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Atmospheric
Chemistry
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Discussions

Interactive comment on "Modelling of base cation emissions, concentrations and deposition in the UK" *by* M. Werner et al.

Anonymous Referee #1

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Referee Comments to the manuscript acp-2010-480.

Modelling of base cation emissions, concentrations and deposition in the UK by M. Werner, M. Kryza1, A. J. Dore, M. Błaś, S. Hallsworth, M, Vieno, S. Tang, R. Smith

The manuscript presents calculations of base cations (BC) for the UK performed with the FRAME model. Accounting for BC pollution is important both in modelling of atmospheric chemistry (e.g. formation of coarse NO3 particles) and in assessment of ecosystem damage and recovery due to the deposition of acidic species, as BC deposition increases the soil alkalinity and thus lessens the effects of acidification. In the calculations, the BC sources included anthropogenic and natural emissions from land and sea spray, with a large focus on the latter. The calculations of air concentrations and wet deposition of Na+, Mg2+ and Ca2+ have been performed for 2003 and 2006

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and compared with observation data from the National Monitoring Network and measurement based estimates of UK deposition budget. The model results were found to be in a reasonable agreement with measured wet deposition of BCs, with somewhat larger scatter for Ca2+. The model calculations somewhat overestimate observed BC air concentrations on the west coast, while underestimate those in the central parts of the UK.

The manuscript addresses a highly relevant issue and present interesting results on BC concentration and deposition levels and distribution in the UK. Those estimates can make a good contribution to the assessment works of BC impact on ecosystem acidification and their influence of PM10 concentrations and composition. The paper is fairly well structured and written, and quite easy to follow. It can be recommended to publication in ACP after the authors considered/addressed a number of comments below.

One of the general comments is that that despite of the title and the aim stated in the Abstract, i.e. modelling of base cations, several parts of the manuscript give an impression that sea salt modelling is largerly in focus. For example, the whole Introduction is dedicated to description of sea spray production (parameterisations); section 3.1 states that “The main purpose of the work is to calculate BC emissions from sea salt...”, and it is clearly seen in the results and illustrative figures. Also, in Summary a considerable attention is paid to sea salt production. The authors should decide firmly what the manuscript is about and be more consistent and balanced in presenting the work.

Connected to the first comment, rather limited details are given regarding BC emissions from anthropogenic sources and soil, i.e. their temporal and spatial distribution, relative contribution to the total emission amount (the emission numbers for anthropogenic BC are difficult to compare with sea spray contribution, and no numbers at all are mentioned for natural BC emissions). Also, it’d useful to indicate the uncertainties if available.

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As to data for evaluation of model calculated BC, it is advisable to provide (in a form of table or a map) details about measurement sites, especially because the differences in model performance for coastal and inland sites are discussed. Also some more relevant details can be given about the CBED measurement-interpolation technique. These would contribute to get a better insight in possible uncertainties in CBED estimates and help in interpreting comparison between those data with model calculations.

Furthermore, model results for BC concentrations and depositions would benefit from more in-depth discussion, including e.g. the relative importance of different emission sources (including anthropogenic and soil), main uncertainties in calculated BC due to input data (emissions, meteorology) and model formulations, which could give better understanding og discrepancies between calculations and measurements.

More specific: Through the whole manuscript, Maårtensson should be corrected to Mårtensson

P. 21990

Line 11: Mårtensson et. al. (2003) for ultra fine particles, Monahan et al. (1986) for fine particles and Smith and Harisson (1998) for coarse particles;

Line 22: whereas for Ca2+ the correlation is poor

Line 23: ..concentrations are represented

P. 21991

Line 1: Introduction talks basically exclusively about sea spray/sea salt

Line 9: salt salt particles with radii smaller than 0.1 um also form

Line 11: The years 1986, 1987, 1992, etc. are not so recent

P. 21993

Line 1: What do you mean by “also”?

Line 8: Probably you could already here specify the “selected parameterisations”.

Line 9: which physical properties are meant by “according to physical properties SSA”

P. 21995

Line 14-15: Are the “annual rainfall fields” used in model calculations? Or some temporal variation is imposed on them?

P. 21996 2.3 Emissions: It would be quite useful to show maps of anthropogenic and soil BC emissions

Line 3: base cations (plural)

Line 9: should probably stay “and coal burning”

Lines 18-23: it’s worth describing more the natural Ca emission estimates, e.g. spatial and temporal resolution, and also their inter-annual variability (since average emissions were used for the calculations).

