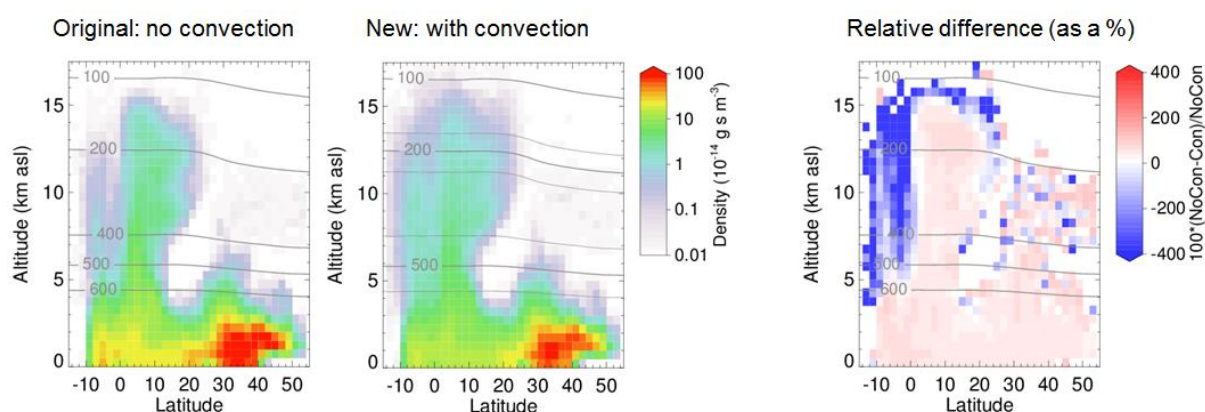


Referee comments are given in black; author comments are in red.

Since submission of this paper to ACPD we have had the opportunity to repeat the calculation described in section 5 using an improved version of the NAME model. The model now contains a more detailed parameterisation for convective transport [see Meneguz and Thomson, 2014, DOI: 10.1504/IJEP.2014.065113], which is clearly important for the topic we have studied.

We find that the results using the new model version are very similar. An example is given in the figure below, which shows from left to right: our original Fig. 5c (no convection scheme), our proposed new version of this plot (with convection scheme), and the relative difference between original and new, expressed as a percentage. The final plot shows that using the convection scheme results in slightly stronger uplift of trajectories (areas shaded blue).



Despite the relatively small differences, we believe showing these new results in a revised paper would be more intellectually rigorous, and therefore plan to do so. As the proposed change does not affect our conclusions, we will not need to make major changes to the text – we only plan to slightly modify our description of the model (section 3, para 1), which we have also been asked to do by referee two.

We will also add Elena Meneguz of the UK Met Office (who developed the new convection parameterisation and performed the new calculation) as a co-author on the revised paper.

We now reply in turn to the comments from the two anonymous referees.

Anonymous Referee #1

This manuscript presents a time series of observations of perchloroethene at a southeast Asian site and connects weekly-scale variations in these observations to variations in air mass source distributions related to cold surges. The authors then provide evidence for subsequent transport of polluted air masses to the tropical upper troposphere. The data and methodology are well-constructed and clearly described. The presentation is concise and generally quite tight, although I feel the authors have erred on the side of brevity in a few instances (see suggestions below for p.30711 and 30713). The text is well-written and the core arguments are easy to follow. I recommend that this manuscript be accepted subject to a handful of mainly editorial corrections.

We thank the referee for their positive comments and address comments individually below.

p.30707, 1.1: I think this sentence would work better as two sentences, split between "recent decades" and "under the prevailing". In either case, I recommend changing the order of the "under the prevailing..." and "these increases have..." phrases.

We will make the suggested change.

p.30709, 1.17: suggest moving "to date" from the beginning to the end of the sentence

We will make this change.

p.30709, 1.24: it would be useful to specify the beginning and ending dates of the timeseries shown in Fig. 1

We will now write: "The data we consider here were collected during one NH winter, between 22 November 2008 and 28 February 2009, and have been averaged over 3 h periods (Fig. 1a)."

p.30709, 1.25: "clear week-scale variations in the data" -> "clear variations in the data at weekly scales"?

We will make the suggested change as part of a revised sentence that also addresses a comment from referee two.

p.30709, 1.26: comma after "Fig. 1b)" is not necessary and disrupts the flow of the sentence

We will remove the comma

p.30709, 1.27: recommend changing format of clarifications from ", of ~1.0–1.5 ppt," to "(~1.0–1.5 ppt)" for easier reading

We will make this change, and a similar change for ~2.0-3.0 ppt at the end of this sentence

p.30710, 1.4: "...shows there..." -> "...shows that there..."?

