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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Scavenging of biomass burning refractory black carbon and ice nuclei in a Western Pacific extratropical storm" *by* J. L. Stith et al.

**Anonymous Referee #2**

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Review of Stith, J. L., Twohy, C. H., DeMott, P. J., Baumgardner, D., Campos, T., Gao, R.-S., and Anderson, J.: Scavenging of biomass burning refractory black carbon and ice nuclei in a Western Pacific extratropical storm, Atmos. Chem. Phys. Discuss., 11, 567-595, doi:10.5194/acpd-11-567-2011, 2011.

General Comments:

The manuscript is a case analysis of aerosol scavenging within a large upper tropospheric storm system over the Pacific. The main conclusions of the manuscript are



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that refractory black carbon and organic-rich aerosol emitted from biomass burning are scavenged by cirrus crystals. In a previous work (Baumgardner et al. 2008), many of the same authors used the same PACDEX dataset as a whole to conclude a similar message, that black carbon aerosol are scavenged by UT cirrus crystals. The additional information provided by this manuscript is data from a CFDC ice nucleation instrument and the comparison of CVI residual concentrations with ambient ice crystal concentrations. The current case study delves more deeply in to the microphysics of one cloud system than the previous overview, and the analysis here is much more robust. Still, it is questionable whether the current manuscript presents a scientifically relevant case that was not covered by the 2008 manuscript. The additional scientific conclusion here is that not only black carbon aerosol but also organic-rich biomass burning aerosol are scavenged.

Scientifically, the distinction between nucleation and inertial scavenging is a very important one in order to constrain cloud formation processes. The previous Baumgardner paper concluded that inertial scavenging was the source of the enhanced BC concentrations in cloud, but the authors did not address the possibility of BC acting as efficient ice nuclei. To address that possibility, one needs to compare the BC number concentrations from cloud residuals with ice crystal concentrations, which the authors present in the current manuscript. Also by measuring ice nuclei from cloud residuals with the CFDC instrument, one can theoretically remove the influence of inertial scavenging. The approach that the authors have taken here is a powerful one.

However, these appear to be very challenging measurements, and the manuscript presents few details in this regard. The authors compare number concentrations of sampled cloud 'residuals' counted behind the CVI to ambient cloud particle concentrations measured using a cloud particle probe. The paper's major conclusions rest on accurate concentrations measured downstream of the CVI and using the cloud probes. These comparisons seem tenuous because of the uncertainties in both concentrations. Furthermore, the data in the manuscript suggests the possibility of contamination or ar-



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tifacts affecting concentrations measured in ice clouds.

Overall, the conclusions are not adequately supported by the analysis in a number of areas, as detailed below. As a result, the manuscript is not publishable in its current form. The authors should consider addressing the major points and resubmitting the manuscript. To conclude scientific relevance beyond the previous Baumgardner paper, the authors must justify that the uncertainties in measured concentrations are small such that the concentration comparisons are relevant, address potential artifacts in the dataset, and provide a more robust identification of biomass burning particles.

Major points:

1) The authors mention that the good agreement between the number concentrations of ice residuals and ice crystals (Fig 5, 575-20) is noteworthy, and much of the paper's conclusions rely on this agreement. This agreement masks the complexity of both the residual and ice crystal measurements. How uncertain are these concentrations, particularly in light of the large enhancement factors used to adjust the raw data? The authors report that the CVI inlet enhances cloud particle concentrations by a factor of "approximately 25" (or x30-50 from the previous publication). Presumably, the authors divide their residual number concentrations by this factor to derive cloud particle concentrations. This large factor is derived from an idealized flow field. These concentrations are then compared to CDP and 2DC cloud particle concentrations. Please explain how this is a rigorous correction factor and how the uncertainties in this factor for realistic flow conditions propagates to the concentration comparisons in the manuscript. Provide a reference to this inlet design if available.

How is the lower threshold size ($\sim$5 um) treated in the CVI transmission? The D50 for any aerosol impactor is not a step function, and the transmission curve can extend to sizes 100% beyond D50 in both directions (eg, Noone et al., AST 1998). How do the uncertainties in the CVI's transmission curve translate into the reported concentrations? This uncertainty may be small in this case since it appears that small ice crystals



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have low relative abundance(?), but please state.

For the ice crystal concentrations, do the authors expect any significant contribution from ice crystal shattering on either the 2DC probe or upstream aircraft surfaces? The images in Fig 6 indicate ice crystals many 100's of microns in size, and shattering on upstream surfaces increases with particle size. The authors address crystal shatter within the CVI inlet (p577), but do not address this important potential artifact. The authors did not report size distributions for the clouds, but the cloud crystal sizes in this altitude range extended to many 100s of um (Fig 6). The larger particles are probably susceptible to shattering (Korolev and Isaac, 2005; Viduarre and Hallet, 2008). How might this affect the ice crystal concentration measurements? Lastly, what fraction of the ice crystal size distribution is measured by the 2DC? A complete size distribution using all available cloud instruments should be presented.

2) The authors seem to rely on processes that are neither well understood nor characterized – ice particle shattering within an inlet to release either scavenged particles or multiple soluble IN species (577, 11-29) – as principal support for their conclusion that ice crystals scavenged biomass burning and black carbon aerosol. The authors state that large crystals will impact on downstream inlet bends and shatter (577,9). What fraction of the crystal size distribution can the CVI sample without shattering? One crystal shattering event can produce 100's of fragments (refs above). This does not appear to be a well-controlled sampling strategy. As a result the authors are forced to make several assumptions regarding fragment and residual behavior during this process (577) that drive the manuscript's conclusions. In one instance, the authors conclude that a the CFDC operating at colder temperatures than ambient gives higher IN concentrations than ambient ice crystal concentrations due to ice crystal shattering upon sampling (577, 25). Is it not a simpler explanation that at colder temperatures, more IN are active?

