Can global models ignore the chemical composition of aerosols?

E. L. Roesler and J. E. Penner

Received 9 August 2010; revised 22 October 2010; accepted 2 November 2010; published 24 December 2010.

The number of cloud droplets formed from a population of aerosols depends on the aerosol number concentration, \( N_A \), the size distribution, and the chemical composition. The cloud albedo effect occurs when increasing \( N_A \) causes increases to the droplet concentration, \( N_D \). We examined the effects of changing aerosol size, composition, and number on \( N_D \) within the United States. We found that changing the water-soluble organic carbon (WSOC) fraction from 50% to 0.05% in the fine mode aerosol and from 50% to 95% in the coarse mode aerosol decreased \( N_D \) by an average of 34%. Our results show that the changes to the aerosol composition cause over a 20% change to \( N_D \), a magnitude previously estimated to cause a 1 W m\(^{-2}\) change in radiative forcing. Given the realistic range of aerosol compositions used here, it is not possible for global models to correctly calculate the cloud albedo effect if composition is ignored. Citation: Roesler, E. L., and J. E. Penner (2010), Can global models ignore the chemical composition of aerosols?, Geophys. Res. Lett., 37, L24809, doi:10.1029/2010GL044282.

1. Introduction

The largest uncertainty in climate change forcing [Forster et al., 2007] is the cloud albedo effect. Global models use empirical relationships based on regional studies or mechanistic activation schemes to calculate \( N_D \) [e.g., Pringle et al., 2009]. A focus of current research is to understand which microphysical variables have dominant roles, thereby eliminating the need for global models to keep unnecessary variables. For example, previous studies have shown that aerosol microphysical variables such as size, number, and small concentrations of coarse mode aerosols in a population of fine mode aerosols dominate in the prediction of \( N_D \) [Chen and Penner, 2005; Dusek et al., 2006; Feingold et al., 1999; Feingold, 2003]. Other studies have shown crustal and organic aerosols also influence \( N_D \) [Ervens et al., 2005; Kelly et al., 2007; Nenes et al., 2002]. Based on these studies, we changed the microphysical variables in a warm microphysics model to identify which variables changed \( N_D \) by 10–20%. These limits of change in \( N_D \) were chosen because a decrease in radiative forcing of \(-1\) W m\(^{-2}\) has been estimated if \( N_D \) is increased by 20% [Facchini et al., 1999].

2. Model Description and Input Parameters

We used the Parcel Undergoing Thermodynamic Transitions (PUTT), a warm microphysics model [Seidl, 1989]. The initial relative humidity of the parcel was 98% and was lifted adiabatically 300 meters from an initial starting pressure of 900 mbars at a speed, \( w \), of 10, 20, 50, 150, or 300 cm s\(^{-1}\). The size distribution of the aerosols was modeled as the sum of two lognormal functions each discretized into ninety bins.

PUTT also calculates the absorption of nitric acid gas, HNO\(_3\)\(_{aq}\), into the aerosol particles. The parcel’s initial gas-phase nitrate concentrations, 0.01 to 31.3 ppbv, were derived from the model results of Feng and Penner [2007] for each region and season. Feng and Penner [2007] found the model overpredicted the observations in North America. A constant value of 0.2 ppbv for each region and season, which would have given better agreement with the observations, was used in a sensitivity test.

The predicted values for \( N_D \) have different responses to internal and external aerosol mixtures [McFiggans et al., 2006], and small concentrations of large aerosols can greatly affect \( N_D \) as well as the formation of precipitation [Feingold et al., 1999]. The IMPROVE dataset does not provide the coarse mode PM\(_{10.0}\) aerosol composition, size distribution parameters, or the mixing state of the fine and coarse mode aerosols. Measurements taken near the Owens (dry) Lake, a saline playa with large and frequent dust storms in the spring and fall [Labban et al., 2004], were used to constrain the coarse mode aerosol parameters. The composition of the fine mode was similar to the coarse mode aerosols [Labban et al., 2004]. The fine mode composition in the IMPROVE regions affected by Owens (dry) Lake dust storms was also similar to the fine mode measurements by Labban et al. [2004]. It was assumed that the fine and coarse mode compositions were equal for these regions in PUTT. Relevant measurements were not available for the composition of the coarse mode for the remainder of the regions. All regions were then assumed to have the same fine and coarse mode composition. This assumption was tested with sensitivity tests where differing fine and coarse mode compositions were used. We assumed the IMPROVE data, when averaged, was an aged background aerosol composition, so external mixtures were not used.

