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Rapid transport of East Asian pollution to the deep tropics

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Abstract

Anthropogenic emissions from East Asia have increased over recent decades, and under the prevailing westerly winds, these increases have led to changes in atmospheric composition as far afield as North America. 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 during the NH winter of 2008/09. We use the NAME trajectory model 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 across > 30° of latitude in 4 days. We then present data from the Monitoring Atmospheric Composition and Climate reanalysis which suggests 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. Convection in Southeast Asia can be enhanced by cold surges, 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 mid-latitude 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 ozone (O_3) precursors (e.g. Granier et al., 2011). The emitted pollutants are known to be transported eastward by the prevailing mid-latitude 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, in both western North America (Cooper et al., 2010) and Hawaii (Lin et al., 2014).

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Southward transport of polluted air masses originating in East Asia, associated with the Northeast Monsoon which occurs during Northern Hemisphere (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 result in a strengthening of the northeasterly monsoon winds in the South China Sea (Zhang et al., 1997; Chan and Li, 2004; Chang et al., 2004). While these surges are known to consistently enhance and affect the distribution of convection in Southeast Asia (e.g. Chang and Lau, 1980; Slingo, 1998; Compo et al., 1999; Chang et al., 2005), 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. This process may be especially important in the context of our study because Southeast Asian air masses are unusually likely to be lifted towards the stratosphere during NH winter (e.g. Levine et al., 2007; Aschmann et al., 2009). 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).

Our analysis is based on observations of an anthropogenic halocarbon, perchloroethene (C_2Cl_4), collected in Borneo during the winter of 2008/09 (Sect. 2). We conduct simulations with a trajectory 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 trajectory 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).

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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, with oxidation by OH the dominant loss mechanism (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).

To date few continuous measurements of atmospheric composition have been reported in Southeast Asia. To fill 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 the NH winter of 2008/09 and have been averaged over 3 h periods (Fig. 1a). There are clear week-scale variations in the data, which occur concurrently at the two sites ($r^2 = 0.85$ in Fig. 1b), and are characterised by a number of abrupt changes between "background" mixing ratios, of ∼ 1.0–1.5 ppt, and "polluted" mixing ratios, of ∼ 2.0–3.0 ppt. We will show in Sect. 3 that changes

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in Northeast Monsoon circulation are the primary cause of these transitions. We have observed similar features in measurements collected in Borneo during subsequent winters and are therefore confident these changes are typical of the region.

Figure 1 shows 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) but Robinson et al. (2014) do discuss the possibility of calibration uncertainty leading to part of this difference. In this study we focus on the variations observed at both locations, which Fig. 1b shows are well correlated, and hence the absolute mixing ratios are of lesser importance.

3 Transport pathways

We use the UK Met Office's Lagrangian atmospheric dispersion model, NAME (Jones et al., 2007), to interpret our C_2Cl_4 measurements. Trajectories are calculated using three-dimensional meteorological fields produced by the UK Met Office's Unified Model (UM) during operational weather forecasts. These fields have a horizontal resolution of 0.5625° longitude by 0.375° latitude and are available at 3 hourly intervals. A parameterisation of turbulence is employed (Webster et al., 2003; Morrison and Webster, 2005). Initially we use NAME to calculate batches of backward trajectories started at each measurement site within an altitude range of 0–100 m. 33 000 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 min 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. Figure 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

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the south of the Philippines during transport from the unpolluted subtropical Pacific. As the C_2Cl_4 mixing ratio at Tawau increases, a larger fraction of trajectories travel from mid-latitudes, and pass through the Philippines. Thereafter, there are two diverging origins. Air masses containing ~ 1.0–1.5 ppt of C_2Cl_4 are largely transported from the Pacific Ocean. In contrast, air masses containing > 1.5 ppt of C_2Cl_4 appear to be affected by an anti-cyclonic 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 does (for both sites $r^2 > 0.7$). Other latitude thresholds (between 25–45° 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 mid-latitudes rapidly to equatorial Southeast Asia.