There is some evidence that ice crystals can generate secondary particles within inlets by ablating the inlet surface (e.g., Murphy et al., 2004, Kojima et al., 2004). This



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can artificially increase particle concentrations within the inlet by large factors. These secondary particles can then be incorrectly counted as residuals by aerosol counting instruments, but artifacts are sometimes resolved by compositional analysis. In the current study were the residuals analyzed by electron microscopy investigated for inlet material such as titanium or aircraft material such as aluminum or zinc? What fraction of chemically analyzed particles showed no evidence of such indicator metals?

The authors contend that scavenging is occurring based on the fact that the SP2 reports BC number concentrations that are factors of ∼2-5 higher than IN or cloud particle concentrations (Fig 7). However, total residual particle concentrations (including non-BC and particles larger than 190nm) are ∼100 times larger than the IN. Do the authors suggest that each ice crystal has scavenged 100 aerosol particles, or is this evidence of a sampling artifact? The authors have estimated size-dependent scavenging rates, but they do not report the results. Do the scavenging rates support this number? Since aerosol scavenging is the principal theme of the manuscript, a separate section should be devoted to a scavenging calculation. Please explain the 100-fold aerosol/IN result. These concentration comparisons provide the main support for the paper's conclusions, since apparently BC was not detected in EM analysis (580, 10), and so provided no confirmation of BC residuals (although 'structureless char' was detected by EM (580, 8) ? State clearly.).

The vertical profile in Fig 8a shows that BC concentrations (measured behind the CVI? please state.) increase dramatically when the cloud transitions from liquid to ice. The authors mention this phenomenon but do not suggest an explanation. Gas phase O3 and CO tracers are constant through this transition, and the airmasses appear similar (578, 12). One possible explanation that should be addressed or refuted is that the particles reported as BC within ice cloud are artifacts produced from shattering of ice crystals. Ice crystals are substantially more massive than cloud droplets and are therefore more likely to ablate inlet and aircraft surfaces, generating metallic aerosols. Is the response of the SP2 to possible artifact material such as titanium dioxide and aircraft



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material known? Some SP2 investigators do not report data within ice clouds due to observed artifacts (eg, Schwarz et al, 2008). State evidence for lack of artifacts from in-cloud sampling of ice crystal fragments. For example, does the SP2 incandescence signal within cloud differ from clear air sampling? Can the authors demonstrate a contrasting case within a cloud containing similar size ice crystals where residuals did not show a large BC increase?

3) The authors have not adequately demonstrated that the collected cloud residuals originated from a biomass burning source. The determination of the characterized aerosols as biomass burning in origin is critical to the principal conclusion of the paper. However, the authors do not use unique biomass burning tracers such as acetonitrile or HCN (presumably not available from the dataset), and instead rely on elevated rBC, organic-rich aerosol, and the presence of potassium (please provide references for this tracer). However, the EM analysis also indicated calcium and magnesium (relative abundance not reported), which are common indicators of crustal material and sea salt (along with sodium and chloride, which were also observed). The ion tracer is unclear, and the other tracers are not unique to biomass burning. Although the combination presents circumstantial evidence that is suggestive of biomass burning origin, airmass back-trajectories intersected large Asian cities, remote continental regions, and dust sources regions, all of which could contribute to the observed chemical tracers.

In order to differentiate a biomass burning source from e.g., urban, dust, and marine sources, and without the availability of typical gas phase indicators, the authors could compare EM compositional elements of aerosol collected during an unambiguous intercept of an out-of-cloud biomass burning plume (high aerosol loading, visible smoke, very high CO, rBC). Because an out-of-cloud sampling is simpler and less artifact-prone than the cirrus sampling case presented here, compositional agreement with an unambiguous out-of-cloud biomass burning case is needed to justify that the cloud residuals are from a biomass burning source. Do those samples show similar abundance of calcium, magnesium, and chloride?



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Minor points:

Section 6 – Composition analysis of collected aerosols – is often unclear and needs to be reorganized or rewritten. Start with clearly stating how many samples were analyzed from which inlets by which EM technique. State the primary data product, limitations, and differences between the EM techniques. For example, aerosol inclusions are mentioned often, but apparently one technique can identify inclusions, where as another infers them via elemental analysis. Other areas in need of help: 580, 18-27; 580,7-12; 581, 1-13. 573, 24: Clearly state the lower and upper size ranges for the 2DC instrument. 577: The Miller and Wang reference has the wrong year. 580, 20-24: Sentences are seemingly contradictory – reword. 581, 14: When referencing the Koehler paper, clarify the relevance of secondary organic aerosol coatings generated via ozonolysis of a-pinene to the present work. 581, 18: Which multi-component particles are you referring to? Are you indicating that inclusions of Na/Ca/K/Mg/S/Cl be detected as rBC by the SP2? Fig 7: What is the significance of the IN concentration correlation with CWC? Since CWC usually correlates with 2DC concentration, this seems to be the same message as Fig 7b, yet the authors highlight it in the text without explanation as to its relevance (577, 22).

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