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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A comprehensive modelling way for assessing real-time mixings of mineral and anthropogenic pollutants in East Asia" *by* F. Lasserre et al.

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Answers to referee #1

4) a * We do take into account the gravitational deposit (Fig.3) in the dust transport process according to the model of Alfaro and Gomes [2001]. The dry deposit rate of each size particle is calculated [Cautenet et al., 2000]. Dust particles with radii larger than 13 μm are not much involved in long range transport because of quick gravitational settling. The sedimentation of a 13 μm radius dust particle is 6 cm.s-1 (the sedimentation of a 20 μm radius dust particle is faster than 10 cm.s-1). We observe that these large mineral particles can reach the anthropogenic pollutant sources when they are close to the desert areas (Hohhot, for instance). However, the largest dust particles

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have a weak optical depth impact as compared to the one of the submicronic mode, due to their (relative) small number and, therefore, to their relatively weak specific extinction surface (compared with the global surface, including all bin sizes). Moreover, large mineral particles won't reach the main SO2 and BC sources of the east part of the studied area.

* Even if our case study is not actually related to a cloudy period, the wet deposit (rain-out and wash-out) of dust particle is explicitly taken into account according to the method of Cautenet and Lefeivre [1994]. This sentence has been added in the final version of the paper.

* The Referee is right but we wonder if the figure would be readable with more details. Figure 3 only describes the anthropogenic/mineral mixing above megalopolis, i.e. far from desert dust sources. The brown dust cloud (which contains dust coated by SO2, etc) on the upper left part of Figure 3 symbolizes the dust arrival - and not the dust source itself - above the cities or any other anthropogenic pollution source: this point will be more clearly specified in the figure caption .

4) b * We voluntarily omitted the in-cloud SO2 oxidation into SO4 because there were few water clouds above the continental area during the case study. Our simplified tool does not consider SO3 from the oxidized SO2 emitted from combustion sources. We know that this kind of emission exists, but we did not find SO3 specific emission data in the EDGAR database or, for the European area only, with the EMEP database (SO2 and SO3 are reported as SO2).

* Vertically, anthropogenic pollutants are homogeneously injected into the 3 first levels of RAMS, i.e. approximately between ground level and 120 m agl. Sensitivity tests (not shown here) strongly suggest that the model results are, to a large extent, insensitive to the injection height. Our mesoscale simulation tries to be spatially relevant but cannot be more accurate than a 25 km resolution in the horizontal scale. Moreover, the GEIA and EDGAR databases provide emission fluxes with a 1° x 1° horizontal resolution. For

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these reasons, it is unfortunately not possible to model the actual 3D grid of emitting points, even for large power plants.

* We unfortunately introduced a misunderstanding with the beginning of the sentence on line 3, p. 11904. In the next version of the paper, we will delete "This aspect of [...]", because this sentence recalls - for the bibliographic overview - that Kulshresta et al. [2003] explicitly detail the mixing of dust and SO2 for predominantly dry conditions. In our simplified model, the oxidation on dust particle surface is not modelled. In the paper, we just suggest what could happen as regards long range transport, by following, for instance, the estimates of Dentener et al. [1996] who model large sulphate formation with mineral aerosol in the vicinity of the dust source regions. If the referee wishes, we can recall that Usher et al. [2002] explain how SO2 captured on dust becomes adsorbed SO32- and/or HSO3- species, before being oxidized into SO42- and/or HSO4- by ozone, but - once again - we don't include such process in our simplified tool. So, in the paper, we did not write that the adsorbed SO2 conversion into SO4 might be instantaneous but we just wanted to recall that the free conversion of H2SO4 into SO4 (line 26, p. 11903) is fast when the relative humidity becomes important (see e.g. Umann et al., 2005, who state that "for RH $<$ 50% Dentener et al. [1996] suggest an uptake coefficient gammaSO2 of only 3 10E-4 (but, in contrast, RH $>$ 50%: gammaSO2 = 0.1)". Now, this value is often exceeded in the coastal region. As suggested by your questions, this paragraph has been rewritten to better explain, on the one hand, what is modelled for free SO2, and on the other hand, what is only suggested for captured SO2.

