Aerosol complexity in megacities: From size-resolved chemical composition to optical properties of the Beijing atmospheric particles

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[1] Megacities need adapted tools for the accurate modeling of aerosol impacts. For this purpose a new experimental data processing has been worked out for Beijing aerosols as case study. Size-resolved aerosol particles were extensively sampled during winter and summer 2003 and subsequently fully chemically characterized. The product is an aerosol model presenting a new particle pattern (mode number, size and chemistry) without any prerequisite constrain either on the mode number or on each mode chemical composition. Six modes were found and five of them consistently appear as internally mixed particles organized around a black carbon or a dust core coated by organic and/or inorganic material. Data were checked by robust comparisons with other experimental data (particle number, sunphotometer-derived derived data). We found the presence of two accumulation modes in different internal mixing and optical calculations show that the Beijing aerosol single scattering albedo (ω_o # 0.90) is significantly higher than expected. Such an approach would allow realistic modeling of atmospheric particle impacts under complex situations. **Citation:** Roger, J.-C., B. Guinot, H. Cachier, M. Mallet, O. Dubovik, and T. Yu (2009), Aerosol complexity in megacities: From size-resolved chemical composition to optical properties of the Beijing atmospheric particles, *Geophys. Res. Lett.*, *36*, L18806, doi:10.1029/2009GL039238.

1. Introduction

[2] Projections show that an important part of population growth will occur in large cities and the picture is particularly straightforward for Asia where the number of megacities (cities over 10 millions inhabitants) is about to jump from 3 in 1975 to 15 in 2015. Such pollution hotspots are likely to exert impacts on the atmospheric environment at all scales. Compiled data for cities in Asia [*Baldasano et al.*, 2003] shows a ''common denominator'' of atmospheric pollution primarily related to very high concentration of particles, which are probably emitted by multiple and intense anthropogenic sources, with possible dust inputs. Numerous gas to particle interactions probably create there a complex mixture of primary and secondary particles whose diversity is far beyond that encountered in developed regions.

[3] Previous works [*Gurjar and Lelieveld*, 2005; *Intergovernmental Panel on Climate Change*, 2007] have strengthened the need to better assess the aerosol impacts caused by megacities. The issue is even more important in fast developing countries like China where particle emissions likely affect the regional climate [*Menon et al.*, 2002]. Hence, in order to model the various impacts of megacity aerosols, and to anticipate their future trends, aerosol emissions and transformations have to be accurately characterized, and for this purpose specific tools need to be created. A key step relies on the availability of an aerosol model that reflects the aerosol phase complexity (e.g., the diversity of sources and the intensity of particle transformation processes) while remaining flexible enough to account both for site diversity and their irreversible changes accompanying their development. Contrarily to urban aerosol models constructed for developed countries, this megacity aerosol model is likely to host several particle modes in external or internal mix and such a complexity has not yet been adequately explored. This new aerosol model is expected to benefit to various fields from air quality management to local radiative forcing and climate impact assessments. Additionally it would enrich aerosols studies based on satellite data inversion.

[4] Beijing (China) is experiencing a rapid development and is now a megacity holding more than 16 millions inhabitants. Since 1998, the municipality is engaged in an ambitious clean-up, and serious efforts have been made to identify and phase out the most important sources of pollution. In 2003, we conducted two intensive field experiments in Beijing during the contrasted winter and summer seasons: Pek1 in January and Pek2 in August-September. Aerosols were sampled on quartz and teflon filters, for carbon and ion analysis, respectively. Allocated Dekati cascade impactors working at a flow rate of 30 $l.min^{-1}$ allowed particle separation into 13 aerodynamic class-fractions (Dp50 in the range 0.03 – 10 μm), thus offering a satisfactory size-resolution in the submicron range. Nineteen sets sampled over 6 hours each, were obtained. For each individual impactor stage, the analytical and subsequent chemical closure protocols developed by *Guinot et al.* [2007a] were applied. Then, it has been possible to obtain the chemical distribution of the main aerosol components (black carbon BC, organic carbon OC, and major ions: sulfate SO_4^{2-}, nitrate NO_3^-, ammonium NH_4^+, and dust). Based on these experimental results, a new data processing enabled to work out an aerosol model that accounts for the microphysical and chemical complexity of Beijing aerosol phase. As detailed hereafter, the model comprises a lognormal distribution showing several modes,