We will make this change

p.30710, 1.6: "but Robinson et al. (2014) do discuss" is a little awkward, maybe ", although Robinson et al. have raised"?

We will make this change

p.30711, 1.1: causation seems backwards here (the change in mixing ratios is caused by the change in parcel source regions, but this sentence reads as though it causes the change in parcel source regions); the sentence should be revised for clarity

We will now write: “As a larger fraction of trajectories travel from mid-latitudes, passing through the Philippines, the C2Cl4 mixing ratio at Tawau increases. Beyond the Philippines, there are two diverging origins.”

p.30711, 1.12: replace "does" with "increases" for clarity

We will make this change

p.30711, 1.17: "focussed" – typo or British spelling?

We are happy to change this to “focused”.

p.30711, 1.25: It would be useful to add a sentence or two briefly describing what contributes to the MACC estimates of CO and O3 in this region. Given that the interpretation is qualitative rather than quantitative, this description need not be especially detailed, but it should be enough to convince the average reader that MACC is a suitable tool for this analysis.

We suggest adding the following sentences: “This dataset is created using satellite observations, emission inventories and chemical transport model calculations. For the two pollutants we consider in this section, CO and O3, Inness et al. (2013) report small negative biases of ~10% and ~20% respectively when the MACC data are compared to available independent observations in the tropical troposphere.”

p.30712, 1.9-11: awkward phrasing, could change to something like "In Fig. 4 we contrast daily mean maps of CO and O3 from the MACC reanalysis (again at 925 hPa) for the two days shaded in Fig. 3: 15 January 2009, when concentrations of C2Cl4 were relatively high, and 20 January 2009, when concentrations of C2Cl4 were relatively low."

We will make this suggested change.

p.30712, 1.19: recommend splitting this sentence into two between "ppb" and "our"

OK – we will make this change.

p.30712, 1.20-23: awkward phrasing, could change to something like "By contrast, simulated levels of CO and O3 and measured levels of C2Cl4 are more representative of the local background (approximately half of the polluted levels) on days when the winds blow from the Pacific, such as 20 January (bottom row of Fig. 4)"

We will make this change.

p.30713: I question the decision not to show the corresponding plot for 200 hPa and/or the corresponding tracer density plots for 20 January. The presentation is already quite concise, so I think that there is room to include these. Leaving them out inspires questions (how strong is the agreement at 200 hPa? how much weaker is the evidence for this type of vertical transport?) that could very easily be addressed by including them.

We are happy to include the additional plots requested by the referee in a revised manuscript. We plan to show time-longitude cross sections for both ~500 hPa and ~200 hPa in a new Fig. 5 and tracer density plots for 20 January (in addition to 15 January) in a new Fig. 6 (4 panels in total). This addition will also require some minor re-wording of the final two paragraphs of p.30713.

p.30714, 1.28: "In order to quantify better" -> "In order to better quantify"?

OK, we will make this change.

p.30715, 1.14-15: awkward phrasing, could change to something like "Further longterm measurements will also facilitate more detailed investigation of the influences of the climatic variations discussed above."

We will make this change.

p.30725: the first sentence in the caption to Fig. 4 is difficult to follow, suggest changing to something like "Daily mean maps of air history calculated using NAME (left), CO at 925 hPa from the MACC reanalysis (center) and O3 at 925 hPa from the MACC reanalysis (right) for 15 January 2009 (top) and 20 January 2009 (bottom)."

We will also make this suggested change.

Anonymous Referee #2

The paper presents observations of marked intra-seasonal variability in the anthropogenic tracer perchloroethene (C₂Cl₄) collected in Borneo during the NH winter of 2008/09. Observed enhancements in C₂Cl₄ are caused by rapid meridional transport. Trajectory calculations show that these polluted air masses are from East Asia and can subsequently be lifted to the tropical upper troposphere. Data from the Monitoring Atmospheric Composition and Climate reanalysis support the results. The paper suggests a potentially important connection between mid-latitude pollution sources and the very low stratosphere.

The paper is significant and appropriate for publishing by ACP. The conclusions and potential impacts of the paper are clear. It is well written, however at some points the authors should be more precise or add some details. The paper is worth to be published in ACP, after minor corrections.

We thank the reviewer for their positive review and address comments below.

General comments:

I recommend to emphasize stronger in abstract and summary that cold surges cause both the meridional transport from the north and the enhanced uplift in the tropics. This is mentioned in the text, but I think the authors should expressly underline this result of the study.

