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Atmospheric
Chemistry
and Physics
Discussions

Interactive comment on “Contribution of carbonaceous aerosol to cloud condensation nuclei: processes and uncertainties evaluated with a global aerosol microphysics model” *by* J. R. Pierce et al.

J. R. Pierce et al.

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Response to Anonymous Referee 3

Referee comments in *italics*

Referee 3, thank you for the detailed read that you gave our paper and the comments that you made on it. We appreciate your attention for detail your help in us making a better paper.

1 General Remarks In their manuscript J. R. Pierce et al. present an analysis of the contribution of carbonaceous aerosol to cloud condensation nuclei based on global modeling studies with the TOMAS

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sectional aerosol scheme coupled to the GISS climate model. For this purpose the authors extend their sectional aerosol model of sulfate and sea salt by primary carbonaceous aerosols (black carbon and organic matter).

The analysis of anthropogenic contributions to CCN is crucial for the understanding of the indirect aerosol effects and therefore a highly relevant research topic well in the scope of ACP. The manuscript is well written and the analysis is innovative - in particular the evaluation of the importance of size vs. composition for CCN predictions is an important quantitative extension of previous work.

However, and in this case unfortunately, to my understanding the manuscript has some major issues that will need a substantial revision of the manuscript before publication. The introduced changes demand for a careful evaluation of the model, in particular as the limited presented evaluation reveals some significant problems: if the simulated CN values are biased high by a factor of 4 the reader needs to be convinced that the (unevaluated) CCN values are reliable. Thus, I would recommend to either significantly extend the manuscript by a basic evaluation, as outlined below, or to split the manuscript into a introductory paper with detailed evaluation and an scientific application of the evaluated model. 2 Major Issues Evaluation The authors extended a microphysical model of sulfate and sea salt by carbonaceous aerosols, a substantial modification from their previous work that increased globally averaged surface CCN by 65-90%. This is essentially a new model - with completely new results that demand for a careful evaluation. To my understanding the authors somewhat leapfroged this step (that typically is a publication on its own) and jump right away into the scientific analysis. This is worrisome as the limited three evaluations performed (surface mass concentrations of BC and OC; surface aerosol number concentration (CN$>$10nm); large scale average marine size distributions) show non-negligible deviations that ask for a more detailed analysis. The model over-predicts CN on average by a factor of 4, so I am missing a supportive analysis why the (unevaluated) CCN predictions should be a reliable basis for their subsequent scientific analysis.

A comparison of simulated aerosol size distributions to observed aerosol size distributions in Europe (Putaud et. al (2003)) has been added to the text (see below). Comparison of accumulation mode number concentrations, in particular, helps explore CCN prediction in this region. Also, the CN bias is something of a worst-case scenario; because the CN-CCN relationship is sublinear, the resulting CCN bias is much lower.

We do feel, however, that interesting science and results may come from imperfect models. Model evaluation is an important pre-requisite, but it is also a never-ending process. We included the interesting science results in which the major conclusions do not depend on the CCN prediction being perfectly accurate (e.g. the sensitivity of the contribution of primary organics to CCN from organic solubility) with the model development paper to make for a more interesting paper. We have added the paragraph "In this section we explore CCN predictions by the model and test the sensitivity of CCN to organic solubility and mixing assumptions. Although the model overpredicted CN globally, the model showed much less bias in the accumulation mode number in Figs. 5 through 8. Furthermore, CCN concentrations tend to vary sub-linearly with CN concentrations allowing CCN errors to be, in general, smaller than CN errors." to the beginning of Section 3.4.

The model predicts also very high CCN surface concentrations, e.g. CCN(0.2%) are in the annual mean $>$ *1000 cm*$^{-3}$ *over large parts of the continents and even* $>$ *2000 cm*$^{-3}$ *in polluted areas. These values seem high given that e.g. Hudson and Yum (2002) report CCN concentrations for polluted air masses as CCN(1%): 1190 cm*$^{-3}$*, CCN(0.1%): 580 cm*$^{-3}$ *and demand for a more detailed evaluation. It is true that CCN measurements are not generally available on an operational basis. Therefore, an evaluation of the aerosol optical depth, that is in these areas typically dominated by the CCN relevant accumulation mode size range, could be an essential first evaluation step.*

Optical depth and angstrom coefficients in general are not good indicators of CCN (Kapustin et al., 2006). We are, however, doing a comparison of our model to optical depth measurements. However, this is being done with a (later) version of the model that includes mineral dust aerosol plus all of the species included here.

