Interactive comment on "Biannual cycles of organochlorine pesticide enantiomers in arctic air suggest changing sources and pathways" by T. F. Bidleman et al.

Anonymous Referee #3

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This manuscript present the results from the re-analysis of air sample extracts for selected chiral OC pesticides from the Canadian air toxics sampler at Alert (Canadian Arctic). The resulting time-series of enantiomeric fractions (EFs) of three chiral OCs are presented and discussed for the 7 year period, 1994-2000. This is exactly the sort of use that a sample archive like this should be subjected to, as EFs are not dependent on sample/solvent integrity etc over the storage period. Given the number of samples/data points for each year of study (n=10s in each case), then this gives a statistical robustness to the observed trends, and it's nice to see use of a trend-fitting model like DF applied this dataset. The core of the discussion is related to understanding the annual variation in EF values, relating seasonal shifts in EFs in arctic air to changes in emissions from secondary sources (soil, water). This is a unique paper and one of a very few to actually report a multi-year time series of EF data. The discussion gives diagnostic insight into contaminant ageing and source type to be affecting the Arctic in the late 1990s. I have no reservations about this manuscript and recommend publication in ACP. The paper is very well written with an excellent narrative style that ends by identifying a clear research need (I'm already looking forward to Part II, 2001-2011!).

Comment: 3.2 Chlordanes (p25038, lines 15-24). In the discussion about TC and CC sources and the role of the temperate ocean, the authors report the preferential depletion of (+)TC and (-) CC in recent water samples (2008) from the N. Atlantic (Zhang et al.2012b). It would be useful if the actual EF values from this study (e.g. mean and range) were reported here, allowing direct comparison to the EF values reported for the Alert air samples in the 1990s. This may help to discount/support the role of northern temperate marine waters as a source of these chemicals to arctic air.

Response: Mean EFs and standard deviations are now included (page 10, lines 18-23).

Suggestion: Is it possible to illustrate EF data for each of the OCs as a box-and-whisker plot? As the n-values are high, plus there doesn't appear to be an underlying multi-year trend in the EFs, then lumping all the annual datasets into 'winter-spring' and 'summerfall' categories would really emphasize the seasonality, perhaps better than a modelled DF trend. Measured outliers could then be investigated. For example, if low value EFs for a-HCH (outliers < 10th percentile) all occurred in July-August (coinciding with ice melt/ice free periods) then this is compelling evidence to suggest a regional arctic seawater signal (akin to the ice-seawater-air work by Wong et al 2011). Ditto, outliers > 90th percentile may be linked to LRT events from afar.

Response: This is a good suggestion, and we have added box-and-whisker plots to the main paper as Figure 1. See response to Reviewer 1. This figure has also been reproduced below for purposes of the interactive discussion. There is difficulty relating "outliers to LRT events from afar", because the air trajectories were only run for 72 hours, and most of them did not get out of the arctic region within that time. See Figure S1 of Supporting Information.

1. *Box-and-whisker plots of monthly EFs over all 7 years: arithmetic mean (square), median (horizontal line), 10th – 90th percentiles (whiskers), 25th-75th percentiles (boxes) and outliers (crosses). The dashed red line indicates a racemic composition.*