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Aerosol regional background in Developing Countries

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To determine the aerosol regional background in three experimental sites in Brazil, South Africa and India modelled and measured surface concentrations of black carbon, organic carbon, sulphate, nitrate and ammonium were analysed.
The South American site is characterised by a seasonal variation of the observed aerosol components. Measured ammonium and sulphate compare well with the model results. The model overestimates the elemental and organic carbon concentrations in the dry season, probably due to overestimation of the biomass burning emission inventories. In India modelled and measured concentrations in the wet season are very similar in level. In the dry season the observed concentrations are much higher than modelled due to air masses passing over New Delhi before arriving at the site. The South African site is influenced mainly by the local anthropogenic sources, thus the model estimates are substantially lower than the measured levels. A strong underestimation of modelled NO_3 concentration is evident in all sites: in warm environment nitrogen shifts from the aerosol to the gas phase, and condenses on larger particles and the model does not take this into account.

1. Introduction

The atmospheric aerosol and its impact at regional and global scales are less investigated and understood in rapidly developing countries than in Europe or North America, countries where emerging economies are combined with serious air quality problems including particulate air pollution.
Regions in Brazil, India, China and South Africa were selected for intensive studies in the framework of the EUCAARI project to fill the gap of knowledge about chemical and physical aerosol properties in those areas (Kulmala et al., 2009). Existing datasets from ongoing and past projects and campaigns have also been collected to help characterising the aerosol regional background.
In this study the aerosol regional background for three of the regions of interest is investigated applying a global Chemistry Transport Model and combining the results with the long-term measurements collected during the project in 2008-2009. Model results have been previously compared to existing datasets to evaluate of the capability of the model of estimating the regional background concentrations over the areas.

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2. Method

The evaluation of the regional background for organic carbon (OC), black carbon (BC), sulphate (SO_4), nitrate (NO_3) and ammonium (NH_4) was carried out over Africa, South America and Asia using the Chemistry Transport Model TM5. TM5 is an off-line global model (Krol et al., 2005) that uses the meteorological data calculated by the ECMWF model. It has a spatial global resolution of 6°x 4° and a two-way zooming algorithm that allows resolving the regions of interest with a finer resolution of 1°x1°. Surface processes are globally treated on a resolution of 1°x1°. Photochemistry and aerosols are coupled in this version of the model. Particles, which are internally mixed and can contain sulphate, ammonium, and nitrate are described using a bulk approach, even though for the calculations of the scavenging processes they are implicitly assumed to be in the accumulation mode. The model treats BC and OC as primary aerosols and does not calculate the Secondary Organic Aerosols (SOA); however, following the suggestions from the AEROCOM exercise (Dentener et al., 2006) SOA are treated as primary and emitted in the model atmosphere. Sea salt and dust are described using lognormal distributions and are externally mixed. The emission inventories are from the Hemispheric Transport of Air Pollution exercise (TF HTAP, 2007). Although defined as "BC" emission inventories the inventories used in this work were constructed using emission factors predominantly based on thermal-optical measurements and therefore they represent a more elemental carbon-like carbonaceous compound rather than BC (Vignati et al. 2010). Elemental carbon and black carbon are operational definitions created by two types of measurement methods utilized for the quantification of soot, depending whether they take advantage of the refractory or the light absorbing properties, respectively. Thus it is more appropriate to compare the modelled "BC" with EC measurements.

The model has been applied for the year 2001 and output of monthly averaged surface fields are used for the analysis.

Measurements describing chemical properties of atmospheric aerosols in South America, Asia, and Africa and from sites representative of the regional background were collected from peer reviewed journals and field campaign archives and merged into one single dataset (Gilardoni et al., 2007). The dataset comprises of concentrations of organic carbon, elemental carbon, black carbon, sulphate, nitrate, chloride, potassium, calcium, magnesium, sodium, dust, sea salt, and aerosol mass. Ground platforms are used for the present model evaluation. Table 1 reports the definition of dry and wet seasons for the regions.

During the project measurements of sulphate, nitrate, ammonium, elemental carbon and organic carbon from Manaus (Brazil), Gualpahari (India) and Elandsfontain (South Africa) were collected.

