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Rapid transport of East Asian pollution to the deep tropics

M. J. Ashfold[1,*], J. A. Pyle[1,2], A. D. Robinson[1], E. Meneguz[3], M. S. M. Nadzir[4,5,6], S. M. Phang[6], A. A. Samah[6], S. Ong[7], H. E. Ung[7], L. K. Peng[8], S. E. Yong[8], and N. R. P. Harris[1]

[1]Department of Chemistry, University of Cambridge, Lensfield Road, Cambridge, CB2 1EW, UK
[2]NCAS, UK
[3]Met Office, FitzRoy Road, Exeter, EX1 3PB, UK
[4]School of Environmental and Natural Resource Sciences, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia
[5]Centre for Tropical Climate Change System (IKLIM), Institute for Climate Change, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia
[6]Institute of Ocean and Earth Sciences, University of Malaya, 50603 Kuala Lumpur, Malaysia
[7]Global Satria Life Sciences Lab, TB 12188, Taman Megajaya Phase 3, 91000 Tawau, Sabah, Malaysia
[8]Malaysian Meteorological Department (METMalaysia), Global Atmosphere Watch (GAW) Danum Valley, 91112 Lahad Datu, Sabah, Malaysia
[*]now at: School of Biosciences, University of Nottingham Malaysia Campus, Jalan Broga, 43500 Semenyih, Selangor, Malaysia

Correspondence to: M. Ashfold (matthew.ashfold@nottingham.edu.my)

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Abstract. Anthropogenic emissions from East Asia have increased over recent decades. These increases have led to changes in atmospheric composition as far afield as North America under the prevailing westerly winds. Here we show that, during Northern Hemisphere (NH) winter, pollution originating in East Asia also directly affects atmospheric composition in the deep tropics. We present observations of marked intra-seasonal variability in the anthropogenic tracer perchloroethene (C_2Cl_4) collected at two locations in Borneo (117.84° E, 4.98° N and 118.00° E, 4.22° N) during the NH winter of 2008/2009. We use trajectories calculated with the Numerical Atmospheric-dispersion Modelling Environment to show that the observed enhancements in C_2Cl_4 mixing ratio are caused by rapid meridional transport, in the form of "cold surges", from the relatively polluted East Asian land mass. In these events air masses can move from ∼35° N to Borneo in 4 days. We then present data from the Monitoring Atmospheric Composition and Climate reanalysis which suggest that air masses high in C_2Cl_4 may also contain levels of the pollutants carbon monoxide and ozone that are approximately double the typical "background" levels in Borneo. 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. This suggests a potentially important connection between midlatitude pollution sources and the very low stratosphere.

1 Introduction

The rapid growth of East Asian economies over recent decades has led to enhanced emissions of various pollutants, including ozone (O_3) precursors (e.g. Granier et al., 2011) and halocarbons (e.g. Wan et al., 2009). The emitted pollutants are known to be transported eastward by the prevailing midlatitude winds, and plumes have been observed at the edge of East Asia (e.g. Akimoto et al., 1996; Tanimoto et al., 2008) and over the Pacific (e.g. Liu et al., 2003; Hudman et al., 2004). East Asian pollution has also been shown to affect atmospheric composition further afield, leading to increased O_3 in both western North America during Northern Hemisphere (NH) spring (Cooper et al., 2010) and Hawaii

