

## ***Interactive comment on “Modelling atmospheric transport of persistent organic pollutants in the Northern Hemisphere with a 3-D dynamical model: DEHM-POP” by K. M. Hansen et al.***

**K. M. Hansen et al.**

Received and published: 30 June 2004

The authors would like to thank the anonymous referee for useful and constructive comments on the paper. Below follow reactions to the individual comments made by the referee.

Referee:

Title: The paper has a title that refers to this as a modelling study of persistent organic pollutants (POPs) [In plural]. In reality, only one single chemical is addressed in the paper (a-HCH). The authors should therefore consider changing the title of the paper by replacing POPs with a-HCH. This may seem like a minor issue, but there is no guarantee that the model will work equally well for other POPs (e.g. those POPs that are more sorbed to particles in the atmosphere).

Answer:

The title of the paper will be changed to: "Modelling atmospheric transport of alpha-hexachlorocyclohexane in the Northern Hemisphere with a 3-d dynamical model: DEHM-POP". See also comment (3) by referee # 1.

Referee:

Page 1340, lines 19-22: An additional criteria that is frequently used and referred to is toxicity.

Answer:

We will include the criteria "toxicity" in the list. The sentence is changed to: "The term persistent organic pollutants (POPs) is used to describe a group of chemical compounds with different origins but common characteristics: semi-volatility, hydrophobicity, bioaccumulation, toxicity and great persistence in the environment (Jones and de Voogt, 1999)".

Referee:

Page 1340, line 24 until page 1341, line 2: Strictly speaking, the environmental fate is also strongly affected by the environmental conditions (OC content etc) as well as temperature in addition to the physical-chemical properties listed.

Answer:

The section will be changed to: "The environmental fate of POPs is determined by environmental conditions such as temperature and soil organic carbon content and by key physical-chemical properties of the compounds, such as the aqueous solubility, the vapour pressure, and the partitioning coefficient between air and water (the Henry's law constant), octanol and water, and octanol and air, where octanol is used as a surrogate for the solid state (Jones and de Voogt, 1999)".

Referee:

[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)[Discussion Paper](#)

Page 1341, line 10: Please consider; POPs may have adverse health effects

Answer:

The sentence will be changed to: "POPs may have adverse health effects...".

Referee:

Page 1341, line 12; The term lipophilic is not very good as most organic chemicals are fairly equally soluble in fat and other organic phases. However, their hydrophobicity may be very different as expressed by Kow. Please, consider to change lipophilic with hydrophobic.

Answer:

We will change lipophilic to hydrophobic.

Referee:

Page 1341, line 14: Potential harmful effects?

Answer:

The sentence will be changed to: "This raises concern especially for the top-predator species due to the potential harmful effects".

Referee:

Page 1341, line 17: It is claimed that the atmosphere is the major pathway of POPs to the Arctic. This is a very firm statement, which I doubt is correct for all POPs in general, and it should therefore not be kept in the paper without references. See e.g. Li et al 2002. Sci Total Environ 291: 229-246.

Answer:

We will rephrase the sentence to: "The most important pathways of POPs to the Arctic environment are the atmosphere, the ocean and the fresh water system (AMAP,

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper

1998). The atmosphere constitutes the most rapid of these pathways; therefore the atmospheric concentration and transport are key factors in the study of the Arctic environmental fate of POPs".

Referee:

Page 1344, lines 6-10: Please consult Li et al 2003, Environ Sci Technol 37: 3493-3498 for more recent information. The authors may also want to consider emphasising: Technical HCH is historically the most used insecticide

Answer:

The estimates by Li et al. (2003) are now included and the sentence is rephrased to: "Technical HCH is historically the most used insecticide worldwide (Li et al., 2000)".

Referee:

Section 2.2.2 (page 1345): Atmospheric reaction is clearly a key process in this model. I miss a clear statement if spatial and temporal variability of OH-radical concentrations are taken into account, and if temperature dependent degradation rates have been considered. I think that the issue of atmospheric reaction of  $\alpha$ -HCH deserves to be discussed in some more detail in the paper, e.g. referring to the data presented by Brubaker and Hites, 1998 Environ Sci Technol 32: 766-769.

Answer:

We will change the section and include a discussion of the reaction rates presented by Brubaker and Hites (1998): "Reaction with OH radicals is assumed to be the most important degradation of  $\alpha$ -HCH in the atmosphere (Atkinson et al., 1999). The degradation rate depends on temperature and OH concentration in air as studied by Brubaker and Hites (1998). However, as an approximation a first order degradation rate is calculated in the model using an estimated mean residence time in the atmosphere due to reactions with OH radicals of  $\tau = 1/(118 \text{ days})$  (Mackay et al., 2000). This value is in agreement with an average atmospheric lifetime of 120 days calculated from

[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)[Discussion Paper](#)

the reaction rates measured by Brubaker and Hites (1998). The degradation rate is applied for the whole model domain and is not seasonal dependent. A transformation of gamma-HCH into alpha-HCH by UV radiation has been speculated (Pacyna and Oehme 1988), but is not taken into account in the model since no direct observation has been made of this reaction in the environment".

Referee:

Degradation rates in soil (p 1347, line 18) and water (p 1348, line 12): It is unfortunate that the authors simply adopt these parameters as used in the earlier modelling study by Strand and Hov without any further attempt to discuss the literature on the subject and discuss these input parameters in some more detail.