However, also the limitation of the evaluation of the size-distributions to marine areas that, according to Fig. 3 and Fig. 6, are almost unaffected by anthropogenic aerosols is insufficient. Suitable datasets are available (e.g. Putaud et al., 2003) and have been used as benchmark in previous modelling studies.

We have added a size distribution comparison with the size distributions published in the Putaud et al. 2003 report and Van Dingenen et al. (2004) (new Figures 5 and 6). These figures show that the overprediction at four European locations in CN comes from particles $<$ 100 nm for all simulations and that the accumulation mode aerosol number is predicted much more accurately. We have added the text, "In order to determine if the model is representing the CCN concentrations more accurately than CN concentrations, we have done comparisons of the aerosol size distribution. Figures

5 and 6 show a comparison of simulated to observed aerosol number size distributions at four of the locations (Jungfraujoch, Aspvreten, Harwell and Hohenpeissenberg) from Van Dingenen et al. (2004) and Putaud et. al (2003) for June, July and August, and December, January and February, respectively. Both simulations and observations show the number size distribution as a function of dry diameter with the exception of the observations at Harwell, which are given as ambient diameter. The data in Van Dingenen et al. (2004) is given as average distributions for the morning, afternoon and night. We have plotted the mean values of these three distributions. The total number at all four locations were shown to be overpredicted in all model simulations in Figure 4. Figures 5 and 6 are consistent with this with the NOCARB, BBASE and IBASE simulations overpredicting the aerosol number in the ultrafine (Dp $<$ 100 nm) size range. The three simulations predict the size distributions more accurately for sizes larger than 100 nm. CCN(0.2%) are, in general, particles of about 80-100 nm and larger, giving us confidence that our model is predicting CCN at these European locations more accurately than the model is predicting CN. Also shown in Figs. 5 and 6 is that the dominating number mode at these locations for all simulations is centered around 20 nm. This corresponds approximately to the primary sulfate emission size, and because we do not get significant boundary layer nucleation, this is likely a major source of the CN overestimation."

Finally, I would have expected some effect of the introduction of carbonaceous aerosols on the distribution of the other species, as modifications in the size distribution and composition directly affect the sinks. It would be a surprising and interesting results if the other species are unchanged. However, if they changed, the changes would need to be discussed in the manuscript and the species maybe re-evaluated.

We have added a table (new table 3) with the budgets of the other species before and after the carbonaceous aerosol was added. The sulfate burden and lifetime decrease on the order of 1% and the sea-salt shows no change.

2.1 Calculation of CCN

The basis of the calculation of CCN in the manuscript is somewhat unclear. In section 3.4 (page 7740, line 25) the authors write: "The CCN(0.2%) concentrations are found using modified Köhler theory as *discussed earlier with the annually averaged size distributions and chemical compositions." I could not find any information if this applies for all calculated CCN concentrations in the manuscript or not - thus I have to assume it does apply. This would to my understanding significantly affect the (interesting) analysis of the effect of aerosol size vs. composition in Section 3.5. If all CCN are derived from annual mean size and composition data, the model effectively uses a (locally refined) bulk scheme for the calculation of CCN. In this case the comparison in Section 3.5 and Fig. 7 would not be a comparison of a bulk scheme vs. an average of CCN from an exact instantaneous activation calculation but rather a comparison with a local bulk scheme for each grid box. Given the non-linearity of the activation process, I would be surprised if this yields the same result.*

If activation diameter did not depend on composition, using annually averaged size distributions to predict the annually averaged CCN would yield the same result as if the instantaneous CCN was used to predict the annually averaged CCN concentration. However; you are correct that this does not hold true when averaging over changing chemical compositions. We have rerun three months of the IBASE simulation printing CCN concentrations every 6 hours and found that the globally average error was about 2%. This indicates that, for typical variations in aerosol composition, the change in the activation diameter is relatively small. We have added the text after the sentence quoted above "Using average size distributions and compositions to calculate average CCN concentrations rather than using the average of the instantaneous CCN concentrations gave results with error on the order of 2% globally when tested across a three month period."

