Investigation of the processes shaping the aerosol number distribution

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Adapted from a Lab exercise taught by Peter Tunved/Christer Johansson
during the 2016 Air Quality course, at the Stockholm University

Overview

As the lifetime of the aerosol in the troposphere is comparably short, properties of the aerosol, such as the aerosol number size distribution will exhibit a large degree of spatial and temporal variability. This is evident by observing time series of aerosol properties at observational sites, but also comparing aerosol properties in measurement sites being very close to each other. Proximity to and nature of sources and meteorological conditions (e.g. vertical mixing, precipitation, cloudiness etc.) are key factors in shaping the aerosol size distribution. In many ways, the aerosol size distribution serves as an aerosol fingerprint regarding what processes have been active (e.g. in-cloud processing, wet removal, nucleation, condensation growth, primary emissions etc.).

During this exercise you will familiarize yourselves with the properties of the aerosol size distribution as observed in two GoAmazon sites. This exercise will aid in the understanding of the factors controlling the evolution of the size distribution by comparing properties of the aerosol size distribution at different locations and over different timescales.

You will use the CALM model to develop more insight in the importance of different dynamical processes shaping the aerosol size distribution. This will include studies of coagulation, nucleation, condensation and dry deposition. Special emphasis will be on coagulation and new particle formation. The goal is to understand how and over which size ranges these different dynamical processes have their largest influence and why, and how this varies between different environments.

Task

At your disposal you will have one year of aerosol number size distribution data (2014, continuous 5-min resolved data) from two different stations around Manaus, namely T2-Tiwa (5km downwind of Manaus) and T3-Manacapuru (80km downwind of Manaus). T2 site receives the fresh Manaus plume, while T3 site is influenced by a more aged plume. The location of the sites is presented in Figure 1. This dataset has already been studied and the plots below are from Cirino et al., 2018 (Atmos. Env. – in press).