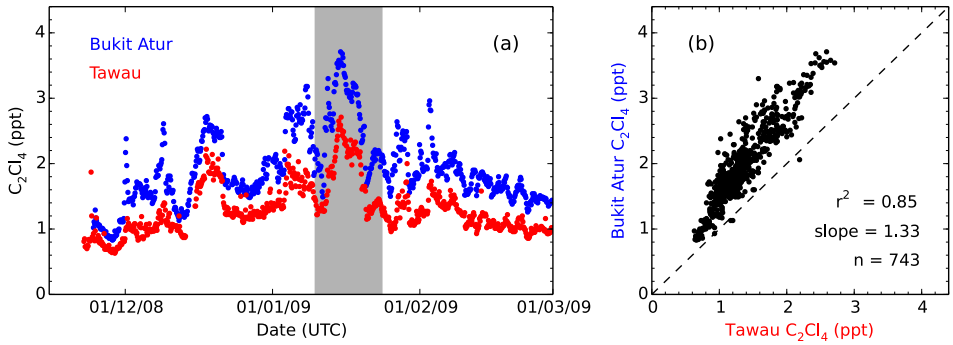

Figure 1. (a) shows a time series of 3 h mean C_2Cl_4 measurements collected at Bukit Atur (blue) and Tawau (red) during the winter of 2008/2009. The period analysed in more detail in Section 4 is shaded grey. **(b)** shows the correlation between the measurements at the two sites. The coefficient of determination (r^2), the gradient of the regression line (slope) and the number of 3 h periods for which data exist at both measurement locations (n) are noted. In constructing this figure, two outlying 3 h mean measurements from Tawau (>6 ppt) have been discounted. Both high mixing ratios are likely to be due to a very local C_2Cl_4 source.

during NH autumn (Lin et al., 2014). Southward transport of polluted air masses originating in East Asia, associated with the Northeast Monsoon which occurs during NH winter, has also been reported (e.g. Liu et al., 2003; Pochanart et al., 2003; Wang et al., 2003).

Here we describe an additional impact of East Asian pollution and show that particularly strong meridional transport events within the Northeast Monsoon, or "cold surges", are able to transport polluted air masses to remote parts of equatorial 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 near the northwest coast of Borneo often highlighted (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.

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.

Our analysis is based on observations of an anthropogenic halocarbon, perchloroethene (C_2Cl_4), collected in Borneo during the winter of 2008/2009 (Sect. 2). We conduct simulations with a dispersion model to confirm the influence of East Asian pollution on our measurements (Sect. 3). We then investigate the wider air quality implications of these transport events by studying data from the Monitoring Atmospheric Composition and Climate (MACC) reanalysis (Sect. 4). In Sect. 5 we use further model calculations to investigate whether the polluted air masses are lifted towards the upper troposphere once they have reached the deep tropics. Finally we discuss the implications of our results (Sect. 6).

2 Observations

C_2Cl_4 is an excellent marker of air masses recently subjected to industrial emissions for several reasons. First, the sources of C_2Cl_4 are predominantly anthropogenic, and largely aseasonal. They span a range of commercial and industrial activities, which include dry-cleaning and metal degreasing (McCulloch et al., 1999). Global emissions, and background mixing ratios, have declined over the past 20 years or so (McCulloch et al., 1999; Simpson et al., 2004, and data available at http://agage.eas.gatech.edu/data.htm). Nevertheless, there is evidence for continued and significant C_2Cl_4 emissions from East Asia over this period (e.g. Barletta et al., 2006, 2009; Shao et al., 2011). Second, C_2Cl_4 has a straightforward atmospheric chemistry which is dominated by reaction with the hydroxyl radical (Singh et al., 1996). Under typical tropospheric conditions its lifetime is ∼3 months (Montzka and Reimann, 2011). This lifetime is both long enough for large-scale transport and short enough for measurable inhomogeneities to exist. Finally, it appears that East Asian C_2Cl_4 emissions are similarly distributed to the anthropogenic portion of carbon monoxide (CO, a key ozone precursor) emissions in the region (e.g. Wang et al., 2003; de Gouw et al., 2004; Shao et al., 2011).

Few continuous measurements of atmospheric composition have been reported in Southeast Asia to date. To fill