We have re-written a sentence in the abstract as follows: “In addition to strengthening the meridional transport from the north, cold surges can enhance convection in Southeast Asia, and further trajectory calculations indicate that the polluted air masses can subsequently be lifted to the tropical upper troposphere.” We are happy with what we have written in the summary in this case, so do not propose any changes there.

Further, a lot of details are discussed in Section 6 (Summary and discussion), but it is not mentioned the important results about the transport times of pollution from tropospheric sources in East Asia into the upper tropic troposphere. I propose to add this in the summary.

We will add the following sentence to the first paragraph in the summary: “Our trajectory calculations suggest that in total, transport from the East Asian boundary layer to the tropical upper troposphere (above 200 hPa) can occur in fewer than 10 days.”

1 Introduction, p. 30708, lines 20: ‘Strong uplift of polluted air masses, and an associated impact on stratospheric composition, has already been demonstrated during the Asian (NH) Summer Monsoon (Lawrence and Lelieveld, 2010; Randel et al., 2010).’ However, the mechanisms for transport into the stratosphere are subject of current debate (see also e. g. Park et al. JGR, 2009; Bourassa et al., Science, 2012, ...)

Agreed, we are happy to add some extra detail to this effect.

We plan to revise this sentence, and re-order the paragraph, as follows: “It is also interesting to consider the possibility of pollutant transport via convection. For example, while the detailed mechanisms for transport into the stratosphere are the subject of current debate (e.g. Park et al., 2009; Bergman et al., 2013), strong uplift of polluted air masses has already been demonstrated during the Asian (NH) Summer Monsoon (Lawrence and Lelieveld, 2010; Randel et al., 2010). Such a process may also be important in the context of our study because Southeast Asian air masses are preferentially lifted towards the stratosphere during NH winter (e.g. Levine et al., 2007; Aschmann et al., 2009), and because of the link between cold surges and convection outlined above.

1 Introduction, p. 30708, lines 24-36: 'East Asian pollution has also been shown to affect atmospheric composition further afield, in both western North America (Cooper et al., 2010) and Hawaii (Lin et al., 2014). ' For which seasons is that valid?

We will add detail on seasons as follows: "East Asian pollution has also been shown to affect atmospheric composition further afield, leading to increased O₃ in both western North America during Northern Hemisphere (NH) spring (Cooper et al., 2010) and Hawaii during NH autumn (Lin et al., 2014)."

1 Introduction, p. 30708, line 5: Please add 1-2 sentences to explain in more detail "cold surges". (...transport of cold air from the north... ?)

We will add detail to this paragraph as follows, with the aim of also addressing a subsequent referee comment related to the impact that cold surges have on convection in Southeast Asia: "These cold surges are typically caused by a southeasterly movement of the Siberian High pressure system, and are associated with movement of cold air masses towards Southern China and a strengthening of the northeasterly monsoon winds in the South China Sea (Zhang et al., 1997; Chan and Li, 2004; Chang et al., 2004). Cold surges are also known to increase convective activity generally in equatorial Southeast Asia, with enhancements in the region around the northwest coast of Borneo often particularly marked (e.g. Slingo, 1998; Compo et al., 1999; Chang et al., 2005). However, their influence on atmospheric composition in the region has yet to be demonstrated."

2 Observations, p. 30710, line 3: 'We have observed similar features in measurements collected in Borneo during subsequent winters...' Please add which winters? (2009-xx)

We will re-write the sentence so that it is clear we have also collected data in the two subsequent winters (2009/10 and 2010/11).

3 Transport pathways: Which vertical velocities are used in NAME?

The vertical velocities are taken from the Unified Model – we will make this clearer by editing the first paragraph of section 3 as follows: "Trajectories are calculated using three-dimensional meteorological fields produced by the Met Office's Numerical Weather Prediction tool, the Unified Model (UM). These fields have a horizontal resolution of 0.5625° longitude by 0.375° latitude, 31 vertical levels below ~19 km, and are available at 3 h intervals. The vertical velocities are obtained from the UM and available at grid nodes, while sub-grid-scale processes are parameterised in NAME. This is the case for turbulence (Webster et al., 2003; Morrison and Webster, 2005) and convection (currently available only in forward mode, see Meneguz and Thomson, 2014a, b).

3 Transport pathways, p. 30711, line 15: What are the typical transport times from > 35N to Borneo?

In days without cold surges the majority of (12 day) back trajectories do not travel to Borneo from north of 35N – this is evident in Fig 2. We will modify the final sentence in this section as follows: "This reinforces the argument made above: cold surges are able to move polluted air from northern mid-latitudes rapidly (over ~4 days in the case study we consider in Sect. 4) to equatorial Southeast Asia."