Figure 1: Location of observation sites

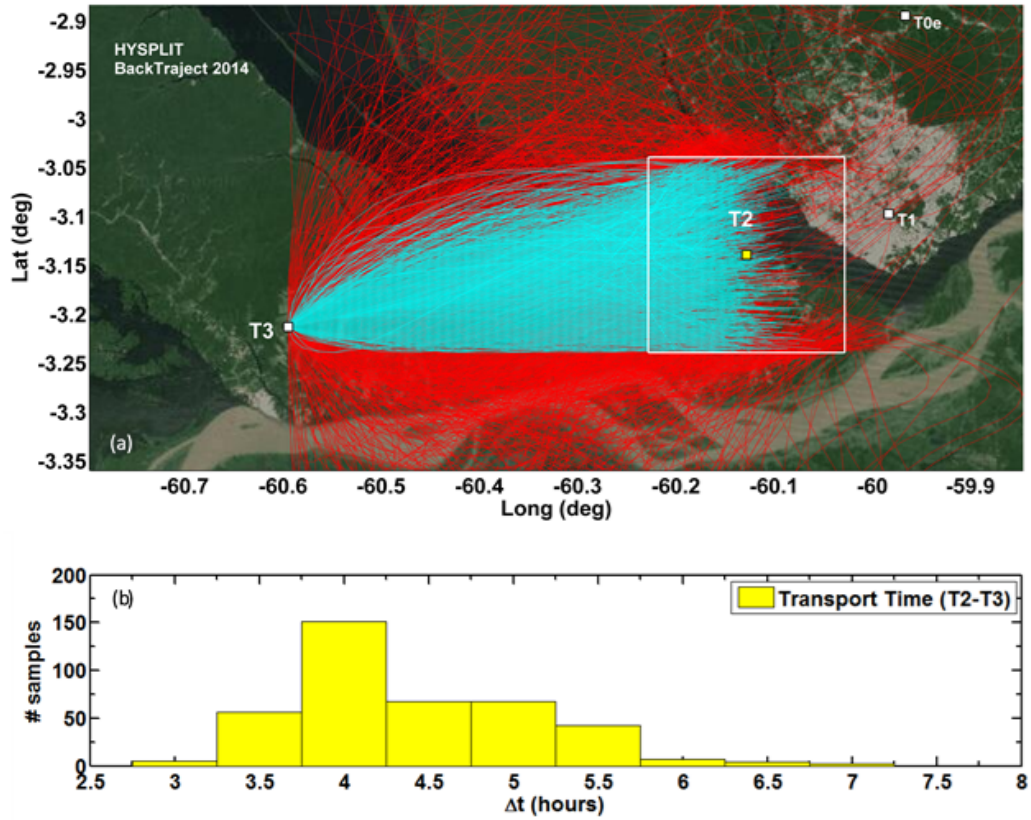

Figure 3. Panel (a) shows the back trajectories calculated for studying the plume evolution. All trajectories eastward from T3 (70 km downwind), inside the boundary layer (altitude < 1000 m) and reaching a 20x20 km box around T2 (8 km downwind) are represented in red. A subset of straight trajectories without precipitation is represented in cyan. The trajectories were simulated as arriving at 100 m above T3. Panel (b) shows the histogram of air mass transport time between T2 and T3 based on the trajectories shown in cyan (a).

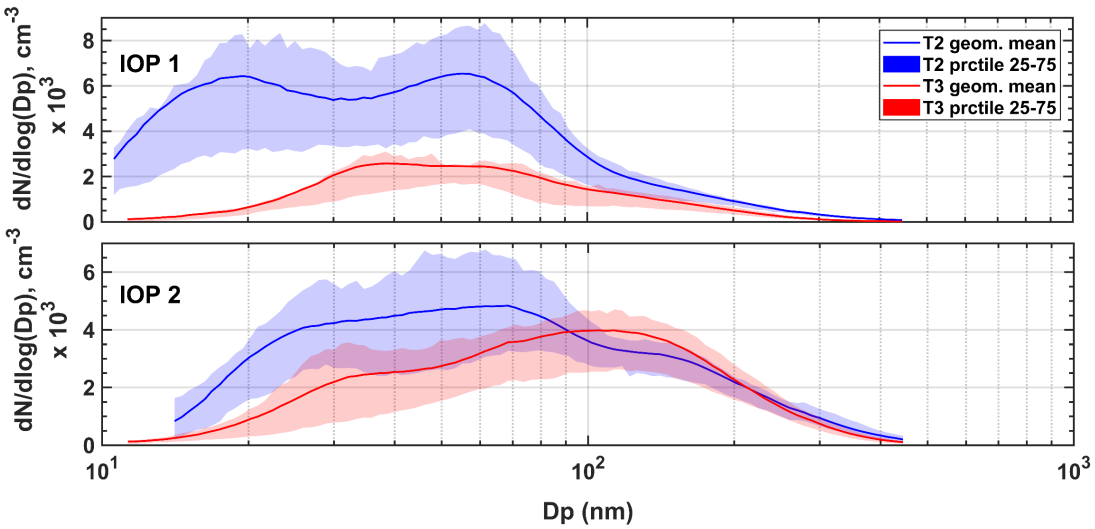

Figure 4. Mean particle number size distribution at T2 (blue line, 8 km downwind) and T3 (red line, 70 km downwind) for the wet (IOP 1, top) and dry (IOP 2, bottom) seasons, considering selected periods under the influence of the Manaus urban plume. Error bars represent standard deviations. The blue and red shaded areas show 25-75 percentiles at T2 and T3 sites, respectively.

IOP1					
T2			T3		
N(cm-3)	GSD	Dg(nm)	N(cm-3)	GSD	Dg(nm)
3070	1.57	18.2	105	1.19	33.3
2818	1.52	56.8	1576	1.80	52.3
416.1	1.46	163.7	138	1.37	162.4
IOP2					
T2			T3		
N(cm-3)	GSD	Dg(nm)	N(cm-3)	GSD	Dg(nm)
1343	1.43	25.4	1024	1.52	35.5
1973	1.52	59.0	200	1.26	69.1
1308	1.59	152.0	2088	1.65	119.8

Table 1: Modal properties for the average number size distribution observed at GoAmazon. Each distribution consist of three modes, each of which is defined by number concentration (N), geometrical standard deviation (GSD) and mean modal size (Dg).