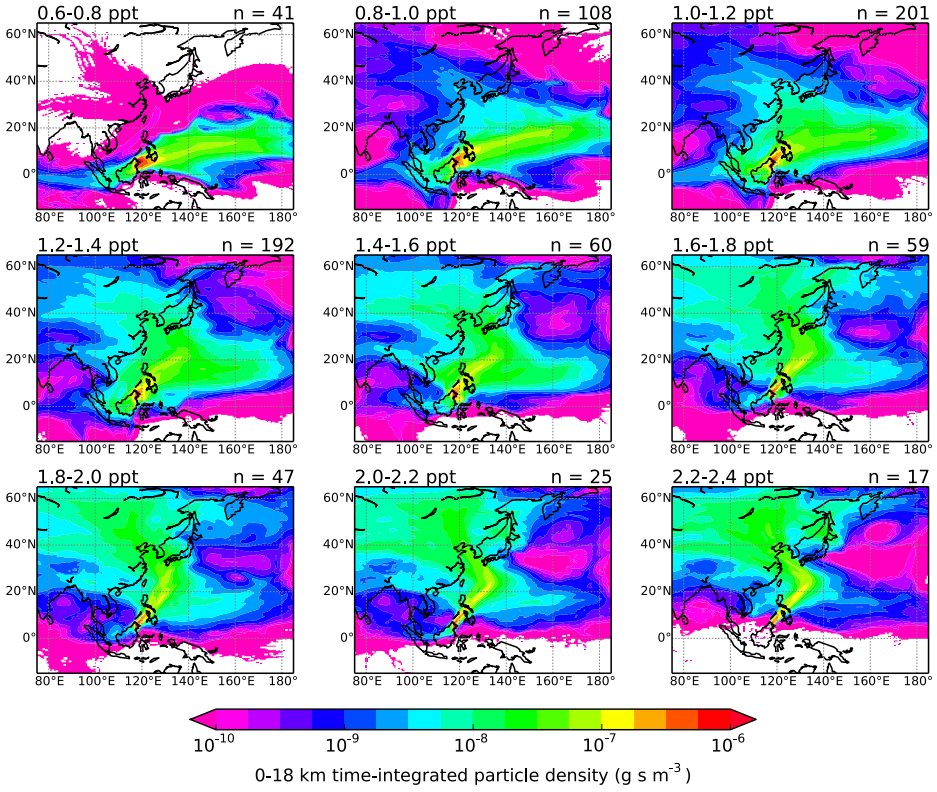

Figure 2. Composite air history maps for each C_2Cl_4 mixing ratio interval of 0.2 ppt at Tawau. The number of 3 h periods, n, contributing to each composite is noted above each panel. A mixing ratio greater than 2.4 ppt was observed in eleven 3 h windows; these periods are not accounted for in the figure.

this gap, we have used University of Cambridge μ-Dirac instruments (Gostlow et al., 2010) to measure a suite of halocarbons at a number of sites in the region (see Pyle et al., 2011; Robinson et al., 2014). Here we focus on C_2Cl_4 observations collected at two locations in Sabah, Malaysian Borneo. The sites, the WMO Global Atmospheric Watch station at Bukit Atur (117.84° E, 4.98° N) and a Global Satria facility near Tawau (118.00° E, 4.22° N), are ∼85 km apart. 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). There are clear variations in the data at weekly scales, which occur concurrently at the two sites (coefficient of determination, $r^2 = 0.85$ in Fig. 1b). This coherence is indicative of changes in composition occurring over relatively large scales, and it suggests that very local emissions of C_2Cl_4 do not have an important influence on the data. Our measurements are also characterised by a number of abrupt changes between "background" mixing ratios (∼1.0–1.5 ppt) and "polluted" mixing ratios (∼2.0–3.0 ppt). We will show in Sect. 3 that changes in Northeast Monsoon circulation are the primary cause of these transitions. We have observed similar features in measurements collected in Borneo during the two subsequent winters (2009/2010 and 2010/2011) and are therefore confident these changes are typical of the region.

Figure 1 shows that there are some differences in the absolute mixing ratio observed at the two sites. Instrument performance is generally good for this compound (Gostlow et al., 2010) although Robinson et al. (2014) have raised the possibility of calibration uncertainty leading to part of this difference. In this study we focus on the variations observed concurrently at both locations, and hence the absolute mixing ratios are of lesser importance.

3 Transport pathways

We use the Met Office's Numerical Atmospheric-dispersion Modelling Environment (NAME, see Jones et al., 2007), a Lagrangian dispersion model, to interpret our C_2Cl_4 measurements. 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 grid resolution of 0.5625°longitude by 0.375°latitude and 31 vertical levels below ∼19 km, and they are available at 3 h intervals. Vertical velocities are obtained from the UM and available at grid nodes. The sub-grid-scale processes of convection (currently

available only in forward mode; see Meneguz and Thomson, 2014a, b) and turbulence (available in both forward and backward modes; see Webster et al., 2003; Morrison and Webster, 2005) are parameterised in NAME.