Answer:

We will change the two sections so they include a discussion of the literature on this subject: "alpha-HCH is subject to biodegradation in soil. This process is not well quantified and the degradation rate in soil is estimated to be:  $k_{\text{soil}} = 1/(1\text{year})$  (Strand and Hov, 1996). This value is generally higher than degradation rates used in other models. A half-life in soil of 120 days at 20 °C, increasing with decreasing temperatures, is used by Wania et al. (1999) and Breivik and Wania (2002). Degradation rates of  $1/(3\text{ months}) - 1/(4\text{ months})$  are used for the temperature range 270 - 301 K by Scheringer et al. (2000)".

"Apart from re-volatilisation, alpha-HCH is lost from the surface compartment by hydrolysis, biological degradation and particle settling. None of these processes is well quantified, and a simple degradation rate of  $k_{\text{ocean}} = 1/(10\text{ years})$  is used (Strand and Hov, 1996). This value is generally higher than estimates used in other models. A half-life of 4 months for ocean water and 3 years for fresh water is used by Wania et al. (1999). Breivik and Wania (2002) use a half-life of 1 year for both ocean and fresh water. Degradation rates of  $1/(5.4\text{ years})$  to  $1/(1.8\text{ months})$  is used for the temperature range 270 – 301 K by Scheringer et al. (2000)".

[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)[Discussion Paper](#)

Referee:

Page 1349, line 13 versus line 26. First a-HCH is described as a compound having great persistence, and next the relative short lifetime in soil is emphasised. This is somehow contradictory.

Answer:

We will omit line 26.

Referee:

Page 1352, line 21. Seasonality is discussed in terms of re-volatilisation processes, but the possible impact of seasonality of emissions is not mentioned in this paragraph.

Answer:

The importance of seasonality in emissions is discussed in the section on page 1354 lines 9-14.

Referee:

Page 1354, line 10. The important assumption of uniform emissions throughout a year should have been explicitly mentioned already in section 2.4.2.

Answer:

This assumption is mentioned in section 2.4.2 on page 1349 lines 2-3.

Referee:

Page 1355, line 11. This is wrong. There have been other dynamical models used to study a-HCH as dynamic may be interpreted as non-steady state, thus facilitating an analysis of the environmental response to changes in emissions. Additional examples are e.g. Wania et al 1999 and Toose et al 2004 as listed in the paper, as well as e.g. Breivik and Wania, 2002 Environ Sci Technol 36: 1014-1023.

[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)[Discussion Paper](#)

Answer:

The use of the word "dynamic" in this context refers to the dynamic meteorological input used to drive the model, i.e. non-averaged data. Since we now realise that the word dynamic also can be interpreted as non-steady state, we will rephrase this sentence, so the meaning will become clear: "Only one atmospheric transport model using dynamical meteorological data as input was used to study the environmental fate of alpha-HCH (Koziol and Pudykiewicz 2001)".

We will also include the results from Breivik and Wania (2002) in the comparison section 4.1 on pages 1354-1355: "A regional scale model covering the Baltic Sea region based on the global distribution model of Wania et al. (1999) was used to study the environmental fate of alpha-HCH for the years 1970-2000 (Breivik and Wania, 2002). This model obtains very good predictions of both seasonal averaged and individual measured air concentrations for the 1980s and 1990s (Breivik and Wania, 2002)".

Referee:

Page 1355, line 4-5. It is claimed that the results in this paper are better than the results from previous models (assuming that this refers to previous attempts using multimedia box models in section 4.1). The term "better" is certainly a relative measure as different models have been developed and used to answer different questions. If the authors really want to keep this firm statement, they should be really cautious and critically compare and contrast how their overall modelling approach and specific results may be considered superior to past studies. As stated by Wania (1999) it is important to note that the low spatial and temporal resolution of multimedia box models often is a deliberate restriction rather than a regrettable shortcoming. This is based on the belief that the predictive capability of numerical models of environmental POP behaviour is not limited by the resolution of atmospheric transport processes, but rather by the uncertainties inherent in emission estimates, physical-chemical properties, degradation rates, and air-surface exchange descriptions of POPs (all of which are correctly recognised as

[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)[Discussion Paper](#)

uncertain parameters and processes in this paper). Adding complexity in the presence of such uncertainty does not necessarily improve a model nor make it better. It would therefore probably be better to regard this study as complementary to past studies, rather than better? Wania, F., 1999: Differences, Similarities, and Complementarity of Various Approaches to Modelling Persistent Organic Pollutant Distribution in the Environment. WMO/EMEP/UNEP Workshop on Modelling of Atmospheric Transport and Deposition of POPs and Heavy Metals, Geneva, Switzerland, 16 -19 November 1999. [http://www.wmo.ch/web/arep/reports/gaw136\\_vol1.pdf](http://www.wmo.ch/web/arep/reports/gaw136_vol1.pdf)

Answer:

We acknowledge the unfortunate use of the word "better" in this sentence. The aim was to emphasise that DEHM-POP is capable of predicting measured air concentrations with as high accuracy as previous models or higher in some cases which is shown by the comparison in section 4.1 on pages 1354-1355. We will rephrase this sentence to: "These results are of equal or higher accuracy than results from previous models describing the environmental fate of alpha-HCH". We will furthermore expand the section on page 1355 line 8-10 concerning the comparison with the box model type models with a discussion of the Wania (1999) paper: "Overall, there is a fairly good agreement between DEHM-POP and the four box models despite the differences in spatial resolution and process description. However, it is important to note that the two types of models should not be seen as competitors but rather as complementing each other, as discussed by Wania (1999)".

---

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 1339, 2004.

[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)[Discussion Paper](#)