3 Specific Issues

Title, Abstract, Conclusions

I think it is important to point out that SOA's are not included (also not as proxy), thus I would recommend to consistently use "primary carbonaceous" instead of "carbonaceous".

This is correct, we have added "primary" throughout the paper including the title.

page 7726

"cloud brightness forcing" is an unusual term. I assume you mean cloud albedo effect? Otherwise please explain.

Yes, this is what we mean. We took the two names of the 1st indirect effect as synonymous. We have changed brightness to albedo.

page 7727

"The modal representation has an inherent disadvantage, however, in treating processes such as activation and cloud chemistry that create discontinuities in the size distribution."

This is certainly true on a local basis. In the grid box average over more than 100x100 km, that global models represent, these discontinuities are rarely observed. This is for example evident in the Hoppel gap of the size-distributions in Fig. 5 that is very sharp in the simulation but log-normal in the averaged observations.

We changed the sentence to, "The modal representation has an inherent disadvantage, however, in treating processes such as activation and cloud chemistry that create discontinuities in the size distribution, at least on a local basis."

As discussed later, the observations in Figure 5 were fit to two lognormal distributions; it is not necessarily true that the raw observations themselves are lognormal.

page 7728

"Carbonaceous particles may affect the CCN concentrations through two different pathways."

This is a simplified view on this issue as other effects, such as surfactants, are neglected.

It is true that we have neglected surfactant effects. We have changed the opening sentence of the paragraph to read "We have simplified the effects of primary carbonaceous particles on CCN concentrations by grouping them into two different pathways." We have added the following sentence to the end of the paragraph. "The two pathways explored here do not include the effects of organics on particle surface tension and the increased organic mass that SOA may partition into, both of which affect CCN."

Section 2.3

The description of the calculation of CCN is very limited and should be extended.

This is correct. There was a brief discussion of CCN in Section 2.1 with a promise that more will be discussed in Section 2.3, however, we overlooked this and forgot to add it. We have added the following paragraph to Section 2.3, "For our mixed aerosol population, we use modified Köhler theory to calculate the number of CCN in the model along with the number of active particles in clouds for wet deposition (Hanel, 1976; Laaksonen et al., 1998; Raymond and Pandis, 2003; Seinfeld and Pandis, 1998). This allows for the calculation of the activation diameter of particles containing various soluble and insoluble components. The hydrophilic OM contributes the appropriate number of solute molecules per OM mass to give an activation diameter of 140 nm at 0.2% supersaturation for a pure hydrophilic OM particle. Sulfate is assumed to be ammonium bisulfate that completely dissociates to give a van't Hoff factor of 3 and sea-salt is assumed to be sodium chloride with a van't Hoff factor of 2. Hydrophobic OM and all EC are assumed to be an insoluble core. In this treatment, we ignore changes in surface tension due to the contribution of surfactants by the organic aerosol."

Section 2.1, page 7731

"we neglect interstitial scavenging in clouds" Could this contribute to the overestimation of CN as it leaves turbulent dry deposition as the only sink for small particles?

Small particles are also lost through below cloud scavenging. Below cloud scavenging is discussed in the paper two sentences after the sentence quoted above. To clarify this we have changed the sentence "Below-cloud scavenging removes particles colliding with falling raindrops" to "Below-cloud scavenging removes particles of all sizes colliding with falling raindrops".

Ignoring interstitial scavenging may cause small particles to be underpredicted. This was explored in Adams and Seinfeld (2002) with a sulfate-only version of the model CN concentrations were 10-20% lower in the boundary layer and a few percent lower in the free troposphere when all particles were assumed to be activated in the cloud (an upper limit on interstitial scavenging). Where-as this will change somewhat with other species added to the model the sensitivity done in Adams and Seinfeld (2002) was an upper limit case for interstitial scavenging, so actual effect of interstitial scavenging should be quite low.

page 7731

“During microphysics, all aerosols are treated as internally mixed.” This is a serious limitation of this study as it will affect the growth of particles to the relevant CCN size. I think this requires some more detailed discussion of the potential implications for the results.