Initially we use NAME to calculate batches of inert backward trajectories started at each measurement site within an altitude range of 0–100 m. A total of 33000 trajectories were started throughout each 3 h period for which measurements were available at a particular location. They ran for 12 days, and every 15 minutes the location of all trajectories within the lowest 18 km of the model atmosphere was recorded on a grid with the same horizontal resolution as the meteorological fields.

The result of the above calculation is that a map of time-integrated and column-integrated trajectory (or "particle") density exists for each 3 h period. Fig. 2 shows composites of these maps when they are grouped according to the 3 h mean C_2Cl_4 mixing ratio at Tawau (maps for Bukit Atur are very similar, and not shown). These maps indicate that air masses containing the lowest mixing ratios (<0.8 ppt) often passed to the south of the Philippines during transport from the unpolluted subtropical Pacific. As a larger fraction of trajectories travel from midlatitudes, passing through the Philippines, the C_2Cl_4 mixing ratio at Tawau increases. Beyond the Philippines, there are two diverging origins. Air masses containing ∼1.0–1.4 ppt of C_2Cl_4 are largely transported from the Pacific Ocean. In contrast, air masses containing >1.6 ppt of C_2Cl_4 appear to be increasingly affected by an anticyclonic circulation, originating from the Asian land mass and containing higher levels of anthropogenic pollution. This difference is qualitatively consistent with the idea of cold surges leading to elevated C_2Cl_4 in Borneo.

In a further, more quantitative analysis we examine the relationship between the 3 h mean C_2Cl_4 mixing ratios and the fraction of trajectory mass (or equivalently of trajectory residence time) north of 35° N in the corresponding individual air history maps. This fraction increases linearly as the C_2Cl_4 mixing ratio increases (for both sites $r^2 > 0.7$, graph not shown). Other latitude thresholds (between 25 and 45° N) were tested and found to yield similar relationships. This reinforces the argument made above: cold surges are able to move polluted air from northern midlatitudes rapidly (over ∼4 days in the case study we consider in Sect. 4) to equatorial Southeast Asia.

4 Wider air quality implications

Thus far we have focused on C_2Cl_4, an industrial pollutant observed in relatively small quantities. It is clearly of interest to consider how the pollution transport pathway identified in Sect. 3 influences air quality in Southeast Asia more generally. However, the continuous air quality measurements collected in northern Borneo that we are aware of appear to be affected by local sources of pollution. As such they are unlikely to reflect the regional mechanism suggested by the coherent variations in C_2Cl_4 observed at our two measurement sites (Sect. 2) and by our trajectory calculations. In this section we therefore use data available from the MACC reanalysis (Inness et al., 2013, downloaded from http://apps.ecmwf.int/datasets/data/macc_reanalysis/). This data set is created using satellite observations, emission inventories and chemical transport model calculations. For the two pollutants we consider in this section, CO and O_3, 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. We concentrate here on a 2-week period containing a particularly strong surge event (10–23 January 2009, shaded in Fig. 1a), during which there are abrupt changes in our data and the highest C_2Cl_4 mixing ratios occur.

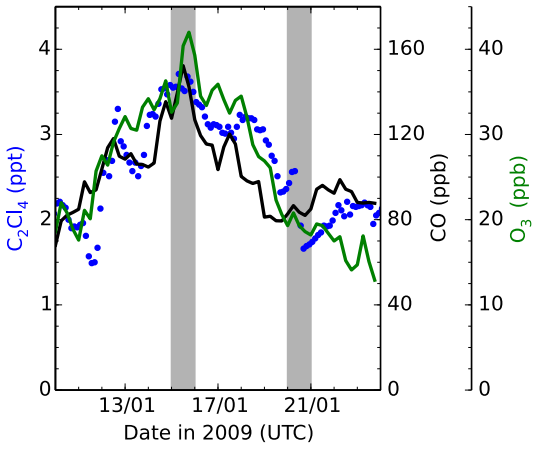

Figure 3. Time series of 3 h mean C_2Cl_4 measurements from Bukit Atur (blue) and 6-hourly CO (black line) and O_3 (green line) at 118° E, 5° N, 925 hPa in the MACC reanalysis. The days analysed in more detail in Fig. 4 are shaded grey.