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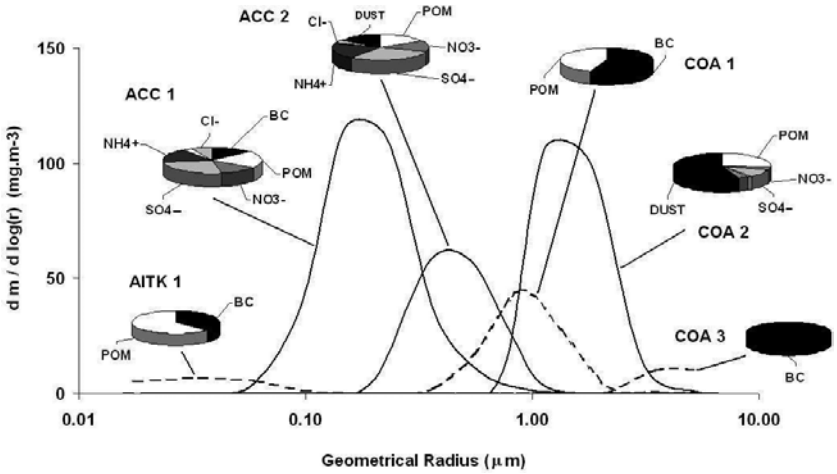

Figure 1. General scheme of the aerosol size-distribution observed for Beijing aerosols (particles sampled with Dekati 13-stage cascade impactors in the range 0.03–10 μm).

each mode being fully defined by its lognormal parameters and its chemical composition. Using adequate algorithms [*Roger et al.*, 2006], these parameters allow retrieval of other properties (aerosol number and optical properties) some of them being compared to direct measurements, thus insuring quality and consistency of data.

2. Mass and Number Size-Distribution of Aerosol and Main Chemical Components

[5] A new process has been developed and applied to aerosol mass and chemical composition in order to sort out all the various aerosol modes and their chemical composition. Based on the size-resolved impactor datasets, and without any pre-defined assessment of mode number, chemical mass distributions consistently end up with 6 modes: one mode in the Aitken size-range (0–100 nm in diameter), two modes in the accumulation size-range (0.1–1.0 μm), and three coarse modes (>1.0 μm). The complex aerosol source pattern in Beijing supports the existence of additional modes compared to models elaborated for urban aerosols of developed countries [*Seinfeld and Pandis*, 1998; *Mallet et al.*, 2003; *Van Dingenen et al.*, 2004, and references therein], and thus reinforces the need of a new approach to account for aerosol complexity in megacities located in developing megacities. Data consistency firstly allows us to assume that two different types of primary particles, either BC or dust, are present in each mode. We suggest that they may serve as cores for condensation or coagulation processes. Then, the next step is to infer the complete chemical composition of each mode. To that end, in each impactor data set, the distribution of major molar associations is carefully scrutinized. $(NH_4)_2SO_4$ and NH_4NO_3 distribution gives consistent evidence for two fine (accumulation) modes organized around a BC core and a dust core separately. For coarse aerosols, dust particles only are found associated to these inorganic ions. Beside these three modes dominated by ion species, there is a clear evidence for three other modes all of them organized around a BC core. One mode is situated in the Aitken size-range while the two others are found in the coarse mode. From this scheme, using an iterative method and a matrix of relative dependence, it is possible to retrieve the distribution of all major aerosol chemical components throughout the six modes, including particulate organic matter (POM) and dust, respectively estimated from OC and Ca^{2+} as suggested by *Guinot et al.* [2007a]. It may be seen in Figure 1 that five out of the six modes could be made of heterogeneous particles whereas only one appears mostly homogenous in nature (more than 95% of BC). The mode related to ultra-fine particles is termed Aitk1. Although not fully determined due to the impactor geometry limitations (about 2/3 of POM and 1/4 of BC are defined) this mode is overwhelmingly dominated by carbonaceous species and is expected to originate mainly from traffic primary particles or from the coagulation of nucleation particles. Both accumulation modes show an internal mix of inorganic salts and POM, with a core of either BC (for the smallest mode Acc1) or dust (for the biggest mode Acc2). A possible origin of Acc2 particles might reside in the background of fine loess particles originating from the plateau West of Beijing and from resuspended fine dust. In the coarse fraction, the main mode (Coar2) is also an internal mix with an important dust core (representing more than 50% of the mass). The heterogeneous nature of this mode is probably the result of dust-acidic gas interactions previously mentioned for the Beijing aerosol [*Guinot et al.*, 2007b, and references therein]. The two other coarse mode Coar1 and Coar3) are almost pure carbonaceous particles and have probably to be related to truck emissions or industrial inputs [*Cachier*, 1998]. They represent a non-negligible portion (25% and 10% respectively) of the aerosol coarse fraction either in winter or in summer.