4 Wider air quality implications

Thus far we have focussed on C_2Cl_4, an industrial pollutant observed in relatively small quantities. It is clearly of interest to consider how this pollution transport pathway influences air quality in Southeast Asia more generally. We are not aware of any continuous air quality measurements in Northern Borneo that are unaffected by local pollution, so instead make use of data available from the MACC Reanalysis (Inness et al., 2013, downloaded from http://apps.ecmwf.int/datasets/data/macc_reanalysis/). We concentrate here on a two 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.

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

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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 the daily mean maps of CO and O_3 from the MACC reanalysis, again at 925 hPa, on 15 and 20 January 2009 (the two days shaded in Fig. 3, with relatively high and low concentrations, respectively). Corresponding daily mean air history maps, in which transport timescales are marked to highlight the strength of the cold surge events, are also presented. Together, the 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, $\sim$ 150 ppb and $\sim$ 40 ppb and our high C_2Cl_4 measurements are an excellent marker of this pollution. In contrast, on days when winds blow from the Pacific, such as 20 January (bottom row of Fig. 4), "background" levels of modelled CO and O_3 and measured C_2Cl_4, of approximately half the polluted values, are present in Borneo.

5 Uplift of polluted air masses

Cold surges are known to affect the intensity 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 mid-latitudes to the tropics may subsequently as-

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cend. Previous work suggests that NAME is a useful tool for analysing this type of vertical transport in the tropics (Ashfold et al., 2012). In this case, such transport could introduce East Asian pollution to the tropical upper troposphere. Forward trajectories (3000 h^{-1}) were released continuously in a kilometre deep surface box covering 100–140° E, 30–50° N. 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 mid-latitudes.

To assess whether the mid-latitude air masses are lifted in the tropics, the shading in Fig. 5a shows the density of particles (i.e. trajectories), between 600–400 hPa (i.e. the mid-troposphere) and between 0–10° 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 mid-latitude air masses coincide with enhanced CO mixing ratios. Similar regions of agreement are found in a corresponding plot for 200 hPa (not shown). These features indicate that mid-latitude pollution is capable of influencing atmospheric composition through much of the depth of the tropical troposphere.

Our argument is supported further by Fig. 5b and c, 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 mid-latitude tracer is indicated with shading. The plots demonstrate that the air masses originating in mid-latitudes can be lifted above 200 hPa within the tropics. Evidence for this type of vertical transport is weaker in similar plots for days without cold surge activity (20 January, for example, not shown). 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.

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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 2008/09. 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 mid-latitude 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 in to 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). 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 mid-latitudes into the tropical upper troposphere and thence to the lower stratosphere during NH winter. Its importance 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 quantify better the future impact of East Asian pollution

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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 program 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 allow the influence of climatic variations identified above to begin to be investigated.

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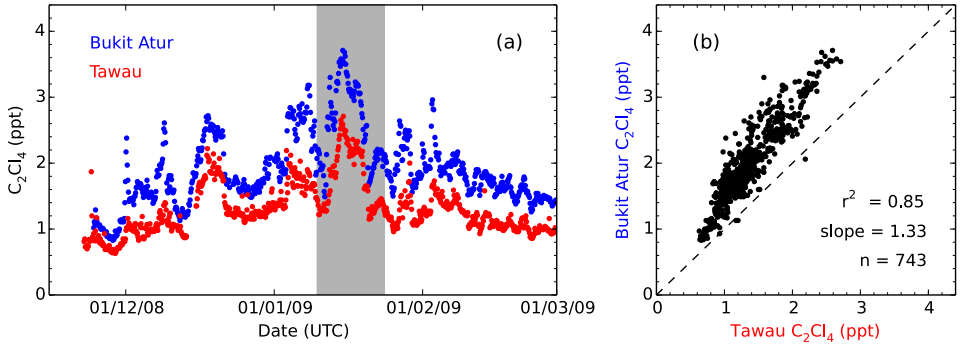

Figure 1. (a) shows a time series of 3 h mean C_2Cl_4 measurements collected at Bukit Atur (blue) and Tawau (red) during the winter of 2008/09. The period analysed in more detail in Sect. 4 is shaded grey. **(b)** shows the correlation between the measurements at the two sites. In constructing this figure 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.

