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Atmospheric Chemistry and Physics Discussions

Interactive comment on "GEM/POPs: a global 3-D dynamic model for semi-volatile persistent organic pollutants – Part 2: Global transports and budgets of PCBs" *by* P. Huang et al.

Anonymous Referee #2

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General comments:

In a "first attempt with a 3-D global POPs transport model" (last paragraph) global budgets and intercontinental transport of three polychlorinated biphenyls (PCBs) are studied. The topic is of increasing interest. The paper introduces a novel budgeting approach. The contribution(s) lack(s) essential parts: Some environmental compartments (media) are captured by the models and others not, but the requirements and possible consequences of omittances are not identified. The model is introduced in part 1 and applied in part 2. The limitations of the methodology (essential) are not discussed in part 1 and the discussion of the results of the model experiment is very

deficient (part 2, see below). The way the study is presented is generally poor. Part 1 is so deficient that the quality of part 2 can hardly be assessed. Some of the text of part 1 is repeated in part 2. A reason to split the paper into two parts is not obvious for this reviewer.

Specific comments:

Introduction (parts 1 and 2): Large-scale multicompartmental modelling was developed in parallel on the atmospheric community and 'box-type' modellers' sides (unlike stated by the authors). The first global model of the first type of models was Strand & Hov, Water Air Soil Poll. 1996, and should be named here. As the historically first 3D global model, Koziol & Pudykiewicz, 2001, was from the same institution than the presented model, it should be explained how these two models relate to each other or what the main differences are (compartments, resolutions, exchange processes, initialisation data). The Max Planck model was published earlier, Semeena & Lammel, Fresenius Environ. Bull. 2003. In the next sentence it is claimed that 'box-type models' proved 'extremely useful (Ĕ) in demonstrating the grasshopper effect and cold condensation'. It seems, that the authors mix up key hypotheses and processes of global scale environmental chemistry such as 'cold condensation', 'global fractionation' and 'global distillation': The latter is a consequence of the combination of 'cold condensation' and other features. It was hypothesized by Goldberg, Proc. R. Soc. London 1975, and Wania & Mackay, Ambio 1993. The latter and later papers, e.g. Calamari et al., Environ. Sci. Technol. 1991, Ockenden et al., Environ. Sci. Technol. 1998, suggested that observed latitudinal trends could be explained by the grasshopper effect. The first attempt to verify/falsify the hypothesis by modelling was Semeena & Lammel, 2005. The statement that 'the investigation of the budgets and deposition patterns (Ĕ) lacks in these 3D studies' is not true, cf. e.g. Malanichev et al. 2004, Shatalov et al. 2005, Semeena et al. 2006. Presentation of the substances chosen (introduction of part 2): One substance is called volatile, two semivolatile. Why ? Substance properties, chemical formulae and degradation rates should be listed including references. Which

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uncertainties are coming with e.g. the choice / uncertainty of Henry coefficients (cf. Bruhn et al., Atmos. Environ. 2003) ?

Methodology / model description:

Several compartments are not mentioned (parts 1 and 2), namely vegetation, sediments, land and sea ice. If ignored, what is the consequence ? Why is this simplification considered to be justified ? These questions should be discussed in the context of earlier studies of PCB cycling (Dachs et al., Environ. Sci. Technol. 1999, Axelman & Broman, Tellus 2001, Malanichev et al. 2004).

Emissions: Are PCBs industrial chemicals only or are they also formed in combustion processes ? Which sources are considered in the emissions ? Why were these substances chosen ? Obviously, physico-chemical properties, observation and emission data are needed for such a modelling exercise. These would have been available for some pesticides, while for the chosen substances the emission data are certainly less well known than for pesticides (despite significant efforts to narrow these uncertainties down).

Atmosphere: Dry deposition of gases is obviously not covered although it is considered to contribute significantly to the overall atmospheric removal of SOCs. Why ? What is the consequence of this simplification ? Gas/particle partitioning: The Junge-Pankow approach is very deficient for lipophilic compounds (cf. e.g. Finizio et al., 1997). Therefore, the parameterisation is severely limiting the applicability of the model for the chosen study. At least needed is a justification why the Junge-Pankow approach was chosen instead of a Koa approach. Conclusions on the role of aerosols on the cycling of the heavier PCBs are not possible. Wet deposition: snow is not mentioned. What does this mean: No snow in the model world or is scavenging by snow treated like in rain? Clouds, although filling large parts of the troposphere, are not mentioned. Again, what does this mean ? No clouds in the model world ? Ocean: How many vertical levels have been considered in the model experiment ? How are vertical transports

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associated with marine particulate matter treated ? This question should be discussed in the context of earlier studies of PCB cycling, e.g. Dachs et al., Environ. Sci. Technol. 1999, Axelman & Broman, Tellus 2001, Malanichev et al. 2004.

Model run initialisation: 'spin-up run', 'assimilated' should be explained for non-modellers in the readership. The way observational data were assimilated should be detailed.