Analysis procedure

Below a number of steps you should follow to reach the goal of this exercise. A useful hint is to regularly save your results you are satisfied with, and doing so also giving the file that you save a useful and informative name that will help you in the coming analysis. A stringent naming convention is of crucial importance in order to be able to navigate through your results when you prepare the report. Feel free to play around with the tools; they can't break (although the can likely provide some errors on occasions). You are encouraged to discuss the problems with your fellow students and supervisors.

1.) Make a directory where you will save your figures.
2.) Download the GUI and the datasets from here:

http://lfa.if.usp.br/ftp/public/2015SummerCourse/

3.) Unzip the GUI.zip file, and start a MATLAB session inside that folder

4.) Let's study how the aerosol is affected by different dynamical processes, including nucleation, coagulation, condensation and dry deposition. All these processes affect the size distribution in their own characteristic way and over characteristic time scales, depending on meteorology, availability of condensing and nucleating gases, and last but certainly not least, but the actual size distribution properties. In this part of the exercise you will study how the aerosol size distribution typically observed in at two different environments vary with respect to different dynamical processes, i.e. Urban and remote Arctic. The model relies on input of so called modal parameters, which is a mathematical representation of the size distribution by a number log-normal distributed modes. The modal parameters are shown above (Table 1), and represents the aerosol size distribution properties observed on average at T2 and T3 during the IOP1 and IOP2.

5.) In the command prompt of Matlab, type "dynamics_toolbox" and press enter.
   a. Starting with T2, write the modal parameters as written in table 1 into the GUI. Set N in mode 4 to zero (you only have three modes in the table above, so the last one is not needed). Press "Calculate size distribution". Your size distribution will appear in the figure below.
   b. in the processes section you can activate and deactivate different processes. Deactivate everything but coagulation. In run parameters, set run time to 4.6 hour, starting time to 00, latitude to -3, longitude to -60 (i.e. Manaus). Press "Launch calculations". Three important figures will be generated: Figure 10 contains starting and ending size distribution, Figure 11 the size distribution time evolution, Figure 22 contains the number and surface evolution.
   c. Increase the number concentration in mode 1 to 7000 particles. Repeat the calculations.
   d. now increase also the number concentration in the largest mode to 1500 particles

   QUESTION: How does the distribution at T2 age with respect to coagulation? By altering the number concentration of smaller and larger particles, coagulation is substantially altered. Discuss why.

6.) Return to your original number concentration values as tabulated in table 1. Activate all processes, make sure nucleation is set to activation type nucleation. Deselect "run with user specified ..." under chemistry and set the concentration of SO_2 to 0.4 ppb. Run the model again.

   QUESTION: Do we produce any new particles? Do we produce new surface? Discuss why/why not.

7.) Increase the SO_2 concentration until significant new particle formation occurs. This might require several ppbs.

8.) Now, set SO_2 to 0.4 again. Change the number concentration to that of a clean environment (table 2 below). Run the model again.

QUESTION: How does the availability of precursor gases and abundance of pre-existing aerosols affect the efficiency of nucleation?

Table 2. Properties (position x_0, integral number concentration N_{CN}, width σ) of Aitken and accumulation modes from the double log-normal fit (compare to R^2) of the total particle size distributions. Values are given as annual means and subdivided into seasonal periods of interest as specified in Sect. 3.3 (compare also to Fig. 6). In addition, values for the position of the Hoppel minimum D_H as well as estimated average peak supersaturation in cloud $S_{cloud}(D_H,\kappa)$ are listed. The errors represent the uncertainty of the fit parameters. The error in $S_{cloud}(D_H,\kappa)$ is the experimentally derived error in S.