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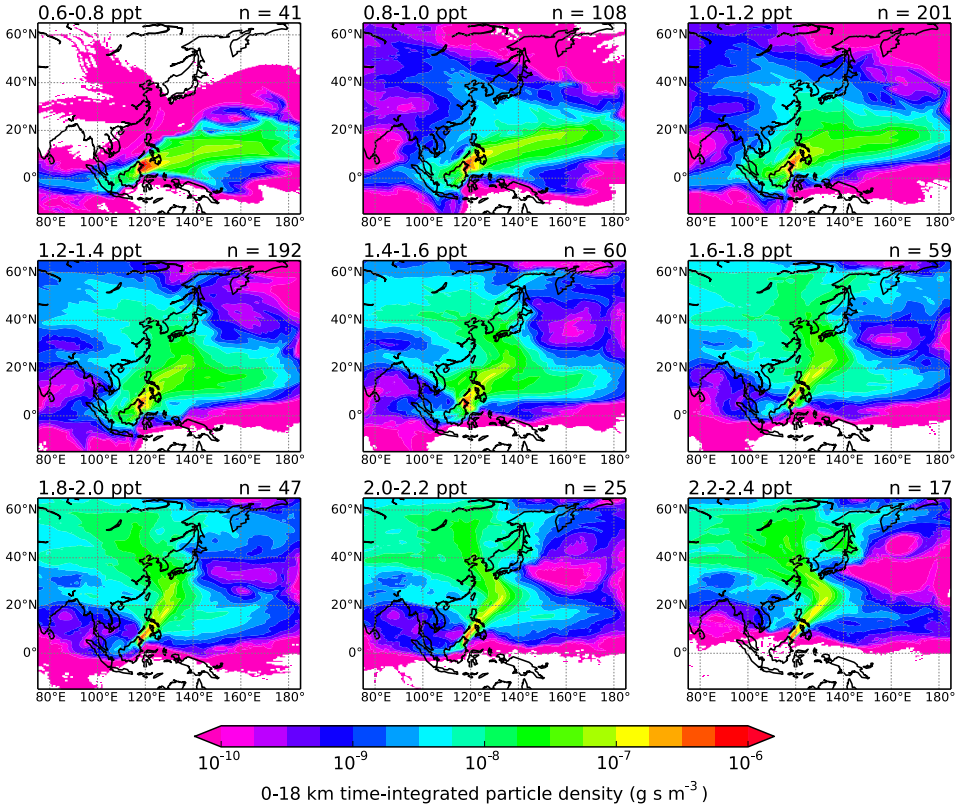

Figure 2. Composite air history maps for each C_2Cl_4 mixing ratio interval of 0.2 ppt at Tawau. The number of 3 h periods, *n*, contributing to each composite is noted above each panel. A mixing ratio greater than 2.4 ppt was observed in eleven 3 h windows; these periods are not accounted for in the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

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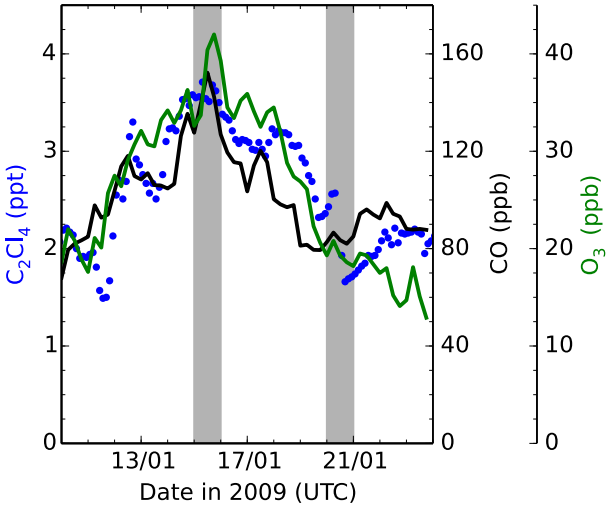

Figure 3. Time series of 3 h mean C_2Cl_4 measurements from Bukit Atur (blue) and 6 hourly CO (black line) and O_3 (green line) at 118° E, 5° N, 925 hPa in the MACC reanalysis. The days analysed in more detail in Fig. 4 are shaded grey.

