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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The lofting of Western Pacific regional aerosol by island thermodynamics as observed around Borneo" *by* N. H. Robinson et al.

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We thank the reviewer for their comments and suggestions for improving our manuscript. We address each of their points below.

1) Starting from the end when circulation over the island of Borneo is combined with aerosol data (Fig. 14). I think there is a certain misinterpretation. Mixed layer (ML) in tropics is cloud free and the fact that it is well mixed is due to dry convection up to cloud base, which is often on top or slightly above top of the ML. The cloud convective layer (CCL) above connects ML with lower troposphere. Not like authors state on page 1235, line 5: "Layer One represents a boundary layer well mixed by turbulence and

shallow convection". There is a clear difference in tropics between marine boundary layer (MBL) depth and diurnal cycle compared the ML depth and diurnal cycle over land. MBL height does not change a lot during day and night, however over the land ML varies from couple of hundred meters during night up to one km or more during midday and afternoon. Over the ocean shallow convection seldom shows similar activity compared to the shallow convection over land (thanks to extra latent heat from evapotranspiration and in this case also Borneo orography). Shallow convection over land usually reaches altitudes similar to authors Layer Two and Layer Three border. The peaks in aersol number concentrations on several figures very likely indicate typical cloud top in the region of profiles and it will be nice if authors can add to the vertical profiles also water vapour mixing ratio or humidity. Where I am heading with this long paragraph is following:

During daytime or when shallow convection is active, ML and CCL (up to top of Level Two) are very closely coupled through clouds. This is nicely shown in this study using changes in aerosol composition. Presented data are as far as I understand from cloud free environment, so it is biased to observations in region of in-between cloud subsidence. It will be nice if authors will use this very nice opportunity and include in discussion paragraph or two on concept of layers using meteorology and thermodynamics compared to layers defined by chemical properties. In the other words, can one say that chemically boundary layer in tropics extends to the top of the shallow convection?

The reviewer's statement of the physics of variation of both MBL and PBL depth as a result of diurnality, surface heating and orographic forcing effects is valid, as is the potential difference between the ML and the height of the cloud convective layer. However, in this paper, we do not seek to directly quantify the intraday variation in boundary layer dynamics and instead seek to discuss composition within the instantaneous mixed layer as diagnosed from the aircraft measurements. When we refer to the mixed layer (ML) in our manuscript, we define this to mean a layer within the planetary or

marine boundary layer that is also below the height of any cloud. For the purposes of our analysis when sub-sampling the aircraft dataset, the accuracy of this definition has been confirmed from the analysis of aircraft thermodynamic profiles (see discussion) and by ensuring that sub-sampled data is below the altitude of cloud base as diagnosed by on-board cloud microphysics measurements.

The reviewer also suggests that by presenting cloud-free data we may be subject to biased sampling in-between cloud subsidence. We can confirm that, for the case studies presented, cloud coverage was very low (less than 10% over Borneo and surrounding waters by analysis of MTSAT infrared satellite imagery) and that we would therefore not expect significant bias of recently subsided air in the sampled data. In summary, our measurements represent a mixed layer that is representative of the column below the height of cloud-base. Therefore, in response to the reviewer's question about the nature of the chemical boundary layer, we cannot justifiably assert (from these measurements alone) that the "chemical boundary layer" extends to the top of the shallow convection. Therefore, to avoid confusion, we now state this when we introduce our definition of the ML (page 1228, line 20).

Also, the reviewer asks that vertical profiles of water vapour mixing ratio are shown - note that these are included in tephigrams included in the existing appendix to the manuscript.

2) At several occasions authors discuss new particle formation, but with the instrumental set up, which covers total aerosol concentration 3 nm (CPC) and then aerosol size distribution from 0.1 to 3 um (PCASP) they cannot show, where new particle formation happened. The difference between both instruments can also involve Aitken mode aerosol, which can be many hours or even days old. I do think that authors are correct with their interpretation, but it should be clearly stated that the interpretation is likely right guess based on previous experiments and published work.

We agree with the point about the limitation of the CPC measurement and will change

the manuscript to reflect this. Specifically:

page 1236, line 12

> The daily increase of aerosol number concentrations is consistent with the formation of new particles and the increasing number concentration with altitude is consistent with the formation of new particles from partitioning of semi-volatile vapours as they are lofted to higher into the atmosphere where it is colder. It should be noted that these particles may have been nucleated at some point upwind before growing to be detectable at the lower size limit of the CPC (3 nm). However, the vigorous uplift observed provides a plausible mechanism for local particle nucleation.

and page 1240, line 16-18

> Aerosol number concentration profiles are consistent with new particle formation aloft throughout the day, which could be due to an increase in both photochemical ageing, and the cooling of semi-volatile organics due to dynamic uplift.

3) In experimental part of the manuscript is mentioned that SLR runs were performed. What is the variability on horizontal scale and how does it compare to vertical variability in different layers?

	All	Upwind
Org Prof	0.58	0.24
Org SLR	0.16	0.13
SO4 Prof	0.41	0.30
SO4 SLR	0.08	0.09

The above table shows the mean of the standard deviations of each profile/SLR. The mean of the standard deviations of all the vertical profiles is more than that of the

SLRs. This supports the assertion made in the manuscript that the different average values reported are due to the vertical structure of the atmosphere, not horizontal inhomogeneity. The greater standard deviation of the organic than the sulphate data is consistent with the contribution of local organic aerosol sources.