A large component of the fine aerosol mass in the IMPROVE network is organic carbon (OC), but the fraction of OC that is water-soluble is not given [Malm et al., 1994, 2004]. We assumed 50% of the OC was WSOC. Measurements have found WSOC fractions in this range [Lowenthal
et al., 2009; Pio et al., 2007]. Sensitivity tests also examine this assumption.

[8] Values for the van’t Hoff factor, molecular weight, density, charge, and soluble fraction of OC were needed for the WSOC. Ervans et al. [2005] suggested that a van’t Hoff factor of one produced the lowest error in predicting $N_D$, and Mircea et al. [2005] showed that the average predicted $N_D$ was 20% smaller than the measured $N_D$ when the organics were assumed undissociated. Ervans et al. [2005] also found that high molecular weight species (M > 400 g mol$^{-1}$) influence droplet concentrations. For simplicity, we assumed the WSOC had a molecular weight of 50 grams mol$^{-1}$, a van’t Hoff factor of one, a density of 2.0 grams cm$^{-3}$, and carried no charge.

[9] Two parameterizations of surface tension, $\sigma_T$, were compared in this study. Mircea et al.’s [2005] parameterization and treating $\sigma_T$ as the sum of the multi-component aqueous solution [e.g., Topping et al., 2007]. PUTT’s treatment of $\sigma_T$ had previously accounted for only the inorganic aerosol components [Seidl, 1989]. We included values of surface tension as a function of WSOC taken under a variety of atmospheric conditions (i.e., polluted continental, remote continental, biomass burning conditions, and wet-season) [Facchini et al., 1999, 2000; Mircea et al., 2005].

3. Description of Sensitivity Cases

[10] Table 1 lists the base cases and test cases we considered. B, N, and NS are the base cases to which other cases are compared. The base cases use the aerosol compositions created from IMPROVE and are different in $N_A$ and $\sigma_g$. Any cases not marked with an S use a geometric standard deviation and mode radius fit to the size distribution of Dusek et al. [2006] in the fine mode ($\sigma_{gf}=1.5$) and of Niemeyer et al. [1999] for the coarse mode ($\sigma_{gc}=1.5$). Cases marked with an S use $\sigma_{gf}=2.0$ and $\sigma_{gc}=3.5$. For cases 1, 3–12, and 21, $N_A$ was calculated for each region from the measured mass concentration in IMPROVE. For cases 2 and 13–20, all regions have a fine and coarse mode number concentration of $N_{A,f} = 1000$ cm$^{-3}$ and $N_{A,c} = 0.75$ cm$^{-3}$, respectively, based on typical continental $N_A$ values [Seinfeld and Pandis, 2006]. For all cases, the fine mode and coarse mode radii are 0.03 $\mu$m and 0.3 $\mu$m, respectively.

[11] Cases marked with a G assume [HNO$_3$]$_{\omega}$ = 0.2 ppbv in every region, otherwise results from Feng and Penner [2007] were used. Cases marked with a C used a simplified composition of 6% H, 48% SO$_4$, 20% WSOC, and 26% insoluble components in every region, derived from a correlation of the droplet numbers with each component of the composition over all regions and vertical velocities in cases B, BS, and BG. Surface tension was calculated using Mircea et al.’s [2005] parameterization, but cases marked st calculate $\sigma_T$ as the sum of the multi-component aqueous solution. Cases marked ln1 used only the fine mode mass and concentration to explore how neglecting the coarse mode mass would affect $N_D$. Cases marked H assumed that a gas-aerosol nitric acid equilibrium is not achieved prior to updraft. For all simulations, the accommodation coefficient for [HNO$_3$]$_{\omega}$ was equal to 0.05 [Xue et al., 2005]. There is uncertainty in the value of the water vapor accommodation coefficient, $\alpha$ [McFiggans et al., 2006]. Cases labeled A set $\alpha$ to 1.0 instead of 0.1. Cases labeled Win assume 50% and 0.05% of the OC in the fine and coarse modes, respectively, is WSOC. Cases labeled W2nd assume 0.05% and 95% of the OC in the fine and coarse modes, respectively, is WSOC. Cases Win and W2nd test ranges of measured WSOC fractions [Lowenthal et al., 2009; Pio et al., 2007].