[6] The mass size distribution is then fitted by a lognormal law and distribution is obtained for dry particles as follows. When atmospheric humidity (RH) is higher than 80%, particle radii of the accumulation modes and of the coarse modes (retrieved from impactor data) have been shown to be RH dependent. However as expected, the smallest particles do not exhibit same RH dependency probably because of the significant evaporation of most of their water content under the vacuum met on the first stages (100–200 mbar). The RH dependency follows the Hänel law that has been therefore used to retrieve the dry aerosol radius. The dry aerosol mass distribution (Table 1) clearly enlightens the dominance of the two Acc1 and Coar2 modes. The mass distribution does not

Table 1. Mean Mass $\langle m \rangle$ ($\mu g.m^{-3}$) and Standard Deviation for Each of the Dry Aerosol Modes in Winter (Pek1) and Summer (Pek2)[a]

		Pek1		Pek2	
		$\langle m \rangle$ (std)	$\langle mc \rangle$/%	$\langle m \rangle$ (std)	$\langle mc \rangle$/%
Aitk 1	BC	0.37 (0.14)	0.11/30%	0.36 (0.10)	0.10/28%
Acc 1	BC	5.24 (3.12)	0.85/16%	4.84 (1.76)	0.73/15%
Acc 2	dust	1.79 (1.18)	0.40/22%	2.49 (2.10)	0.83/33%
Coa 1	BC	0.79 (0.62)	0.50/63%	0.37 (0.27)	0.28/76%
Coa 2	dust	2.96 (2.16)	1.48/50%	5.75 (2.43)	4.50/78%
Coa 3	BC	0.16 (0.11)	0.15/94%	0.14 (0.07)	0.13/93%

[a]The chemical nature (BC or dust) of the probable core is indicated; $\langle m_c \rangle$ (%) stands for the core mass and percentage. Total number of samples: 19.

Table 2. Dry Particle Radius for Each Mode, Respectively for the Volume ($\overline{r}_v$) and the Number ($\overline{r}_n$) Size Distributions During the Two Seasons[a]

	$\overline{r}_v$		$\overline{r}_n$		STD	
	Pek 1	Pek 2	Pek 1	Pek 2	Pek 1	Pek 2
Aitk 1	0.035	0.037	0.013	0.014	1.7	1.8
Acc 1	0.15	0.15	0.065	0.075	1.7	1.6
Acc 2	0.42	0.44	0.295	0.300	1.4	1.4
Coa 1	0.97	0.95	0.505	0.540	1.6	1.6
Coa 2	1.15	1.02	0.60	0.660	1.6	1.5
Coa 3	4.56	3.87	3.330	2.730	1.4	1.4