Table 1 Dry and wet seasons

Regions	Dry	Wet
Africa	April-October	November-March
South America	June-October	November-May
India	November-April	May-October

The sampling site in Brazil is in a mostly pristine rain forest, 60 km north of Manaus, in the Central Amazonia (Gilardoni et al., in preparation). The measurements were collected continuously with 3 day average sampling, from February till June and from mid-August till the end of September. In India the sampling was performed in Gualpahari, an urban - background location 10 km from New Delhi from April 2008 till March 2009. The samples in Gualpahari were collected with a frequency of 1 day every 6 days, therefore the measurements were not continuous. The South African sampling site (Elandsfontein) is located 200 km to the east of Johannesburg. Data here reported were collected from March till October 2009; the samples were collected 1 day every 6 days, therefore also in this case not continuously (Laakso et al, 2009).

3. Results

Monthly averages of surface concentrations of black carbon, organic carbon, sulphate, nitrate and ammonium are sampled from the TM5 model output at 1°x1° resolution, representing the given site. They are reported in Figure 1 with the PM2.5 measurements collected during the project campaigns for the three EUCAARI experimental sites; the modelled concentrations are assumed to be in the accumulation mode, therefore they are comparable to the measured PM2.5 fraction.
Due to the scarcity of available observations, modelled surface concentrations of organic and inorganic compounds have been compared to measurements collected not only in the regions surrounding the EUCAARI sampling sites but also to other grand sites in the respective continents.

3.1 Manaus

At Manaus modelled BC, ammonium, OC and sulphate have a seasonal variation, being higher in the period July-December corresponding to the dry season. Ammonium and sulphate compare well with the model results, showing similar concentrations in both seasons. There are a few stations in South America with annual averaged SO_4 and NH_4 observations; although the stations are very close and fall within the same model grid the measurements show a great variability. Thus, it is not trivial to evaluate the model accuracy when the observations show such a scatter in the levels.
At Manaus the model overestimates the EC concentrations in the dry seasons. The same overestimation in the dry season is detected for OC. The peaks in modelled values correspond to the biomass burning seasons.The biomass burning emission inventories used in the model are constructed based on the climatological data (GFED) of the years 1997-2001 (van der Werf et al., 2004). In the last years the Amazonian area has experienced a constant reduction in deforestation; in 2008 the forested area was cut by 30% less then in the period 1997-2001 (http://www.inpe.br/ingles/index.php). As a consequence fewer fires are initiated and thus the GFED emission inventories might overestimate the concentrations for the site. It should be mentioned that for other sites in Amazonia (Cuiaba, Alta Foresta (Echalar et al., 1998)) where measurements were collected in the years before 2002, the comparison with model results showed an underestimation of the modelled BC concentrations. Deforestation in the years 2002-2004 reached the highest levels since 1996-97 with a consequent high burning activity; therefore the emission inventories might underestimate the emissions during the period 2002-2004.