Comparison with observations: These results should be discussed in the context of the input data uncertainties. As it is probably covered by the emission uncertainties, a discrepancy of a factor of 10 does not indicate a disagreement. (?) Table 1 of part 1: soil types and soil levels should be addressed in the caption. Table 2 of part 2: is it measured or modelled ? Drop two or more digits. Fig. 2 of part 1: Caption needs to identify the parameter (probably atmospheric ground level concentration). Drop 'master'. Fig. 4 of part 1: Replace 'fractionation' in the caption by what it is supposed to be

Results: The results' presentation in part 2 is limited to the PCB concentration fields produced in one model simulation. Most of the results presented on intercontinental transport are well known as given by the large-scale atmospheric transports controlling the global scale distribution of tracers. The relevant knowledge should be briefly presented with appropriate citations. Then the new, in particular the substance specific insights should be presented. How do these contrast with the distributions of conventional pollutants ?

The study would greatly merit from discussing the results in the context of previous studies into PCB regional and global fate (e.g. Wania & Daly, Atmos. Environ. 2002). In Section 3.1 some findings are mixed with expectations in a confusing way. The vertical distributions of the tracers are a major asset of the study. It would greatly benefit from a table which quantifies the fractions accumulated above certain levels (including the tropopause). The long-range transport potential is addressed, partly in quantita-

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tive terms. Movements, e.g. meridional in Section 3.1, however, are not quantified. Migrations in degree north as a function of season and relative to the distributions of emission would be the adeaquate way. The last sentence of Section 3.1 addresses the very difference between environmental chemistry and passive tracer transport studies: It needs to be detailed and elaborated: which processes and properties favour which distribution trends ? The seasonal variations (end of Section 3.2) should be discussed in the context of the seasonal variations of the emission patterns. Generally, as none of the common LRTP indicators are used, the results on LRTP cannot be used to compare with the results of other studies of the same substances or between substances.

Figure captions: units should be harmonized (g and s or kg and years). (a), (b) not visible in Fig. 3. Table 1 of part 2: Caption needs specifications: is it about fluxes ?

Global budgets (Section 4 of part 2): The first sentence(s) of Section 4.1 should be part of a methodological section of the manuscript. The emission estimates' uncertainties should be quantified. The senarios should be explained more detailed.

More technical corrections: Sloppy written: Terminological and notation rules largely ignored and frequently poor grammar. Science communication requires to restrict to precise wording otherwise we are lost, which is what happens sometimes in this manuscript. The authors are advised to refer to accepted standard terminology (e.g. IUPAC, SI). Some examples for this type of weakness are: "physiochemical" and "physico/chemical" are there but none of these is an English word, sorry; "partition" is used as a verb, a noun and an adverb (please use "partitioning" as the noun exclusively in the title of 2.2.4 of part 1 and elsewhere); 'ND' should be replaced by upper bounds / limits of quantification in caption of table 1 of part 1 (cf. IUPAC rules); "exchange", although bidirectional by definition, is used unidirectional, too (title of Section 2.2.3 in part 1); concentrations are formulated as C, as [x] (even in the same equation, i.e. (2) of part 1) and called either concentrations or loadings (the latter is not recommendable as used with different meanings in different sub-communities); two Henry coefficients are used (in Section 2.2.2 of part 1) of which only one is identified (as dimensionless)

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but not really told what it is standing for; “fraction” and “fractionation” are used often without telling what it is: quotient (of what?), spatial or temporal trend ? Section 4.2 of part 1 (in combination with Fig. 4) is titled ‘Fractionation of PCBs’ but there is no chance for the reader to guess what it is about. In this case it does not help that the introduction seems to refer to global fractionation, although without explicitly saying so (mixed up with cold condensation ? see above). Section 4.3 of part 1 refers to ‘spring’ - even in the tropics. The following sentence lacks any logic. The long-range transport potential may be higher or lower but not ‘more’ (last sentence of abstract, part 2), the tropics inner or outer but not ‘most’. Please accept that particles together with the gas-phase are the aerosol and that the atmosphere contains gas, particulate matter and cloud water and ice. Hence, the word aerosol should be used when both phases are meant, ‘particulate phase’ when the condensed phase is meant only. ‘particles of 79 kg’, ‘total gas of 148 kg’ is inacceptable. Please use ‘multicompartmental” instead of “multi-compartment”. English grammar: almost no sentence which is grammatically correct. It is also unclear whether it is meant to be British, American or an other variety of the language (e.g. usage of ‘vapour’ and ‘vapor’ in the same text). It seems that a native speaking scientist and/or a professional language editor was not involved prior to submission, which is somewhat disappointing. Typos: ‘behaviours’, ‘potentials’, PM2.5 (use capitals), ‘meridinal’, ‘metrological’, ‘over three following areas’, many hyphens are misplaced many articles are lacking.

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Interactive Comment