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IN-CLOUD AND BELOW-CLOUD NUMERICAL SIMULATIONS OF SCAVENGING PROCESSES AT AMAZON BASIN DURING LBA-SMOCC

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ABSTRACT

Atmospheric scavenging processes have been investigated, taking into consideration a numerical simulation through the model Regional Atmospheric Modeling System (RAMS) and the below-cloud and in-cloud scavenging model (B.V.2) to Amazon Basin region. The RAMS modeling was off-line coupled with a one-dimensional (1-D) below-cloud and in-cloud scavenging model in order to simulate the scavenging processes. RAMS modeling results were used in order to obtain cloud structures. Real-time measurements of ammonia, nitric acid, hydrochloric acid, sulfur dioxide and the water-soluble inorganic aerosol species, ammonium, nitrate, chloride, and sulfate were performed at a pasture site in Amazon Basin (Rondonia, Brazil) during LBA/SMOCC campaign (Large-Scale Biosphere Atmosphere Experiment in Amazonia - Smoke Aerosols, Clouds, Rainfall, and Climate: Aerosols From Biomass Burning Perturb Global and Regional Climate - September 12 to November 14, 2002). The concentrations of gases and particles in the samplings, as well as the meteorological parameters obtained during the campaign LBA/DRY-TO-WET (September and November 2002), that happened in the transition period, between the late dry season and the onset of wet season, were used as input data in the scavenging model and for atmospheric simulation comparison purpose. Gamma distribution adjusted for the each case and Levine & Schwartz, 1982 droplet spectra were also used in order to compare the rainwater concentration species due in-cloud scavenging. The aim of modeling was to predict the average concentration of three chemical species found in rainwater, SO_4^{2-}, NO_3^- and NH_4^+, scavenged from the atmosphere. Another objective was to compare modeled and observed rainwater composition. In this work, two precipitation events (October, 9 and 10) were studied. The preliminary results based on scavenging modeling are in the following: a) Droplet spectra are quite important to nucleation processes, b) The incloud scavenging processes dominates (between 80% and 97% of total in rainwater concentration), c) Rainwater modeled sulfate presents a better fit to the observed data compared to ammonium and nitrate. Results shows good agreement between modeled and observed data.

1. INTRODUCTION

In the last years, mainly after industrial revolution period, the industrial activity growing has caused abrupt changes in environment, mostly due large emissions to atmosphere. In remote regions where forests were dominant, fires even due naturals and anthropogenic causes are important sources of gases and particulate matter (PM) to atmosphere (Yamasoe et al., 2000). Those substances bring so many consequences to climate and weather dynamics, as greenhouse effect and changes in precipitation regimes. Direct observations have been revealed differences in cloud formation patterns over regions where land use has changed (Cutrim et al., 1995).

Scavenging process by wet deposition has been widely studied around the world. Those studies concentrate efforts mainly in rainwater chemical analysis, generally in sequencial samplers performing distinction between composition and building relations with possible sources emissions (Pruppacher et al., 1983; Frank et al., 1990; Naik et al., 1994).

For Amapá/Brasil amazonic region, Gonçalves (Gonçalves et al., 2003) numerically simulated rainwater composition using a scavenging model (B. V. 2) and an atmospheric model (RAMS) associated. The authors have found that in-cloud process dominates when compared to below-cloud. Afterward, ionic species studied by authors, sulfate was that model results has better agreement with

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observed data collected during Serra do Navio experiment in Amapá (May, 1995 – June, 1997 period).

The present work aim to use scavenging model to simulate the rainwater composition in Rondônia (sulfate, nitrate, and ammonium) that were removed by wet deposition and study physical process evolved. The Regional Atmospheric Model (RAMS) is used in order to obtain atmospheric parameters to be used as input to scavenging model. Comparisons are made using observed data collected during *Large Scale Biosphere Atmosphere Experiment in Amazônia / Smoke Aerosols, Clouds, Rainfall, and Climate: Aerosols From Biomass Burning Perturb Global and Regional Climate* (LBA-SMOCC) campaign.

2. METHODS AND MATERIALS

2.1. The Rondonia`s region

Field measurements were performed at a pasture site in Rondonia state (Brazil), at Fazenda Nossa Senhora Aparecida (FNS - 10º45'44''S, 62º21'27''W, 315 m asl) located in the southwestern part of Amazon Basin (Trebs et al., 2004) during LBA/DRY-TO-WET campaign (September to November 2002) and LBA/SMOCC campaign (September, 12 to November, 14 – 2002). The site is characterized by small seasonal temperature variations, ranging from 23–24ºC at the end of the wet season (June/July) to 25–26ºC during the dry season (September/October). Annual relative humidity values are high, with averages of about 70% in the dry season and 80% during the wet season. Precipitation in this region shows a seasonal cycle with lowest values in July and is highest from November to April (up to ~300 mm per month) (Andreae et al., 2002).