P. 21998

Line 5: why “particle- size distribution”

Line 6-7: use for example “denoted as r80 or r0 ”

Line 11: correct O’Dowed to O’Dowd

Line 13-14: I think it is quite right to state that “The emissions of drops from breaking wave whitecaps and sea foam is quite well known”. The uncertainties are considerable!

Lines 15-18: many repetitions; e.g. no need to say non-lineary if it was described as power-low just before.

Line 19: somewhat slack formulation “sea salt aerosol production (PM10)”

Line 21: Which radius is referred to here, r80 or r0 ?

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P. 22000

Line 5: what is the size range of calculated SSA? In particular, what is the upper size limit of sea salt particles used for describing PM10?

Also, nothing is said about how SSA emissions were distributed between model vertical layers.

Line 19: suggested to change to “Data for model evaluation”

Line 20: for both years 2003 and 2006

Line 28: what is meant by “ two stations were eliminated because of monthly data”?

P. 22001

Line 4-8: Please elaborate a bit more regarding “large coefficient of variation fro Ca: what are plausible the reasons (e.g. large temporal variations due to windblown dust? Or measurement artefacts?) and implication of this.

Line 9: “not large change in number of stations”? From 10-12 in 2003 to 30 in 2006.

Line 17: check on “The main purpose of the work..”!

Line 19-20: what exactly was tested in those tests? And how the results of the tests were further used in the work, e.g. in interpretation of model calculations. I am not sure it is very necessary to include these tests in the section about results (they only shift the focus even more to the issue of sea spray source function parameterizations).

Lines 15-26: Unclear what results are discussed here? If those in Fig.2, reference should be given here. I think it is a bit confusing that authors talk here about emissions from three individual source functions. Have not they be combined in a single scheme in FRAME? As to the numbers given for emissions of ultra-fine, fine and coarse SSA, do they correspond to the same grid cell/area, or they describe just random grids?

Line 16: who is “it” in “it gives”?

Line 24: what is meant by “the UK sea domain”? Any formal definition, e.g. 100 or 200 nautical miles?

P. 22003

Line 4: Is the larger SSA production due to larger wind speeds in 2006 compared to 2003 ?

Line 14 : Is that a map from the run with total (land + sea) emissions? It would be interesting to include a map showing the relative contribution of e.g. sea salt source; and for Ca+2 it would be useful to also see separate contributions from anthropogenic and soil dust sources.

P. 22004

Line 5, 7: “BC” shows up for the first time without explanation.

Line 26-27: suggested “calculated concentrations are mostly within a factor of two of observed values”.

P. 22005

Lines 1-15: it would be worth discussing why the results for (only) Ca compares with measurements rather differently for 2003 and 2006; due to uncertainties in natural dust emissions? What was particular about 2006 in this respect, e.g. dry and windy? Or the anthropogenic emissions?

Why Ca in precipitation is so underestimated, while Ca in air so much overestimated in 2006? The suggestion that the land emissions are underestimated does not explain the latter.

Line 3-4: how can we see that if none description of site locations is provided?

Line 16: the authors write that it was “possible to compare model results with estimations of.. CBED budgets” to evaluate its performance despite acknowledging large

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uncertainties of in the CBED calculations. Is it known how large those uncertainties could be and what kind of biases they could cause in the estimates? The comparison shows some rather severe disagreements between FRAME and CBED dry and wet depositions, but which of those two should we trust more?

P. 22006: Summary and conclusions:

Again, much of the first paragraph is about sea salt implementation in the model, and nothing about other BC sources.

Lines 10-11 say “Maps of base cation emissions (Na+, Mg2+, Ca2+, K+), extracted from the SSA, were used as the input data in the FRAME model”. Are not sea salt is calculated online, along with the other BC components?

Line 20-21: “The larger scatter for calcium may be connected with underestimation of the land emission” - rather arguable statement; I’d say due to emission uncertainties. Could the authors say which of the land emissions (anthropogenic or natural) contribute most to calcium concentrations/ depositions and their uncertainties?

Line 22-23: “It is also possible that measurements for Ca2+ have greater uncertainty” - please explain why? And what kind of artefacts (positive/negative) are expected?

Figure 1: (a) explain the colours, (b)what radius? wet/dry? Also, the text on the axes and legends are rather difficult to discern - can be recommended to increase font size.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 21989, 2010.

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