[a]Standard deviation unchanged by the mass-to-number conversion.

exhibit significant changes between winter and summer, particularly for the combustion derived modes formed around a BC core (Acc1, Coar1 and Coar3). The lognormal mass distribution $d\,m/d\,\log(r_v)$ is fully characterized for each mode by the mass, the mean radius and its standard deviation. Surprisingly, the dry aerosol radius does not exhibit any seasonality (Table 2) which might denote the presence of a background of pollution all year long, with likely smoothing effects during the two opposite seasons (photochemical production in summer and thermal condensation in winter).

[7] For a purpose of validation, particle number concentration N ($p.cm^{-3}$) was calculated from mass size-distributions following *Mallet et al.* [2003]. Particle number is obtained for each mode, using particle density (accounting for the heterogeneous nature of particles and weighing the proper density of each chemical component) and the lognormal mass-distribution parameters (r_v, standard deviation σ). Estimated N values are shown against direct optical measurements obtained from CNC/DPS and GRIMM optical counters (Figure 2) and comparison appears satisfactory. Mass size-distributions may be transformed into number size-distributions and radius data are shown in Table 2. Comparison of measured and retrieved volumic radii of the different modes satisfactorily highlights a good agreement between the two datasets (in auxiliary material).[1] As a matter of comparison too, the procedure used to sort out the 6 log-normal modes from the impactor concentrations has been applied to the monthly aerosol volume data available in the AERONET database (http://aeronet.gsfc.nasa.gov/). Similarly, no pre-requisite parameters were set regarding the number of modes, their size or their amplitude. AERONET data strikingly provide same information as impactor data and repeatedly enlighten 2 accumulations modes, and 3 coarse modes (as expected, the very small Aitk1 mode is not easily extractable from AERONET data). The two accumulation modes likely correspond to the Acc1 and Acc2 modes experimentally defined (Table 2). For coarse particles, the smaller AERONET coarse mode may stand for Coa1 and Coa2 that are of very similar size and the second AERONET coarse mode to Coa3 whereas the fourth one corresponding to particles over 10 μm in diameter is not sampled by the impactor. The two accumulation mode radii are comparable to those retrieved from AERONET volume size-distributions (Figure 3a). Mean Acc1 radius (0.14 and 0.15 for Pek1 and Pek2 periods) fairly compares with the 0.15 value found with our sampling-derived procedure. For Acc2 too, AERONET mean radius (0.40 and 0.51) show satisfactory agreement with experimental values (0.40 and 0.42 for Pek1 and Pek2) recalling that sunphotometer data apply to ambient (and not dry) aerosols. Data suggest too that high RH (prevailing in summer) may cause a significant increase of Acc2 particles whereas Acc1 particles appear less prone to water capture.

3. Derived Aerosol Optical Properties

[8] Aerosol radiative effects primarily rely on their single scattering albedo ω_o and their asymmetry parameter g. It has been shown recently that using adequate algorithms, these two parameters could be retrieved based on the chemical composition of each aerosol mode [*Roger et al.*, 2006]. Calculations may account for the heterogeneous nature of particles which are also considered as spheroids [*Dubovik et al.*, 2002]. Evaluated complex refractive indexes show quite similar values during the two experiments for both the real and the imaginary parts which allows assessing a single value over the whole (Pek1 and Pek2) experiment (Table 3). Calculated ω_o and g values at 550 nm and at two different RH for the total aerosol and for each of the 6 identified modes are shown too in Table 3. A comparison with values retrieved from AERONET data was also conducted and the agreement between both data sets appears satisfactory (Figure 3b). As expected, the two accumulation modes (Acc2 and Acc1 particularly) are the most radiatively important modes as