To begin, we extract time series for CO and O_3 (Fig. 3) in the grid cell nearest to Bukit Atur in the MACC reanalysis. Our C_2Cl_4 measurements from Bukit Atur are also plotted for comparison. We obtained MACC data at 925 hPa (rather than 1000 hPa) because Bukit Atur, at an elevation of 426 m, often appears to be more representative of the free troposphere (Pike et al., 2010). In Fig. 3 the mixing ratios of C_2Cl_4, CO and O_3 all follow the same pattern, with a sustained increase to maximum values on 15 January, followed by a decline back to more typical background mixing ratios in the subsequent days. This suggests that the air masses arriving in Borneo with high levels of C_2Cl_4 also contain a range of other pollutants and significantly impact air quality in this part of the tropics.

Next, in Fig. 4 we contrast daily mean maps of CO and O_3 from the MACC reanalysis (again at 925 hPa) for the 2 days shaded in Fig. 3: 15 January 2009, when concentrations of C_2Cl_4 were relatively high, and 20 January 2009, when

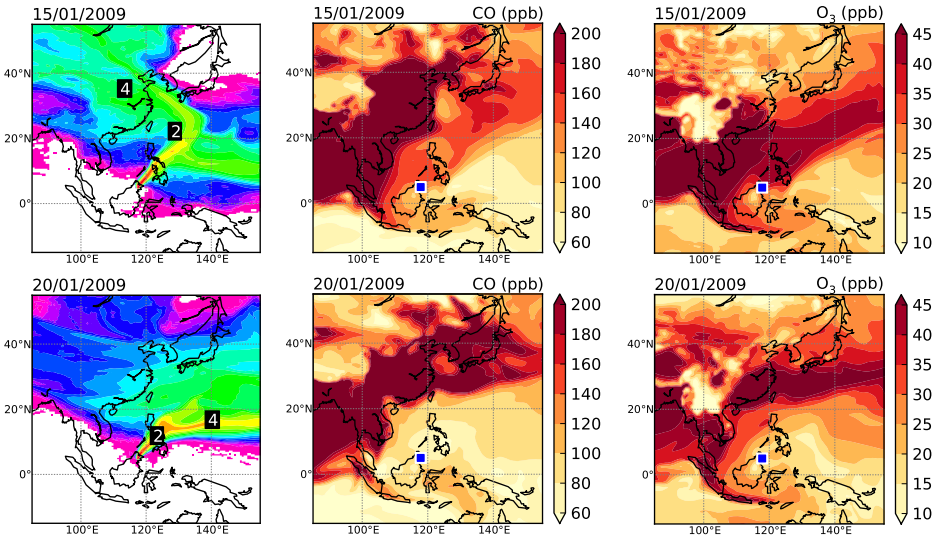

Figure 4. Daily mean air history maps calculated using NAME (left), CO at 925 hPa from the MACC reanalysis (center) and O_3 at 925 hPa from the MACC reanalysis (right) for 15 January 2009 (top) and 20 January 2009 (bottom). The colour scale for the air history maps is the same as in Fig. 2, and the mean horizontal location of the trajectories after 2 and 4 days is marked. In the CO and O_3 panels the location of Bukit Atur is marked with a blue square.

concentrations of C_2Cl_4 were relatively low. 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. All together, the six maps demonstrate that our measurement sites in northern Borneo sit near the edge of a steep pollution gradient associated with contrasting polluted air masses from East Asia and cleaner air masses from the tropical Pacific. On days when air masses are moved rapidly by cold surges from East Asia towards Borneo, such as 15 January (top row of Fig. 4), air quality is significantly reduced. According to the MACC reanalysis, mixing ratios of CO and O_3 can reach, respectively, ∼150 and ∼40 ppb. Our high C_2Cl_4 measurements are excellent markers of this pollution. By contrast, modelled levels of CO and O_3 and measured levels of C_2Cl_4 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).

5 Uplift of polluted air masses

As noted in the Introduction, cold surges are known to affect the characteristics of convection in Southeast Asia (e.g. Chang et al., 2005). A further NAME calculation was performed to assess whether air masses that move rapidly from midlatitudes to the tropics may subsequently ascend. 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). In this case, such transport could lift East Asian pollution to the tropical upper troposphere. Forward trajectories (3000 h^{-1}) were released continuously in a kilometre-deep surface box over East Asia, covering 100–140° E, 30–50° N. These trajectories were not subject to any chemical losses and their travel was recorded for 12 days. The aim was not to simulate any particular pollutant but to consider in a simple manner the transport of air masses originating in the polluted East Asian midlatitudes.

