loading significantly on a local to regional scale. The fixed aerosol climatology also does not allow representation of all feedback processes and aerosol-cloud interactions such as changes in formation of new clouds or changes in aerosol properties themselves after a large fraction of the aerosol has been removed by scavenging. In addition, the current method of using prescribed size-distributions for marine and continental aerosol to calculate particle numbers from aerosol mass concentrations cannot account for changes in the aerosol size-distribution caused by mixing of different air masses as in the case of outflow of polluted air from the continents over the ocean. The next step in improving the representation of aerosol-cloud interactions and our assessment of the indirect aerosols effect over the eastern Pacific will include: 1) comparison between aerosol climatology and in-situ observations from recent airborne field campaigns, PASE (Pacific Atmospheric Sulfur Experiment) and VOCALS, 2) more dynamic representation of primary and secondary aerosols emission and/or formation as well as their transformation and removal by wet and dry
deposition.

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**Table 1.** Log-normal distributions used in iRAM to calculate particle number from aerosol mass concentrations. Particle numbers are scaled to match total aerosol mass. $z$ denotes the height above sea level (km).

<table>
<thead>
<tr>
<th>aerosol compound</th>
<th>diameter mode 1 (μm)</th>
<th>sigma mode 1</th>
<th>diameter mode 2 (μm)</th>
<th>sigma mode 2</th>
<th>number ratio mode 1 : mode 2</th>
<th>reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>SO$_4$, maritime boundary layer</td>
<td>0.033</td>
<td>1.48</td>
<td>0.15</td>
<td>1.51</td>
<td>1:1.11</td>
<td>McNaughton [2008]; Heintzenberg et al. [2000]</td>
</tr>
<tr>
<td>SO$_4$, maritime free troposphere</td>
<td>0.04 · exp$\left(-\frac{z-2}{7.2}\right)$</td>
<td>2.0</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>McNaughton [2008]; Caffrey et al. [2006]</td>
</tr>
<tr>
<td>carbon (BC, POM) + SO$_4$, continental</td>
<td>0.028</td>
<td>1.81</td>
<td>0.069</td>
<td>1.80</td>
<td>1:4.72</td>
<td>McNaughton [2008]</td>
</tr>
<tr>
<td>carbon (BC, POM) + SO$_4$, maritime</td>
<td>-</td>
<td>-</td>
<td>0.142</td>
<td>1.59</td>
<td>-</td>
<td>Clarke et al. [2004]</td>
</tr>
<tr>
<td>mineral dust, boundary layer</td>
<td>-</td>
<td>-</td>
<td>0.53</td>
<td>2.2</td>
<td>-</td>
<td>McNaughton [2008]</td>
</tr>
<tr>
<td>mineral dust, free troposphere</td>
<td>-</td>
<td>-</td>
<td>0.57</td>
<td>2.1</td>
<td>-</td>
<td>McNaughton [2008]</td>
</tr>
<tr>
<td>sea salt, marine boundary layer</td>
<td>-</td>
<td>-</td>
<td>0.5</td>
<td>2.12</td>
<td>-</td>
<td>McNaughton [2008]</td>
</tr>
</tbody>
</table>

**Table 2.** Changes in domain averaged cloud properties over the ocean due to anthropogenic emissions of aerosols and precursor gases for the time period August-October 2006 and the autoconversion schemes from Khairoutdinov and Kogan [2000] (KK2000) and Liu et al. [2007] (L2007). CF$_{\text{net}}$ is the net TOA cloud forcing calculated as the sum of longwave and shortwave TOA cloud forcing, CDNC and $r_{\text{eff}}$ specify the number concentration and effective radius, respectively, of cloud droplets of low clouds. Mean values and standard deviations.
calculated from the individual ensemble members are given. Changes marked by an asterisk (*) are not statistically significant compared with the variability of the individual ensemble members.