Is there a difference between measurements upwind over the ocean and over the Borneo?

The upwind data also show greater mean standard deviations for profiles than for SLRs. There seems to be generally lower variability upwind, however, given the differences in size of the upwind vs over-island datasets, we do not think any conclusions should be drawn from this.

4) Discussing changes in aerosol composition, authors often argue with influence of cloud processing and wet removal. There is very likely extensive observational log for each flight maybe including also if there have been in vicinity precipitating and non-precipitating clouds. Can this be used in the analysis?

We have made some informal discussion of cloud observations within the manuscript. In general there was a layer of shallow cumulus clouds approximately at the boundary between Layers One and Two (Fig. 4; p 1230, line 12;). It has been stated when this was not the case, that is, the two profiles where there was also cumulonimbus present. We are loathed to draw too much upon precipitation observations, largely because of the highly changeable and localised weather (also see next answer).

Minor comments: Page 1222, Data handling part: With respect to available data, why potential and/or equivalent potential temperature was not used to define atmospheric layers prior involving aerosol in their definition? Both are very good tools to do it and easy to include them in vertical profiles figures. Using aerosols as one of the key parameters to define layers and then discuss role of meteorology using these layers involves a danger of circular reasoning and bias thanks to assumptions how the profile and layers should look like based on aerosol.

We appreciate the point about the potential risks of defining layers using aerosol composition data. However, we feel we minimised this risk by drawing boundaries using thermodynamic data in the form of tephigrams and wind vector profiles, as well as the aerosol data. We feel that it was best to use several complementary metrics. As stated on p1222, lines 16-24, the inhomogeneity of the local atmosphere, with aerosol potentially being introduced/removed locally upwind of the measurements, means that thermodynamic data alone would not suffice to characterise these atmospheric layers.

With respect to the comment on the use of potential/equivalent potential temperature, while we accept that this is an effective alternative way of presenting thermodynamic profiles, we chose to represent this using tephigrams given the additional information provided on moist/dry mixing.

Page 1227, line 6: instead of "sub-degree" should be "sub-grid"?

We thank the reviewer for bringing this to our attention and will amend the manuscript accordingly.

*Page 1229, "upwind" and Fig. 6: Several thousand pp*cm-3 is very high aerosol number concentration for MBL. Typically remote tropical MBL contains around 300 – 600 pp*cm-3. Where the air came from? Is sulphate really resulting from DMS oxidation as discussed later in the paper or it can be long range transport of polluted air?*

It is certainly possible that the air upwind of Borneo is influenced by pollution. The relative contributions of different regional sources is discussed in depth in Robinson et al. 2011. In that analysis back trajectories did not show any flow over major pollution centres during the preceding week. In addition, while sulphate concentrations were found to be greatest in marine air masses, long range pollution tracers (CO, black carbon) were found to be greater in terrestrial air masses. The conclusion was that it is unclear what the regional off-island source of sulphate is, but that one might expect DMS oxidation to be a significant source. However, we accept the argument made about the relatively high upwind sulphate number concentrations. We will change p1229, line

20-22, where we do not discuss the regional source of sulphate, to read:

> Number concentrations are low throughout the profiles compared to those measured downwind, which is consistent with the aerosol population being dominated by aged long range transport aerosol.

and we will change the mention of DMS on p1234, lines 25-30 to

> [Robinson et al.] conclude that the sulphate is potentially from a variety of sources, with one likely source being the production of dimethyl sulphide (DMS) from processing of phytoplankton emissions.

Page 1234, lines 17-18: Cumulonimbus or cumulus formation? Data shown extend up to approximately 5 km altitude. Typical height of Cb clouds in tropics is 10+ km. Then the question is of the described effects on aerosol profiles is due to lateral ventilation of the CB clouds or due to more intensive shallow convection in vicinity of Cb clouds, which is often the case.

The profiles presented in Fig. 9 were recorded in the vicinity of cumulonimbus clouds which were observed visually and recorded in the flight log. It is certainly possible that the aerosol could be uplifted as the Cb cloud was building, however, we felt this level of mechanistic detail would be overly speculative given the measurements made. We have left the paper as stating that aerosol may be uplifted in the vigorous convection associated with these types of clouds, without stating if we think it is from lateral ventilation or intensive shallow convection.

Page 1241, lines 15-16: Can Doppler lidar data can be used to show that during MCS regime the shallow convection, which is likely controlling the exchange between ML and CCL is less active compared to ITC-regime?

Unfortunately the Doppler lidar stationed at the Bukit Atur ground site was not on-line when the aircraft measurements were performed (p 1236, line 16).

Robinson, N. H., Newton, H. M., Allan, J. D., Irwin, M., Hamilton, J. F., Flynn, M., Bower, K. N., et al. (2011). Source attribution of Bornean air masses by back trajectory analysis during the OP3 project. Atmospheric Chemistry and Physics, 11(18), 9605-9630. doi:10.5194/acp-11-9605-2011

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