There are two ways that particles may grow: condensation and coagulation. In the model, we assume that the sulfate condensation accommodation coefficient does not depend on particle composition, nor are we aware of any evidence to suggest that it depends on particle composition. We do not have SOA in the model, so organic partitioning is not an issue. Except for the 2nd order effect of water uptake, condensational growth of a particle should not depend on whether the particle is internally or externally mixed. A similar argument follows for coagulation (coagulation coefficients depend on aerosol size not composition) where water uptake is the only means for particle growth to change. Because of this we don’t expect growth to a given CCN size to vary greatly.

The externally mixed population will have to grow to a larger size to become a CCN; however, this is the case even if we did not treat the aerosols as internally mixed during microphysics.

Sulfate condensing onto the carbonaceous externally mixed population causing it to grow would cause it to move towards internally mixed (lowering its activation diameter). The same may be said for the carbonaceous particles coagulating with the inorganic particles. In our externally mixed simulation, we continue to treat these populations as externally mixed even after these processes occur, which, although idealized, is a bounding case for the affect of external mixing on CCN. As shown in Figure 6, and discussed in Section 3.4.2, this extreme external mixing assumption does not greatly change the number of CCN predicted.

We have changed the last portion of section 2.4 to “This assumption does not appreciably alter microphysical growth rates because condensation and coagulation rates depend primarily on aerosol size, not composition. A second-order effect is the effect of aerosol mixing state on water uptake and, therefore, on condensation and coagulation growth rates. This effect is neglected in the current study; nevertheless, these sensitivity simulations provide insight about the importance of mixing state on cloud processes."

Section 2.2, page 7731

Please give the base years of the emission datasets. Given the availability of significantly improved satellite based wildfire emission datasets, the provide the observed seasonal distribution, the used inventories seem somewhat outdated.

We have added the following sentence for IPCC, "The base year for these emissions is 2000 (IPCC, 2001).", and the following for Bond, "The base year of the Bond et al. (2004) emissions is 1996 for fossil fuel and biomass burning and the open burning is based on fire counts during 1999-2000."

Emissions, page 7732

I am somewhat surprised by the choice of the emission size-distributions from near source measurements. Given that the initial evolution of aerosols occurs on much smaller scales than the 5 degree grid boxes of the model, the assumption of somewhat aged size distributions would appear more appropriate. This could be the reason for the strong overestimation of CN in the model. Also, in particular downwind biomass burning size distributions have typically much larger sizes, a factor that has been taken into account in the recommendations for e.g. the AeroCom emission size distributions Dentener et al. (2006).

This is an issue that we are working on exploring with the model, as well as trying to understand the cause for the large overestimation of CN. The next version of the model is being run with AeroCom emission size distributions as one of its test scenarios.

We have added the sentence, "Also, uncertainty also arises due to use of near-source size distributions as opposed to the size distribution of particles well mixed within the grid-cell" to that paragraph.

Mixing state, page 7733

Are hydrophobic and hydrophilic OM really always assumed internally mixed as has to be assumed from *this description, dividing the carbonaceous aerosols into a pure EC population and an internal mixture of all other components? How can the internal mixture remain hydrophobic?*

Both OM types are assumed to be internally mixed during all simulations except for the BEXT and IEXT simulations. The hydrophobic OM is classified as such because it is assumed to not dissolve in water (i.e. "hydrophobic" in this case refers to the solubility of the OM itself, not to the hygroscopicity of the overall particle). The particle that it is internally mixed with may take up water; however, there will be no additional water uptake due to the dissolution of hydrophobic OM. It is indeed artificial to have a population of pure EC particles. However, we choose to err on the side of treating more aerosol components as internally mixed since single particle mass spec and TDMA data indicate that CCN-sized particles are internally mixed much more often than not.

This discussion should be clearer now with the addition of the discussion about Kohler theory in Section 2.3.

Mixing state, page 7733

The choice of the BC aging timescale seems to be based on a relatively old source. Recent model based estimates, e.g. Riemer et al., yield shorter timescales. This could also contribute to the simulated relatively long BC residence time.

The shorter timescale determined the in the Riemer paper was found using a mesoscale model over a relatively smaller region of Germany. This may not be representative of the global mean. The aging timescale of 1.5 days is still significantly shorter than the average lifetime of aerosol, so even reducing this to time to zero should lower the lifetime by 20-30% and increase the burden by the same factor (Cooke and Wilson, 1996).