<table>
<thead>
<tr>
<th>autoconversion scheme</th>
<th>Δ cloud cover (%)</th>
<th>Δ LWP (g m⁻²)</th>
<th>Δ CDNC (cm⁻³)</th>
<th>Δ SCF (W m⁻²)</th>
<th>Δ LCF (W m⁻²)</th>
<th>Δ CFnet (W m⁻²)</th>
<th>Δ rain rate (mm d⁻¹)</th>
<th>Δ r_eff (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>KK2000</td>
<td>0.8 ± 0.5</td>
<td>4.0 ± 0.6</td>
<td>11.3 ± 0.4</td>
<td>-2.9 ± 0.7</td>
<td>0.9 ± 0.4</td>
<td>-2.0 ± 0.5</td>
<td>(*) 0.026 ± 0.033</td>
<td>-0.36 ± 0.02</td>
</tr>
<tr>
<td>L2007</td>
<td>(*) 0.4 ± 0.4</td>
<td>2.7 ± 0.4</td>
<td>11.7 ± 0.2</td>
<td>-2.6 ± 0.3</td>
<td>1.0 ± 0.4</td>
<td>-1.6 ± 0.3</td>
<td>(*) 0.040 ± 0.051</td>
<td>-0.39 ± 0.01</td>
</tr>
</tbody>
</table>
Figure 1. Mean vertical profiles of aerosol mass concentrations for sulfate, mineral dust and sea salt particles as well as total particle number (CN) of particles > 10 nm from measurements during INTEX-B near Hawaii (a) and over the Gulf of Mexico (b) [McNaughton, 2008] in comparison to the model climatology used in this study. The aerosol mass concentrations are given in µg std-m⁻³, particle numbers in std-cm⁻³. Error bars show the standard deviation of the measurements calculated from 1-minute averages.
Figure 2. Zonal cross sections along 10°S of the August-October 2006 averaged (conditional average, cloudy grid cells only) cloud liquid water content (a) and cloud droplet number concentration (b). Black contours show the temperature (K), the white contour depicts the average boundary layer height. The model orography is in black.

Figure 3. August-October 2006 average total cloud cover. Left: iRAM simulation, right: International Cloud Climatology Project (ISCCP) satellite observations [Rossow et al., 1996].
Figure 4. August-October 2006 average vertically integrated cloud liquid water content over the ocean in g m\(^{-2}\). Left: iRAM simulation, right: Special Sensor Microwave Imager (SSM/I) satellite observations [O’Dell et al., 2008].

Figure 5. August-October 2006 average daily rainfall over the ocean in mm d\(^{-1}\). Left: iRAM simulation, right: TRMM Microwave Imager (TMI) satellite observations [Wentz, 1983]. TRMM data have been reduced to 0.5° x 0.5° to match the model horizontal resolution.
Figure 6. August-October mean of the layer averaged cloud droplet number concentration of marine low clouds in cm$^{-3}$. Left: iRAM simulation (year 2006), right: MODIS satellite observations (multi-season average, 2003-2007).

Figure 7. Mean diurnal cycle of liquid water path (LWP) in g m$^{-2}$, total cloud cover and cloud...
droplet number concentration (CDNC) in cm$^3$ in the marine stratocumulus deck off South America in October 2006. Solid lines: iRAM simulation, dashed lines: Ship observations obtained during the Stratus Ocean Reference Station (20°S, 85°W) mooring recovery and deployment cruise in October 2006 [de Szoek et al., 2009].

Figure 8. Vertical profiles of cloud droplet number concentration (a), cloud liquid water content (b), and cloud droplet effective radius (c) from aircraft measurements during EPIC 2001 [Raymond et al., 2004] and simulated by iRAM. The aircraft measurements have been binned to a 0.5° x 0.5° grid and 50 hPa layers.
Figure 9. August-October 2006 average shortwave (solar spectral range, top) and longwave (thermal spectral range, bottom) cloud forcing at the top of the atmosphere (TOA) in W m$^{-2}$. Left: iRAM simulation, right: Clouds and the Earth's Radiant Energy System (CERES) satellite observations [Wielicki et al., 1996].
Figure 10. August-October average anthropogenic sulfate aerosol fraction at 850 hPa obtained from multi-year model simulations with a global aerosol model [Lauer et al., 2007].

Figure 11. Changes in the 3-monthly mean (August-October 2006) number (a) and effective radius (b) of cloud droplet in low clouds between model runs with present-day and pre-industrial aerosols. Differences that are not significant at 99% confidence level compared with the variability of the individual ensemble members are masked out in gray.