Season	Mode	N_{CN} (cm^{-3})	κ	x_0 (nm)	σ	R^2	D_H (nm)	$S_{cloud}(D_H,\kappa)$ (%)
Year	Aitken	397 ± 31	0.13 ± 0.03	69 ± 1	0.44 ± 0.02	0.99	97 ± 2	0.29 ± 0.03
	accumulation	906 ± 29	0.22 ± 0.05	149 ± 2	0.57 ± 0.01			
LRT	Aitken	231 ± 8	0.14 ± 0.04	67 ± 1	0.63 ± 0.01	0.99	109 ± 2	0.23 ± 0.02
	accumulation	232 ± 10	0.28 ± 0.08	172 ± 1	0.51 ± 0.01			
Wet	Aitken	246 ± 9	0.13 ± 0.02	70 ± 1	0.53 ± 0.01	0.99	112 ± 2	0.22 ± 0.02
	accumulation	145 ± 8	0.21 ± 0.05	170 ± 2	0.42 ± 0.01			
Transition	Aitken	405 ± 24	0.14 ± 0.02	65 ± 1	0.42 ± 0.01	0.99	92 ± 2	0.34 ± 0.03
	accumulation	668 ± 24	0.24 ± 0.04	135 ± 1	0.53 ± 0.01			
Dry	Aitken	483 ± 49	0.13 ± 0.03	71 ± 2	0.42 ± 0.03	0.99	97 ± 2	0.29 ± 0.03
	accumulation	1349 ± 47	0.21 ± 0.04	150 ± 2	0.58 ± 0.01			

From: Pohlker et al., ACP 2016. Note: width = 1 + σ

9.) Repeat step 5-7, but now use Zeppelin size distribution. Lat/lon are the same.

QUESTION: How does nucleation efficiency differ between the clean environment compared to the polluted urban environment? Is it reasonable, do you think, to have concentrations of 0.4 ppb SO2 in the clean Amazon? Try to find out how low the SO2 concentration can be without removing nucleation completely.

All these tasks will require educated guesses and estimates; however, the important thing is not to write a paper on the aerosol size distribution. Rather, the tools supplied should be used to build a basic understanding on temporal and spatial variability of the aerosols size distribution, and of course also understand the importance of the controlling mechanisms.

10.) Now lets study the plume evolution. Go back to the simulation of the T2 case, but do a simulation of just 4.6h (average flight time). Set start time to 15h UTC (i.e. 12h local time). Write a Matlab script to take the size distribution at the end of the simulation and compare it with that measured at T3 in the same plot. Try turning on/off the different processes, and try also changing the concentrations of gas precursors.

QUESTION: Are you able to reproduce what is observed at T3 after the day-time evolution of the plume from T2 to T3? Which processes are most important? What did you have to change about the gas precursors? Is there evidence of new particle formation? Are those inorganic or organic?

Home exercise:

Investigate what happens with the plume evolution from T2 to T3 for different levels of SO2 at T2. Is nucleation more likely to occur for higher or lower concentration of SO2? Why?

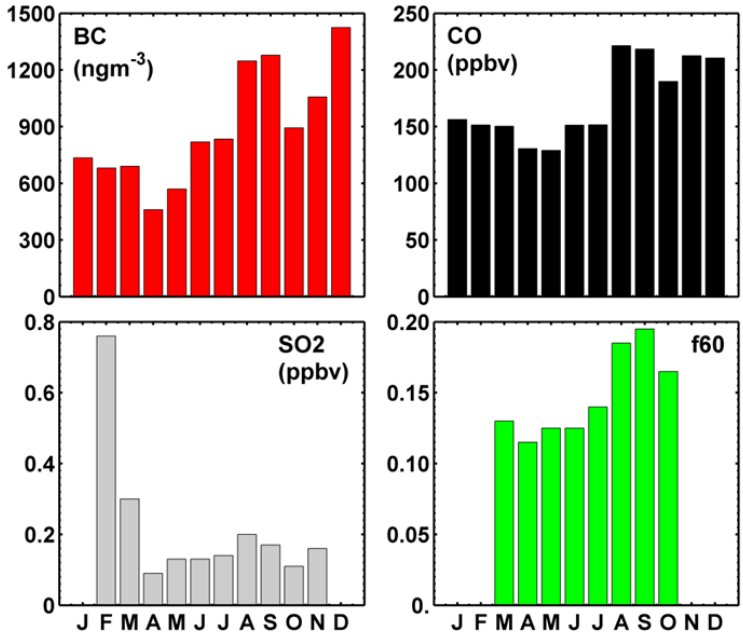

Figure. Average monthly BC, CO, SO_2 and f60 at T2 site.