We have added a sentence to the end of that paragraph, "This timescale is shorter than the mean lifetime of particles in the atmosphere, so uncertainties in the aging timescale should have only a modest affect on the carbonaceous burden."

Section 2.4, page 7734 I was missing a description of the simulation setup here. How long has the model *been integrated, resolutions, boundary conditions, such as SST need to be described.*

We have added "All simulations are spun up for six months followed by one year of simulation time." to the first paragraph in Section 2.4. The model resolutions are given in Section 2.1. In Section 2.1 we have added, "Sea-surface temperatures are specified as the mean values from 1979-1993."

Section 3.1, page 7736

It would be important to show the budgets of all components here.

Added, see discussion above.

Section 3.2, page 7736

Please be more specific about the sampling of the data (also in the captions). Is the evaluation also done on an annual mean basis? Why not as monthly means that are available at least from IMPROVE?

We added the following sentence to Section 3.2, "Sampling for IMPROVE includes twenty four hour aerosol samples that were taken twice a week (on Wednesdays and Saturdays). The observation data are averaged over 3 years from March 1996 to February 1999. The sampling of the rural, remote and marine sites are averaged over various time periods and details are given in Tables 10-15 in Chung and Seinfeld (2002)."

A detailed seasonal evaluation of the emissions datasets in the model is beyond the scope of this paper.

Section 3.3, page 7738

Please be more specific about the evaluation. How was the sampling performed? Do the measurement instruments have the same lower cut-off? I am somewhat puzzled by the strong overestimation of CN and surprised that they authors have not repeated the simulations with updated emission size distributions after the significant overestimation of CN by a factor of 4.

We have added the text, "The CN observations were done using a condensation nucleus counter (CNC) in the case of the GMD and AIRMAP data and using a CNC with various size scanning devices in the case of the European sites. The low limit cutoff for the CNCs in the GMD and AIRMAP data is 10 nm (which corresponds to the lower size limit of the model). The lower size limit for the CNCs used in the Van Dingenen et al. (2004) paper vary, however, they have corrected their number counts for a lower cutoff of 10 nm using the size distribution measurements."

Work is currently being done on the emissions size distributions and will be published in a future paper. Added, "The bias in CN by the model is large and we are currently addressing this in our future work."

Figure 5, page 7740

The simulated marine size-distributions seem to have a distinct discontinuity, probably at the critical radius of activation, and do not look log-normally distributed - while the observations show smooth log-normal distributions. I am surprised that this is the case in the annual mean values. Is this due to the assumption of fixed supersaturation in clouds?

This is correct, the discontinuity is due to the fixed supersaturation of clouds. The activation diameter varies due to the composition of aerosol, but not enough to smooth the discontinuity in the average values.

The "observations" show a smooth log-normal distribution because the raw observational data is fit to two lognormal modes by Heintzenberg (2000); the original observations themselves are not necessarily lognormal.

Section 3.4, page 7740

As discussed before, I see issues with the calculation of CCN from annually averaged aerosol data.

See our discussion above indicating that the error is 2%. The primary dependence of CCN concentrations on the number size distribution is linear (for a fixed supersaturation). The nonlinearity resulting from (relatively small) shifts in the activation diameter based on time-varying aerosol composition induces the small difference.

Section 3.4, page 7740

I think the authors need to make significant effort to evaluate the predictive skill of the model in terms of

CCN - as CN have a significant bias of a factor of 4.

Please see our discussion in the "Major issues" section. Size distributions and accumulation mode number concentrations are now evaluated in marine and polluted continental areas.

Section 3.4.1, page 7742

It might be interesting to show ratios here, as would be easier to identify the regions of importance.

There are definite advantages to showing ratios, however, they also would make regions that start with very low CCN concentrations that get only modest increases in CCN look artificially important. We have corrected panel A, which was inadvertently the same as panel B. This may have been a cause of not being able to identify important changes in the results between simulations

Section 3.5, page 7744

As discussed before, I see issues with the calculation of CCN from annually averaged aerosol data.

See our discussion above.

Conclusions, page 7746

They authors attribute the overestimation of CN on the primary sulfate emission size distribution. On what basis? How do the primary particle number compare to the carbonaceous flux?