4) c * Chemically speaking, BC is considered in the model as an inert species (Fig.3). We include BC in the model as only EC, and do not include primary Organic Carbon. According to the GEIA database, global BC annual amount is about 12 Mt.yr-1 and BC is of course not the predominant anthropogenic PM10 in mass over East Asia. Sulphates are roughly 1 to 10 orders of magnitude more concentrated in this area than BC. We modelled a 5 day period with a dusty/less dusty/dusty sequence. As the dust,

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if present, is much more concentrated than anthropogenic pollutants, our comparisons with PM10 observations (Fig. 10a - with the (a) - corrected on line 23, p.11918, and Fig. 10b) concern the actual overall observed PM10 species versus the only modelled dust concentrations. It is thus logical to obtain model concentrations close to the observed ones during the dust storm episodes and to obtain model concentrations slightly inferior to the PM10 observations when dust concentration is lower, but still present. To know the contributions of BC and sulphates which could be added to the dust concentrations to complete the PM10 estimation, please refer to Figure 13 of the submitted paper (not shown anymore on final version). With these concentration values added to the dust values, Figure 10 would not be significantly modified and these modifications would be completely irrelevant according to the accuracy of the local observations compared to the horizontal accuracy of the model

* We are very sorry to let believe that we consider BC as a tracer for all anthropogenic emissions because we thought that we deeply insist on the major role of sulphates. We have replaced the BW figures 14 (a) to (e) by their coloured versions We hope that the respective contributions of BC, dust, free SO4 and sulphates on dust will be more emphasized now.

* The use of level 1.0 AOT data is relevant because it appears - by clear sky conditions, which is the situation of our case study, except over Liangning city - to be the only way to get real-time fast variations of AOT. We took care about the condition of no hydrometeors and we used the 1.0 AOT because it is not possible to observe the short-time maxima of AOT with 1.5 and 2.0 AOT. For example, the AERONET data for April, 28, 2005, concerning the first dust storm over Beijing: it appears as a sharp event with 1.0 data (http://aeronet.gsfc.nasa.gov/cgi-bin/type_one_station_opera_v2_new?site=Beijing&nachal=0&year=13&month=3&day=27&aero_water=0&level=1&if_day
but it is an "invisible" event with 1.5 data because the post-processing makes the data not accessible at this time. Our work tries to retrieve real-time observations, so the level 2.0 - usually used for long period studies, using mean values over

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days or weeks - are less interesting than the raw 1.0 data. In these figures, the strong peak (almost 3 in AOT) at 0500 (in level 1.0) is totally removed in level 1.5. The value of the Angström exponent (which lies between -0.2 and 0.1) strongly suggests the signature of dust (cf. observations - see below - and models). Meteorological observations in Beijing related to this event (real-time sky observations): http://english.wunderground.com/history/airport/ZBAA/2005/4/28/DailyHistory.html?req_city=NA&req_state=NA&req_state

6) We hope that the answer to questions 4 will complete and answer this remark 8) We propose a new title, more relevant to this study: "A comprehensive model tool for assessing real-time mixings of mineral and anthropogenic pollutants in East-Asia: case study of April 2005." 12) corrected on revised version to be submitted.

13) * We hope that with the answers of the questions above, it becomes clear now that the BC contribution in the optical impact cannot be ignored and thus cannot be removed from the paper because it is an inescapable issue as regards the Asian pollution case. To focus more directly on the modelled AOT, we will shorten the paper in section 5.3 whose main results are the derived AOT on section 5.4, but we keep Table 2 summary. * As asked by the referee, we propose new figures 14 (a) to (e) (now numbered figures 13) with larger sizes and coloured version to show - beyond the other aerosols - how BC impacts the total modelled AOT and very significantly contributes to the retrieval of the real-time magnitudes of the observed AOT. It then validates - with all the meteorological hypothesis of the case study - how our simplified tool based on average chemical and physical features gives realistic evolutions of the AOT and how each of the two optically most important anthropogenic pollutants contribute to the pollution issue when dust is more or less present.

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