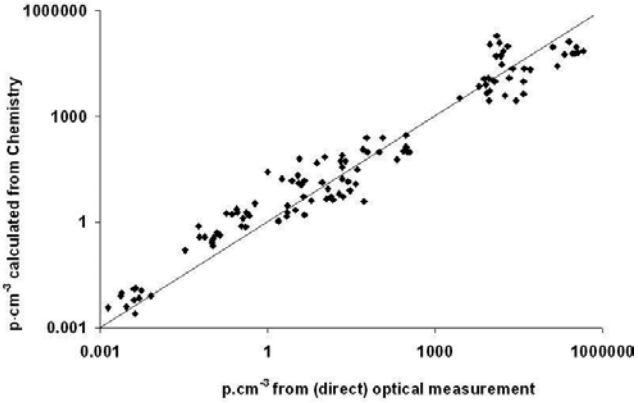

Figure 2. Comparison of direct measurements of particle numbers (CNC/DPS (TSI) and GRIMM optical counters) versus calculated concentration numbers retrieved from mass distributions obtained by impactors ($y = 0.88x + 0.20$; $r^2 = 0.85$).

[1]Auxiliary materials are available in the HTML. doi:10.1029/2009GL039238.

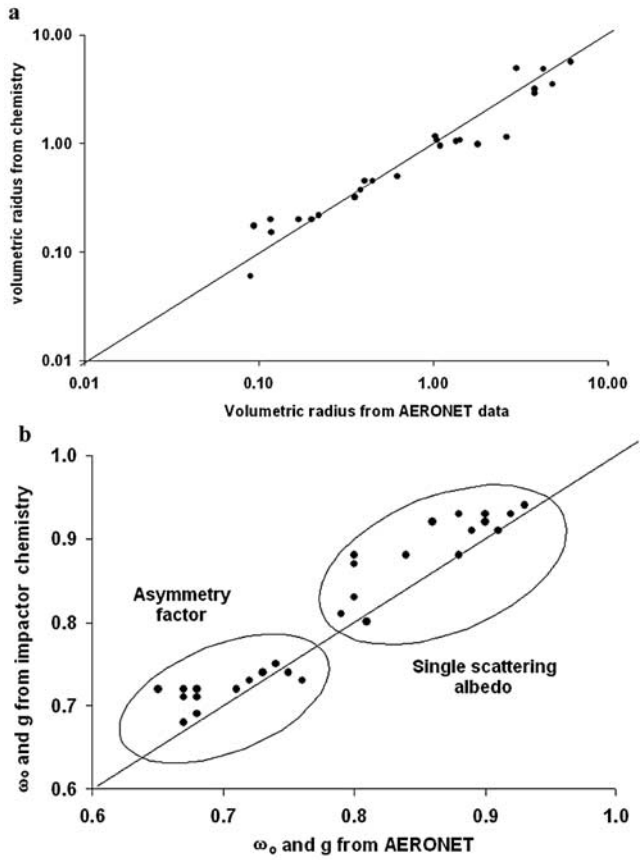

Figure 3. Comparison of AERONET and impactor chemical measurement retrievals. (a) Volumic radius of the different aerosol modes ($y = 0.90x + 0.008$ $r^2 = 0.87$). (b) Single scattering albedos and asymmetry factors of the total aerosol at 550 nm.

they are shown to drive 84% and 80% of the scattering and the absorption, respectively, whereas the role of the two extreme size-modes (Aitk1 and Coar3) is not significant (Table 3). Our optical calculations show that the scattering albedo ω_o of Beijing aerosol is close to 0.90, which is significantly higher than those obtained for other megacities, such as Mexico (ω_o # 0.75) (Loeffler et al., unpublished material), New Delhi (mean ω_o # 0.68) [*Ganguly et al.*, 2006] or Kanpur (ω_o # 0.76) [*Tripathi et al.*, 2005]. Such a high ω_o value, despite the significant BC content in the Beijing aerosol, may be partly explained by the thick coating of scattering species onto BC particles. BC, the main absorbent species, is thus likely to lose adsorption efficiency and this loss produces an increase of ω_o. Furthermore, significant amounts of fine dust particles are also likely to drive ω_o towards higher values. As for the asymmetry parameter g, the value obtained in our work (0.70) is significantly higher than common values obtained for anthropogenic aerosols. The presence of a significant coarse dust component may explain the importance of scattering in the forward peak. These findings point to the need for an accurate updating of these parameters to be used for the assessment of aerosol optical and radiative properties in Chinese urban areas.