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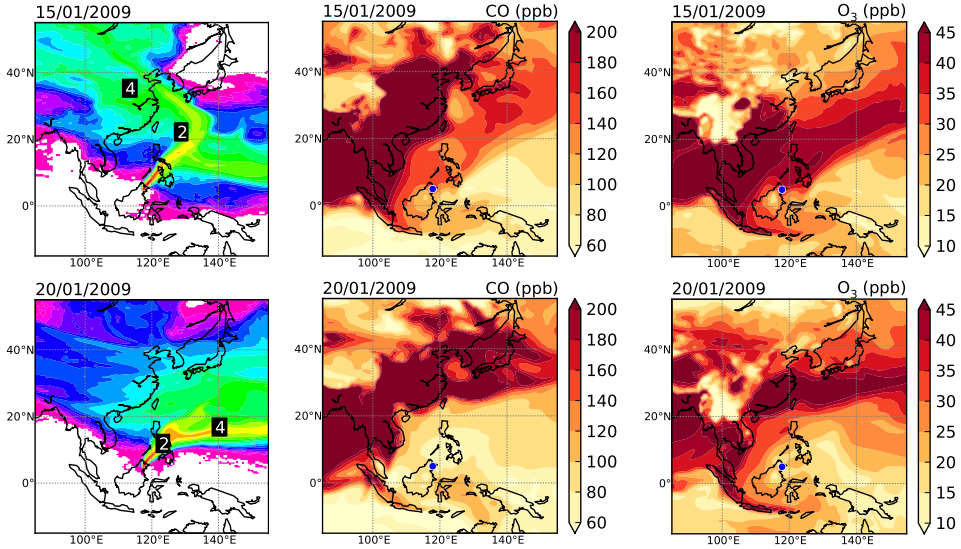

Figure 4. For both the 15 and 20 January 2009 (shaded in Fig. 3), daily mean air history maps calculated by NAME, and daily mean CO and O_3 at 925 hPa from the MACC reanalysis. The colour scale for the air history maps is the same as in Fig. 2, and the mean horizontal location of the trajectories after 2 and 4 days is marked. In the CO and O_3 panels the location of Bukit Atur is marked with a blue circle.

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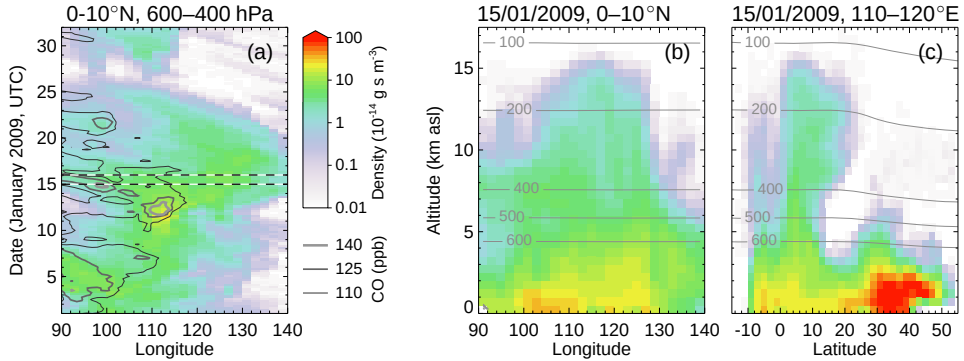

Figure 5. Various views of time-integrated particle (i.e. trajectory) density resulting from a mid-latitude particle source in NAME. **(a)** shows particle density between 600–400 hPa, averaged between 0–10° N, as a function of longitude and time through January 2009. The contours show CO from the MACC reanalysis at 500 hPa for the same spatial and temporal dimensions. **(b)** shows a longitude–altitude slice of particle density for 15 January 2009 (marked with dashed lines in plot **a**), also averaged over 0–10° N. **(c)** shows a latitude–altitude slice of particle density, also for 15 January 2009, and averaged over 110–120° E. Pressure contours (in hPa) from NAME's driving meteorological data are marked with grey lines in both **(b)** and **(c)**.

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