To assess whether the midlatitude air masses are lifted in the tropics, the shading in Fig. 5a shows the density of particles (i.e. trajectories), between 600 and 400 hPa (i.e. the mid-troposphere) and between 0 and 10° N during January 2009 for Southeast Asian longitudes. Also plotted are CO contours from the MACC reanalysis at 500 hPa in the same time–longitude space. Within the figure there are periods, notably around 12–13 January near the time of a strong cold surge (Figs. 3 and 4), when peaks in simulated midlatitude air masses coincide with enhanced CO mixing ratios. Similar regions of agreement are found at the same time in a corresponding plot for ∼200 hPa (Fig. 5b). These features indicate that East Asian midlatitude pollution is capable of influencing atmospheric composition through much of the depth of the Southeast Asian tropical troposphere.

Our argument is supported further by Fig. 6a, b, which show, respectively, longitude–altitude and latitude–altitude slices through the Southeast Asian atmosphere on 15 January 2009 (our selected cold surge day in Fig. 4). Again, the density of the midlatitude tracer is indicated with shading. The plots demonstrate that the air masses originating in midlatitudes can be lifted above 200 hPa within the tropics. Evidence for this type of vertical transport is weaker in similar

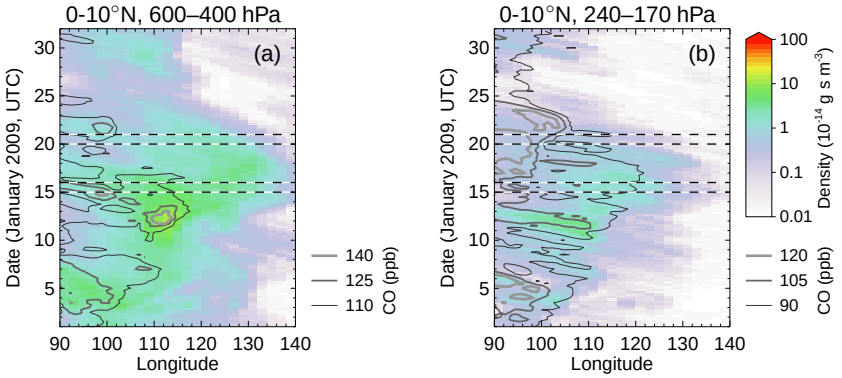

Figure 5. Time-integrated particle (i.e. trajectory) density, as a function of longitude and time through January 2009, resulting from a midlatitude particle source in NAME. **(a)** shows particle density between 600 and 400 hPa averaged between 0 and 10° N. **(b)** shows particle density between 240 and 170 hPa, also averaged between 0 and 10° N. The contours show CO from the MACC reanalysis at, respectively, 500 and 200 hPa, for the same spatial and temporal dimensions.