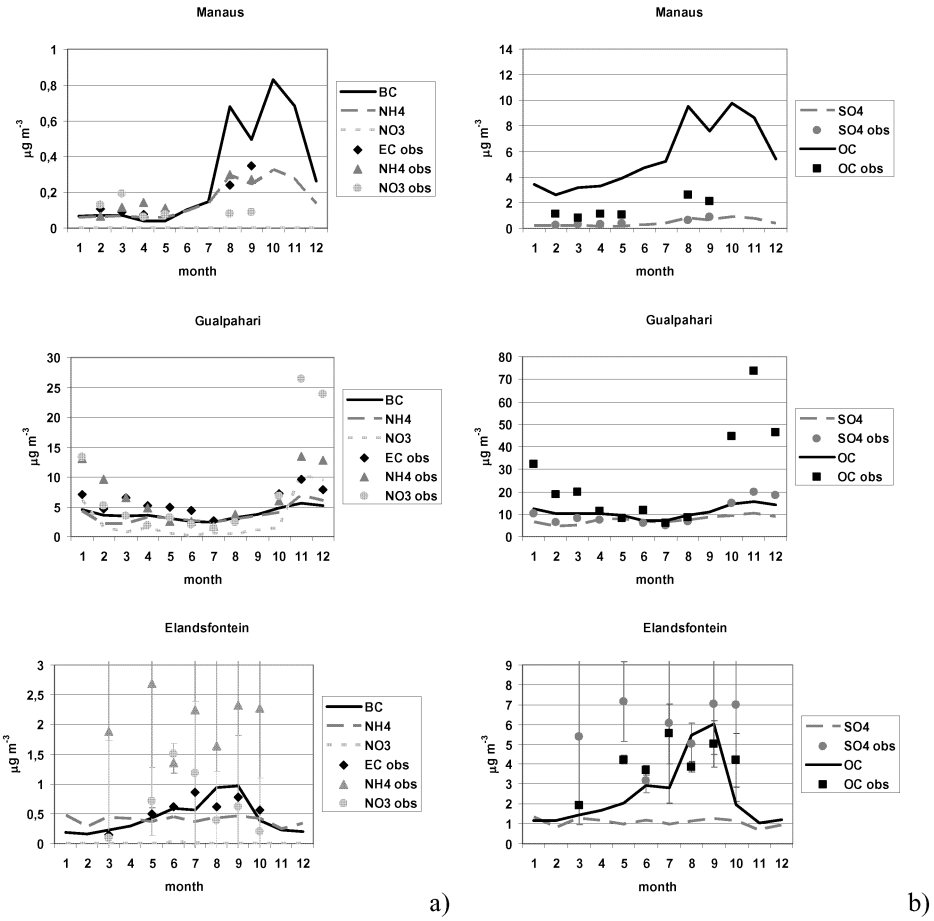

Figure 1. Time series of modelled and measured BC (EC observations), NH_4, NO_3 (a) and OC and SO_4 (b) concentrations ($\mu g\ m^{-3}$) at Manaus, Gualpahari and Elandsfontein.

A strong underestimation of modelled NO_3 concentration is evident: in warm environment nitrogen shifts from the aerosol to the gas phase, and condenses on larger particles. In TM5 the formation of coarse nitrate is not included, thus the modelled aerosol concentrations remain very small, since nitric acid remains in the gas phase.

3.2 Gualpahari

The comparison with other Indian stations shows that the model predicts fairly well BC, SO_4, and OC in the dry season, they are underestimated in the wet season. NO_3 is again strongly underestimated except a few cases in the wet season. Ammonium is slightly overestimated.

In Gualpahari (India) the model predicts a seasonal variation with minimum values in the wet (May-October) and maximum in the dry (November-April) season, although the increases in the dry season are not pronounced, except for nitrate. Observations and model show similar levels in the wet season for all compounds. The concentrations rise during the dry season especially for OC and nitrate in the period November-January.

Analysis of air mass trajectories has revealed that during the wet season (May-August) the air masses came from the South, while during the remaining months they came from the North, therefore the urban emissions from New Delhi influence the atmospheric concentrations of the air arriving to Gualpahari and the resulting aerosol levels may not be representative of the regional background. On the other hand the emission inventories may not contain the correct values for New Delhi.

3.3 Elandsfontein

The dataset contains measurements of Cape Point (South Africa), which is a regional background site. The comparison is done for SO_4 and NH_4. Sulphate is underestimated by the model in the wet season and ammonium only slightly overestimated. Therefore we might expect similar behaviour in Elandsfointein.

Modelled BC and OC have a peak of 1 and 8.5 μg m^{-3}, respectively, and generally large concentrations in the period June-August, corresponding to the most intense biomass burning period, while sulphate and ammonium show a quite flat behaviour around the year. The concentrations are quite high, with a large mass fraction in the coarse mode for EC, OC, and nitrate. Modelled nitrate is expected to be strongly underestimated. The site is influenced by the local anthropogenic sources, especially from coal-based power production, petrochemical industry and the megacity of Johannesburg: therefore it is generally not a background site. However when the air masses came from the NE-E-SE sector they were only little contaminated by pollution sources and the concentration levels were much lower, as seen in the figure for June, and comparable to the modeled background, although only one sample (8 June 2009) was available from the clean sector (Laakso et al, 2009).

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