4. Case and Regional Comparisons of $N_D$

[12] Table 1 lists the average difference between each test case and base case normalized by the mean of the base case. Mean droplet number increases with vertical velocity. The largest differences in absolute percentage values for base case B is that with test cases N and BW2nd. The largest differences in absolute percentage values for base case NS is that with test cases N and NSW2nd. An average increase in $N_A$ in test case N creates more droplets than in base cases B and NS. In cases BW2nd and NSW2nd, the amount of

---

Figure 1. (a) The 28 regions created from the 187 IMPROVE network locations. (b) Droplet concentration, $N_D$ (cm$^{-3}$) for $w = 20$ cm s$^{-1}$ average of seasons for case B. (c) Same as Figure 1b but with case NS. (d) Same as Figure 1b but with case BW2nd. (e) Same as Figure 1b but with the spring season compositions.
soluble mass was decreased in the fine mode and increased in the coarse mode causing the larger, but fewer, more soluble aerosols to form droplets at the expense of the smaller, more numerous, less soluble fine mode aerosols. The third largest difference for \( N_D \) for base case B is with test case BS. This is due to the increased width of the size distribution and higher concentration of large-radii aerosols forming droplets at the expense of the small-radii aerosols. The remainder of the sensitivity tests did not have average differences greater than 20% for \( N_D \) between the base cases and test cases.

The inter-regional variation (standard deviation divided by the mean \( N_D \)) shows how the changes in composition between regions or changes in the microphysical variables affect \( N_D \). A high inter-regional variation value of \( N_D \) for a case implies an empirical relationship of \( N_D \) based on a region’s value would not be accurate if applied to other regions. Figures 1b–1e show \( N_D \) (cm\(^{-3}\)) in every region for a subset of the test cases from Table 1. Changes in composition between regions cause an inter-regional variation in \( N_D \) of 8% when all the seasons are averaged (Figure 1b), and a 15% variation of \( N_D \) in the spring (Figure 1e). The mean \( N_D \) is increased by 6% to 534 cm\(^{-3}\) in spring compared to the annual average mostly due to a factor-of-two average increase in [HNO\(_3\)](g). The \( N_D \) in spring is increased by 20% along the eastern U.S. in regions 2 and 16 primarily due to an average increase in [HNO\(_3\)](g) from 13 to 26 ppbv. The \( N_D \) in regions 8, 9, and 19 also increased by 15% due to the increase in [HNO\(_3\)](g) from 7 to 14 ppbv. Changes to the composition caused changes to \( N_D \) within and between regions by 10–20%.

The annual average of \( N_D \) for base case NS, shown in Figure 1c, has the same aerosol composition as the annual average base case B (Figure 1b), and \( N_D \) is also unique in every region which causes an inter-regional variation of 48%. The average \( N_D \) decreased by 12% compared to case B due to an average decrease in \( N_A \) in case NS. Figure 1d shows the BW2nd case which has the largest inter-regional variation of 60%. From Table 1, case BS has the third largest average difference in \( N_D \) from base case B but has a negligible inter-regional variation (not shown in Figure 1). This is due to the increased width of the size distribution and higher concentration of large-radii aerosols forming droplets at the expense of the small-radii aerosols.

5. Changes to \( N_D \) for Different \( S_{\text{max}} \)

Figure 2 shows the computed \( N_D \) at the maximum supersaturation, \( S_{\text{max}} \), for test cases 5–12 against base case
Acknowledgments.
We thank Yang Chen for his contributions.

References


E. L. Roesler and J. E. Penner, Department of Atmospheric, Oceanic, and Space Sciences, University of Michigan, Space Research Building, 2455 Hayward St., Ann Arbor, MI 48109-2143, USA. (eroesler@umich.edu)