2.2. Available data

Rainwater compositions analysis and PM and gaseous data compositions from atmosphere were used as input data for B. V. 2 scavenging model. For RAMS atmospheric model as used the NCEP reanalysis (Kalnay et al., 1996) with 2.5°x2.5° horizontal resolution and 6 hours time intervals, as initial and boundary conditions.

2.3. Events and studied pollutants

For such study two precipitation events were chosen: October 9 and 10, 2002. The selected events were based on the available data sets for both aerosol and rainwater samples. The chemical species to be simulated in rainwater are the following: sulfate, ammonium and nitrate, through the atmospheric scavenging of sulfur dioxide, nitric acid and ammonia gases and of particulates ones, sulfate, ammonium and nitrate for wet deposition.

2.4. Scavenging process modeling

The methodology used in this work could be itemized following:

a) Numerical modeling of atmospheric conditions, including cloud structures during the selected events, using RAMS:

Two simulations has been performed, each one for such event, both using RAMS version 5.0, which has been configured using four nested grids, centered at FNS site, using 64, 16, 4 and 1 kilometer horizontal resolution, Grell convective parameterization active in mother domain and microphysics using 5-form parameter in nested grids.

b) Below-cloud scavenging numerical modeling:

Below cloud scavenging modeling has been performed using the one-dimension "*Below Cloud Beheng Version 2*" (B. V. 2), described by Gonçalves (1997). This model performs rainwater composition forecast based on the gaseous and PM initial conditions, as atmospheric profiles obtained from RAMS simulations.

c) In-cloud scavenging numerical modeling:

For that case B. V. 2 model adaptation was needed to work properly with in-cloud scavenging. The most principal changes were: 1) Atmospheric parameters model input changes; 2) Gaseous and PM initial concentration changes using exponential decrease (Seinfelds & Pandis, 1998); 3) Implementation of two droplets distribution functions, the first one, an exponential function determined by Levine & Schwartz (1982), and the second one, a gamma function adjusted with 5-form parameter to the collected observed data during the flights carried through in campaign LBA-SMOCC.

d) Integrated process study:

Considering the concentration exponential decline with time (Seinfeld & Pandis, 1998), the total amount of pollutants removed from cloud can be gotten integrating the relations in each cloud layer and adding the below-cloud removed amounts of gas and particulate matter.

3. RESULTS AND DICUSSIONS

RAMS simulations for both events presented spatial distribution and rainfall intensity similar to that shown on weather radar maps and satellite images (not showed). However, in both the cases, the model underestimated the amount of precipitation observed in the experimental site.

3.1. October 10th event

This event had beginning 18:06h UTC and ending 18:19h UTC, lasting ~20 minutes. The total rainfall amount for this event was 0.39 mm. Thus, it was found a sulfate total of 1689.1 μg/l, while that the nitrate total was 4718.8 μg/l in rainwater analyzed. However, it did not have measurements of ammonium and ammonia due equipment failure, for this reason, these will not be evaluated for this event.

In order to verify the improvement in physical representation of sulfate and sulfur dioxide scavenging due the implementation of a droplets distribution function adjusted to the observed conditions, two simulations had been carried through: the first one using the droplets distribution function proposal for Levine & Schwartz (1982) and second, using the droplets distribution by gamma function with 5 form parameter.

Figure 1 shows an improvement of 23.2% in scavenging results due implementation of a more realistic droplets distribution function. Analyzing the contribution of each removal process stage separately (nucleation, in-cloud and below-cloud), it is observed a predominance of processes that occurs in-cloud, being these responsible ones (nucleation + in-cloud) for ~95% of sulfate concentration found in rainwater for this case.

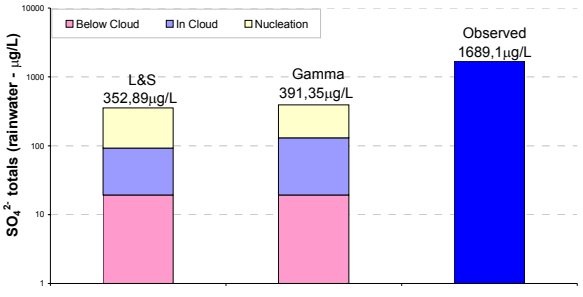

FIGURE 1 - SO_4^{2-} AND SO_2 WET DEPOSITION RESULTS COMPARISON FOR SCAVENGING MODEL USING DROPLETS GAMMA DISTRIBUTION AND LEVINE AND SCHWARTZ DISTRIBUTION FOR OCTOBER 10TH, 2002

For nitrate, however, the simulation with droplets spectra representation proposed by Levine & Schwartz (1982) was not carried through, since it was verified previously that gamma function adjusted to observed data represents this spectra better than the other one.