The basis for this was Figure 4, where CN is already overpredicted in the polluted regions in the simulation without primary carbonaceous aerosol. The statement of attributing the too many CN from primary sulfate particles follows the sentence "A comparison to a network of total aerosol number measurements shows that the model predicted number concentrations were on average about a factor of 4 too high, although even without carbonaceous particles included, the number concentrations are a factor of 3 too high."

We have changed the sentence attributing the error to, "This overprediction of CN may be due to the emission of too many particles through primary sulfate emissions and

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aided by incorrect emission size distributions of carbonaceous particles. The sensitivity of CN and CCN to these emissions is being performed in future work."

Conclusions, page 7746

I was missing a reference to the caveat of the simplifying assumption of internal mixing in the microphysics and the potential implications here, and in the abstract.

Please see the discussion given for the earlier comment on external mixing.

Conclusions, page 7746

I was missing a discussion of the sensitivity of their results to the emission size distributions.

This paragraph now reads, "Predicted primary carbonaceous aerosol mass and aerosol number concentrations were compared to observations. Errors in predictions of OC and EC masses were a factor of 3 on average and OC predictions were biased towards too little mass whereas EC predictions showed little bias. A comparison to a network of total aerosol number measurements shows that the model predicted number concentrations were on average about a factor of 4 too high. Even without carbonaceous particles included, the number concentrations are already a factor of 3 too high. A comparison of the simulated aerosol size distributions to observations at several European sites showed that the overprediction of CN at the sites was due to large overpredictions in the number of particles with diameters smaller than 100 nm, whereas the accumulation mode particles were predicted much more accurately. This overprediction of CN may be due to the emission of too many particles through primary sulfate emissions and aided by incorrect emission size distributions of carbonaceous particles. The sensitivity of CN and CCN to these emissions is being performed in future work. In contrast, a comparison of CN to marine observations showed very little overprediction (<30%).

Figure Captions

Most of the figure captions are not self explaining and do not give basic information, such as sampling periods, etc.

Done

References

Bond, T. C., Streets, D. G., Yarber, K. F., Nelson, S. M., Woo, J. H. and Klimont, Z.: A technology-based global inventory of black and organic carbon emissions from combustion, Journal of Geophysical Research-Atmospheres, 109, ISI:000222916600001, 2004.

Cooke, W. F. and Wilson, J. J. N.: A global black carbon aerosol model, Journal of Geophysical Research-Atmospheres, 101, 19395-19409, ISI:A1996VE25800041, 1996.

Hanel, G.: Single-Scattering Albedo of Atmospheric Aerosol-Particles as a Function of Relative Humidity, Journal of the Atmospheric Sciences, 33, 1120-1124, ISI:A1976BV24200020, 1976.

Kapustin, V. N., Clarke, A. D., Shinozuka, Y., Howell, S., Brekhovskikh, V., Nakajima, T. and Higurashi, A.: On the determination of a cloud condensation nuclei from satellite: Challenges and possibilities, Journal of Geophysical Research-Atmospheres, 111, ISI:000235966500001, 2006.

Laaksonen, A., Korhonen, P., Kulmala, M. and Charlson, R. J.: Modification of the Kuhler equation to include soluble trace gases and slightly soluble substances, Journal of the Atmospheric Sciences, 55, 853-862, ISI:000072288400011, 1998.

Putaud, J. P.: A European aerosol phenomenology; physical and chemical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe, 2003, European Commission, EUR 20411 EN., http://ies.jrc.cec.eu.int/Download/cc

Raymond, T. M. and Pandis, S. N.: Formation of cloud droplets by multicomponent organic particles, Journal of Geophysical Research, 108, 4469 4476, 2003. Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics, John Wiley and Sons., New York, 1998.

Van Dingenen, R., Raes, F., Putaud, J. P., Baltensperger, U., Charron, A., Facchini, M. C., Decesari, S., Fuzzi, S., Gehrig, R., Hansson, H. C., Harrison, R. M., Huglin, C., Jones, A. M., Laj, P., Lorbeer, G., Maenhaut, W., Palmgren, F., Querol, X., Rodriguez, S., Schneider, J., ten Brink, H., Tunved, P., Torseth, K., Wehner, B., Weingartner, E., Wiedensohler, A. and Wahlin, P.: A European aerosol phenomenology-1: physical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe, Atmospheric Environment, 38, 2561-2577, ISI:000221265500011, 2004.