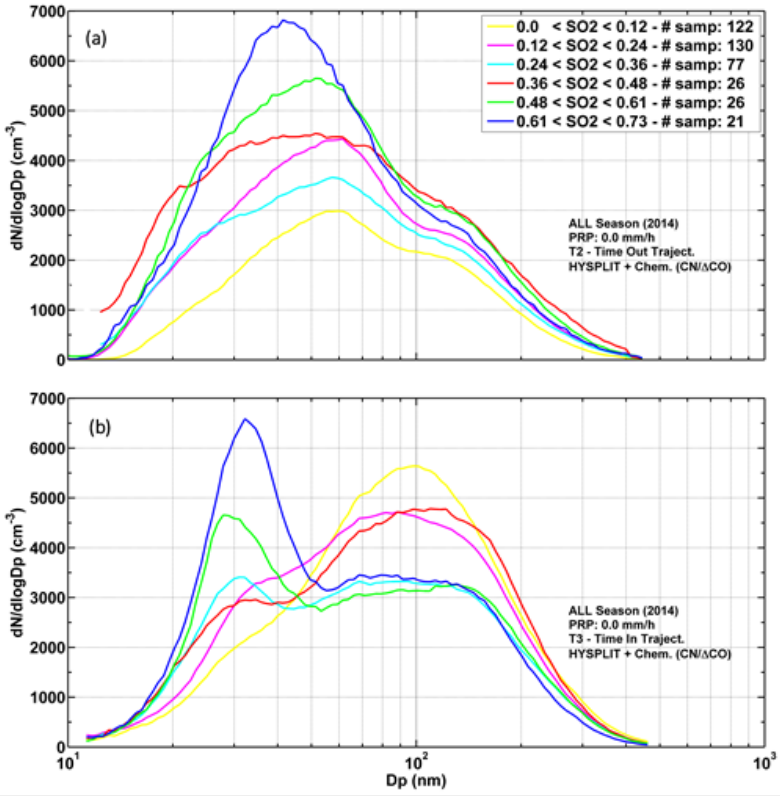

Figure. Aerosol size distributions measured at T2 and T3 as a function of the increase of SO_2 concentration in the atmosphere during the events of downwind transport of pollution from Manaus to Mancapuru. Data is grouped by the concentration of SO2 at T2, ranging from 0 to 0.73 ppbv. Events with precipitation were excluded.

Table S2. Changes on the concentration of aerosol chemical components resulting from evolution of the Manaus plume between the sampling sites (T2 and T3) during IOP 1 (1 February to 31 March 2014) and IOP 2 (15 September to 15 October 2014). Concentrations are shown at departure and arrival times of air masses leaving the T2 site and reaching the T3 site.

Chemical Species	T2 Concentration at departure		T3 Concentration at arrival		T3/T2 Relative Factor	
-	IOP 1	IOP 2	IOP 1	IOP 2	IOP 1	IOP 2
OA - μgm^{-3}						
Median	1.9	7.0	1.9	8.9	1.0	1.3
Average	2.1	7.3	1.9	9.5	0.9	1.3
Std. Deviation (±)	1.0	1.9	0.9	3.2	-	-
SO4 - μgm^{-3}						
Median	0.3	1.4	0.2	1.9	0.5	1.4
Average	0.4	1.6	0.2	2.1	0.5	1.4
Std. Deviation (±)	0.2	0.5	0.1	1.0	-	-
NO3 - μgm^{-3}						
Median	0.1	0.3	0.0	0.2	0.4	0.6
Average	0.1	0.3	0.0	0.2	0.4	0.6
Std. Deviation (±)	0.0	0.1	0.0	0.1	-	-
NH4 - μgm^{-3}						
Median	0.1	0.5	0.1	0.5	0.8	1.1
Average	0.1	0.5	0.1	0.6	0.7	1.2
Std. Deviation (±)	0.1	0.2	0.0	0.2	-	-