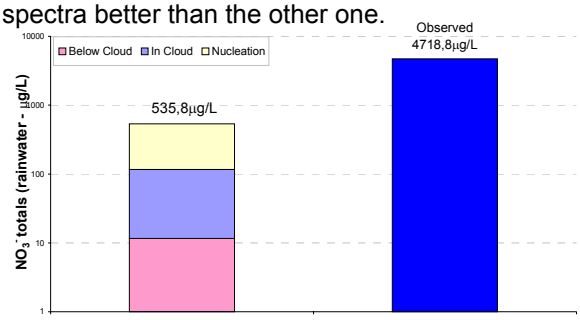

FIGURE 2 - NO_3^- AND HNO_3 SCAVENGING RESULTS FOR OCTOBER 10TH, 2002

Figure 2 analyses indicate that the model tends to underestimate the sulfate concentration in rainwater, having represented only 11.4% of total sulfate observed in the experimental site for this event. Additionally, it was observed a bigger contribution of in-cloud scavenging also for this element, being this responsible one (together with the nucleation of droplets) for ~98% of nitrate concentration in rainwater analyzed.

3.2. October 9th event

This event had beginning 20:06h UTC and ending 21:15h UTC. In this interval (~ 1 hour), the amount of rainfall was 18.5 mm. Thus, it was found a total sulfate of 126.72 μg/l and total nitrate of 1034.16 μg/l, while that the total ammonium was 233.46 μg/l in this rainwater.

Figure 3 displays a good improvement in simulation results of sulfate and sulfur dioxide atmospheric scavenging. In this case, model represents about 25% of total sulfate founded in rainwater. However, a contribution of each wet deposition stage of process is similar than the previous event.

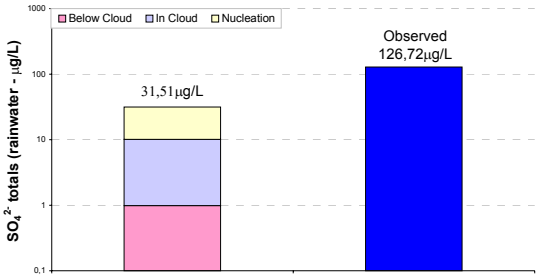

FIGURE 3 - SO_4^{2-} AND SO_2 SCAVENGING RESULTS FOR OCTOBER 9TH, 2002

For this event, it was observed that nitrate had the worst result than the other species studied (Figure 4). In this case, model represents only 2.5% of total nitrate in the analyzed rainwater.

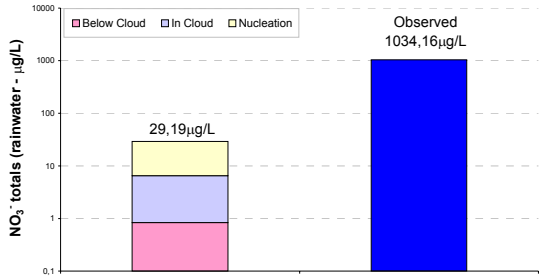

FIGURE 4 - NO_3^- AND HNO_3 SCAVENGING RESULTS FOR OCTOBER 9TH, 2002

Finally, the Figure 5 shows the result for the ammonium and ammonia atmospheric scavenging simulation. In this case, the model represents only about 20% of the ammonium total in the rainwater analyzed. Additionally, it was possible to verify the predominance of in-cloud scavenging processes, so that also could be observed for sulfate and nitrate scavenging.

5. CONCLUSIONS

A sensible improvement in simulation results for sulfur dioxide and sulfate scavenging was observed due the implementation of droplets gamma distribution in wet deposition code.

In general, the simulations for all chemical species studied had tended to underestimate the concentrations of these in rainwater, when compared with observed data in both events analyzed.

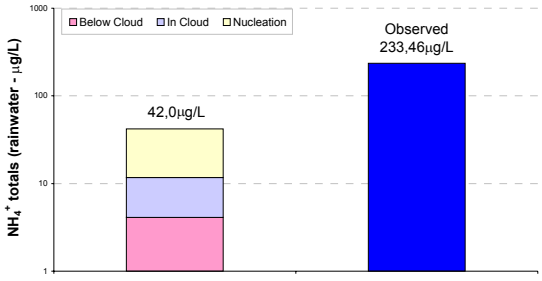

FIGURE 5 - NH_4^+ AND NH_3 SCAVENGING RESULTS FOR OCTOBER 9TH, 2002

Results lead to attribute an in-cloud scavenging dominance (between 89% and 99%), due the great difference in clouds structure and microphysics that develops in Amazon region during biomass burning period and the ones that develops in a metropolitan region of middle latitudes.

Comparing in-cloud and below-cloud results for the two studied events, results shown a great similarity between them. However, it can be observed for October 9th, the percentage associated with below-cloud scavenging was still lesser (about only 2%).

Comparisons amongst species, sulfate modeling shows better agreement with observed data, while ammonium and nitrate shows modeled amounts underestimated when compared with concentrations found in rainwater.

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