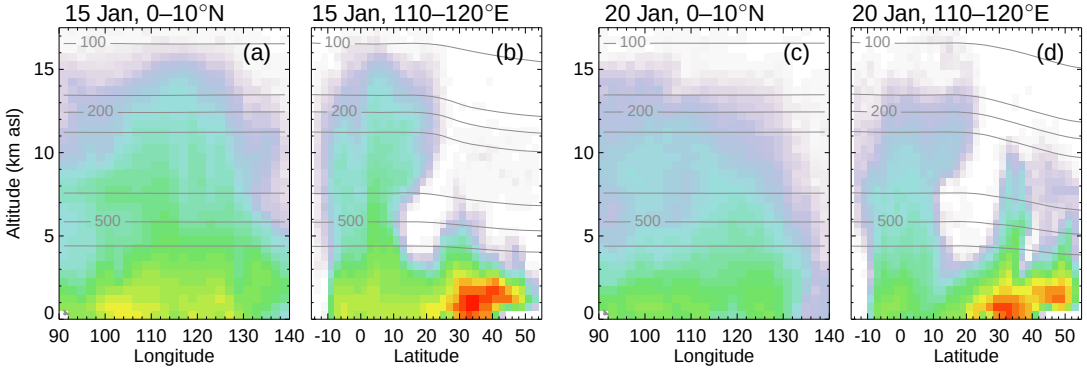

Figure 6. Time-integrated particle (i.e. trajectory) density resulting from a midlatitude particle source in NAME, on 15 and 20 January 2009 (marked with dashed lines in Fig. 5). The colour scale is the same as in Fig. 5. **(a)** shows a longitude–altitude cross section for 15 January 2009, averaged over 0–10° N. **(b)** shows a latitude–altitude cross section, also for 15 January 2009, and averaged over 110–120° E. **(c)** and **(d)** show the same for 20 January 2009. The altitude scale is kilometres above sea level (a.s.l.). Pressure contours (in hPa) from NAME's driving meteorological data are marked with grey lines (contours without labels bound the pressure ranges used in Fig. 5).

plots for days without cold surge activity (20 January, for example, Fig. 6c and d). An additional analysis of the trajectory timescales shows that it can take fewer than 10 days for air masses to travel from the East Asian boundary layer to the tropical upper troposphere (i.e. above 200 hPa). This is a sufficiently short time for this process to be important for even relatively short-lived pollutants.

6 Summary and discussion

We analyse observations of C_2Cl_4, an excellent tracer for anthropogenic pollution, collected at two sites in Borneo during the NH winter of 2008/2009. Backward trajectories calculated using NAME show that measurements of high C_2Cl_4 mixing ratios were associated with rapid cold surge transport towards the Equator from polluted midlatitude East Asia. Data available from the MACC reanalysis demonstrate that the polluted air masses may contain levels of CO and O_3 that are approximately double the typical background levels in Borneo. Steep gradients in atmospheric composition have been identified before in this region (Hamilton et al., 2008), but the gradients identified here appear to be mobile (Fig. 4) and capable of periodically reducing air quality significantly in Southeast Asia. Once in the deep tropics, we show that the polluted air masses can be lifted out of the boundary layer into the mid- and upper troposphere. This process may be enhanced by the more vigourous regional convection typically induced by the cold surge circulation, as considered before within the framework of Borneo Vortex events (Braesicke et al., 2012). 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. The tropical upper troposphere is a key gateway to the lower stratosphere (e.g. Fueglistaler et al., 2009). The route we have identified may therefore transport polluted air from NH midlatitudes into the tropical upper tro-

posphere and thence to the lower stratosphere during NH winter. To illustrate the potential importance of this route, air masses enriched in C_2Cl_4 may also contain high levels of various other relatively short-lived chlorine-containing gases (e.g. Wang et al., 2014) that could have a negative impact on stratospheric ozone. The significance of this transport pathway therefore needs to be investigated further.

Pollutant emissions from East Asia have increased over recent decades, and they are unlikely to decline significantly in the next 20 years or so (e.g. Zhao et al., 2014). At the same time, rising greenhouse gas levels could lead to changes in the regional climate. There remains, however, significant uncertainty in how the various large-scale processes that influence East and Southeast Asian climate variability will change this century (Christensen et al., 2013). Any climatic changes are likely to influence the importance of the pollution transport pathway we have identified. To illustrate, the El Niño–Southern Oscillation is known to moderate the strength of cold surge activity (e.g. Zhang et al., 1997). In order to better quantify the future impact of East Asian pollution on the tropical atmosphere, there is a need to increase our understanding of both past multi-decadal variations (e.g. Huang et al., 2011) and possible future changes (e.g. Park et al., 2011) in cold surge frequency.

Observational evidence from other parts of Southeast Asia will be needed to assess more fully the influence these transport events have on regional atmospheric composition. One step in this direction is afforded by a new programme of long-term measurements of a suite of compounds (including the halocarbons measured by μ-Dirac) at the University of Malaya's Bachok Marine Research Station on the east coast of Peninsular Malaysia (http://www.ioes.um.edu.my/research_facilities.html). Levels of pollution associated with the prevailing northeasterly flow through the South China Sea may be more severe in, for example, Peninsular Malaysia than those in Borneo. There are clearly possible implications for human health, though the effect of pollutants such as O_3 on tropical forests and crops is yet to be well understood (e.g. Ainsworth et al., 2012). Further long-term measurements will also facilitate more detailed investigation of the influences of the climatic variations discussed above.

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