4 Wider air quality implications, p. 30711, line 20: 'We are not aware of any continuous air quality measurements in Northern Borneo that are unaffected by local pollution...' What is the impact of local sources on the C₂Cl₄ observations?

We suggest adding the following sentence in section 2 to highlight this point: “This coherence is indicative of changes in composition occurring over relatively large scales, and suggests very local emissions of C2Cl4 do not have an important influence on the data.”

4 Wider air quality implications, p. 30712, line 12: ‘Corresponding daily mean air history maps, in which transport timescales are marked to highlight the strength of the cold surge events, are also presented.’ Please add the transport timescales in the text.

We will now write: “Corresponding daily mean air history maps are also presented. In these maps the mean horizontal locations of the back trajectories after 2 and 4 days are marked to highlight the strength of the cold surge event.”

5 Uplift of polluted air masses, p. 30712, line 25: ‘Cold surges are known to affect the intensity of convection in Southeast Asia (e.g. Chang et al., 2005).’ Please explain briefly how the convection is affected. What exactly is enhanced? The convection height or its spatio-temporal distributions? Is there a shift of the location of convection by the cold surges?

We plan to add the requested detail, as described in our response to a referee comment above, to the introduction (and will remind the reader of the introduction in this particular sentence).

5 Uplift of polluted air masses, p. 30713, line 1/2: ‘..analysing this type of vertical transport ..such transport’ What means by ‘this type of vertical transport’? Convection? Is the convection in NAME described by the upward transport in the Unified model?

As noted in our response above, we have revised our description of the model, with particular reference to vertical transport by a combination of resolved vertical velocities from the UM and the new NAME convection scheme. We will clarify this sentence as follows: “Previous work suggests that NAME is a useful tool for analysing vertical transport of relatively short-lived compounds in regions of tropical convection (Ashfold et al., 2012).”

5 Uplift of polluted air masses, p. 30713, line 4: ‘surface box..’ Please add ‘over East Asia’
We will make this change.

5 Uplift of polluted air masses, p. 30713, line 6: ‘The aim was not to simulate any particular pollutant,..’ Does that mean that ‘Chemistry is not calculated along the trajectories’ ?

Yes – the trajectories are inert. We will clarify this as follows: “These trajectories were not subject to any chemical losses and their travel was recorded for 12 days.” We will also clarify in the text that the backward trajectories described in Sect. 3 were inert.

5 Uplift of polluted air masses, p. 30713, line 7: ‘originating in the polluted mid-latitudes’
Please add ‘in East Asia’

The same in line 16.

We will make both these changes.

6 Summary and discussion, p. 30714, line 19: ‘Its importance needs to be investigated further.’ What would be possible impacts of enhanced C2Cl4 in the stratosphere?

C2Cl4 alone is unlikely to have a large impact in the stratosphere, but air masses that are high in C2Cl4 are also likely to contain high levels of various other pollutants. To clarify this point in the text, we will write: “To illustrate the potential importance of this route, air masses enriched in C2Cl4 may also contain high levels of various other relatively short-lived chlorine containing gases (e.g. Wang et al., 2014) that, in sum, could have a negative impact on stratospheric ozone. The significance of this transport pathway therefore needs to be investigated further.”

Figure 2:

What is the altitude/pressure range of the trajectories?

These back trajectories are started near the surface (0-100 m) but, as noted in the legend, the maps in Figure 2 show trajectory density between 0-18 km. In practice, few trajectories actually leave the lower troposphere.

Minor comments:

Observations, p. 30709, line 1: Please add 'Perchloroethene C₂Cl₄'...

We have already spelt out the chemical name for C₂Cl₄ in the final paragraph of the introduction, and so do not think we need to do so again.

Observations, p. 30709, line 26: 'r² = 0.85).' 'r' is not introduced.

We will add this information.

5 Uplift of polluted air masses, p. 30713, line 3: 'introduce ..to the upper tropical troposphere' -> 'uplift ...to the upper tropical troposphere'

We will make this change

5 Uplift of polluted air masses, p. 30713, line 4: ' kilometre' -> 'kilometer'

OK

Figures:

In general, r and n is not always explained in the figure captions.

We will add an explanation to the caption

Figure 4:

The blue circle is difficult to see?

We will modify the figure to make the location marker larger

Figure 5b:

What is the meaning of 'km asl'?

km above sea level – we will add this explanation to the appropriate figure caption.