4. Conclusion

[9] Complex situations such as those encountered over Megacities need adapted tools for the accurate modeling of aerosol emissions and transformations. For this purpose a new experimental data processing has been elaborated and validated through various inter-comparisons for the Beijing aerosol and is presented here as a case study. The process produces the distribution of the aerosol particle mass-concentration over several modes which total number is not a-priori assessed. Each mode is defined by its lognormal distribution parameters and its full chemical composition comprising the main components: black carbon (BC), particulate organic matter (POM) and major ions (SO_4^{2-}, NO_3^-, NH_4^+, Ca^{2+}, Cl^-). The methodology is likely to apply to other megacities at locations where different intense and natural sources, produce high concentrations of particles and precursors, in addition to different atmospheric gas-to-particles processes. The original scheme obtained for Beijing aerosol shows a complexity reflected in the presence of 6 distinct aerosol modes, a situation never encountered in western countries. From the strikingly consistent and repeatable results obtained, it may be inferred that the same sources impact Beijing city whatever the season, with somewhat variability in their relative intensity only. The main straightforward finding relies on the presence of two accumulation modes both bigger ($d \geq 600$ nm) than what is generally inferred for urban aerosols. Furthermore, 5 of the 6 identified modes are found as internal particles organized around a BC core or a dust core. They also point to the intensity of gas-to-particles. Retrieval of aerosol optical properties shows significantly high scattering albedo ω_o (# 0.90) and asymmetry parameter g (# 0.70) values in Beijing, underlining the need for appropriate aerosol studies in complex multi-source situations. The aerosol model may be used for local or regional modeling of aerosol transport and impacts, and

Table 3. Optical Properties of the Beijing Aerosol Modes Calculated at 550 nm[a]

	Refractive Index		ω_o		g		Contribution to the Total Aerosol	
	Real	Imaginary	Dry	Wet	Dry	Wet	Scatt.	Abs.
Aitk 1	1.65 ± 0.04	0.17 ± 0.07	0.62	0.71	0.27	0.30	4%	4%
Acc 1	1.60 ± 0.02	0.10 ± 0.03	0.89	0.92	0.66	0.70	60%	78%
Acc 2	1.54 ± 0.01	0.01 ± 0.01	0.98	0.99	0.75	0.76	24%	2%
Coa 1	1.73 ± 0.04	0.36 ± 0.06	0.53	0.54	0.88	0.89	1%	14%
Coa 2	1.55 ± 0.01	0.01 ± 0.01	0.97	0.98	0.71	0.72	10%	1%
Coa 3	1.85 ± 0.01	0.57 ± 0.01	0.59	0.59	0.89	0.90	1%	1%
TOTAL			0.86	0.91	0.69	0.73		

[a]Complex refractive index of the dry particles (uncertainty stands for standard deviation observed over the whole dataset), single scattering albedo (ω_o) and asymmetry factor (g) for two RH values (dry = 40% and wet = 80%). Contribution of each mode to the total scattering and absorption is also presented.

scientists constructing elaborated air quality models including particle chemistry in megacities may benefit from this work. In particular, due to its sensitivity in the submicron-size fraction, the model may be adequately used to run realistic abatement scenarios for prediction of radiative impacts and may help to develop appropriate pollution controls. The aerosol model may also serve as a first step to evaluate aerosol deposition in the respiratory track and the detailed chemical description of the fine modes might be considered as a useful tool too for research in the field of biological effects of the respirable aerosol [*Ramgolam et al.*, 2009].

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