The review article of Bartels-Rausch et al. is an important contribution to a special issue of ACP (New Perspectives on Air-Ice Chemical Interactions) and brings together an impressive list of authors from the fields of snow/ice physics and chemistry. The manuscript covers a variety of topics related to the physical and chemical processes occurring in snow and how these are impacted by the fundamental/molecular nature of the snow itself. Synthesizing a topic of this magnitude is no small feat, and the authors are to be commended for the timely, updated, and comprehensive review they produced. In general the manuscript is well written and organized, with a few exceptions which I outline below. Overall the topics are well described and recent and relevant studies are discussed. The review is an important contribution to the literature and should be published after considering the suggestions/clarifications outlined below:

General points:

1) I don't see Figure 1 as the best representation of the various microstructural "domains" in snow/ice for someone who is not already well-versed in this topic. Stars and boxes and circles are not very illustrative, and perhaps will cause a misconception for newcomers as they are not shown to scale. The text written over the crystal structure doesn't actually point out carefully the different domains as it is hard to do this with a 2-D image (e.g. disordered interface vs surface). It is great to show an actual microscopic image of a true sample, but I would perhaps include this as a two-panel figure which also includes a drawn "schematic" of the domains using a more 3-D representation … then you could include pictorially areas like micropockets, solid solutions, etc. which later in the manuscript become important topics of discussion.
2) In general in section 2, I think it is important to be very explicit regarding properties dealing with a pure water ice vs situations where solutes are present. As you discuss later, solutes play a big role in changing the microphysical properties, but early on in section 2 it isn't clear that you are mainly referring to pure ice without solutes.
3) Throughout the manuscript there is some inconsistency is using molecular formulas for describing a chemical, or using the written name (e.g. CH3COOH vs acetic acid). I would choose one (probably formulas) and stick to it.
4) In section 3.3.1 I get completely lost in a lot of jargon. I would suggest rewriting this section with a keen eye to making it a summary that a snow physics non-expert can grasp.
5) There are a lot of areas where sentences get quite long and verbose. In a revision, I'd suggest a careful read-through to minimize run-ons.
6) Throughout the manuscript there are areas where commas are used and shouldn't be (or vice versa). E.g. page 30470 should be: "As a result, indirect arguments are

used to explain observed shifts in ice core signals that occur over long time periods." I won't point out all instances of this, but carefully check the revision.

7) I would suggest ending the physical properties section in the same manner as you did for the chemical processes (a conclusions section with numerically listed "take-home points"). This is a great way for a reader to quickly get the main ideas summarized.
8) In general I found the chemistry section had much better flow and organization than the physical section. There was less jargon and fewer instances of repeated discussion points. A revised version should really focus on tightening up the descriptions and making sure the physical section is written so that someone who is not an expert in this field can still appreciate and learn from what is being presented.
9) Processes like sintering are referred to multiple times in the paper. I would quickly describe what this is at its first use.

Specifics:

Page 30412, line 16: "…, the properties of which and their changes with time, critically affect both physical processes and chemical reactivity." This is awkward wording.

Page 30413, line 4: "Other substantial effects include the emission of nitrogen oxides from snowpacks". This statement implies only NOx is emitted, but we know a whole host of species are generated in and emitted from snowpack.

Page 30416, line 23: "…the snow chemistry (Grannas et al., 2007b)…", delete the word "the"

Page 30418, line 8: overlaying should be overlying

Page 30419, line 3: "…a number of different approaches that have been used to overcome this limitation…". Will this be discussed later? This statement leaves the reader hanging.

Page 30420, line 6: "Yet, a statistically sound demonstration of impurities distribution…" What exactly does "statistically sound" mean here?

Page 30421, line 20: The way this is worded, it is not clear if the ice was doped with NaCl, or if the liquid nitrogen is doped with NaCl. (From context and for someone familiar with the field it should be obvious which you meant … but change the sentence so that the wording is clearly reflecting what you mean).

Page 30421, line 30: I would suggest showing on Figure 4 what you mean explicitly when you say "dihedral angle".

Page 30422, line 7: "data is", change to "data are"

Page 30425, line 17: “is elemental specific”, change to “is element specific”

Page 30427, line 7: “from the disorder”, change to “disordered”

Page 30427, line 23: “is the thickness varied across the surface”…change to “does the thickness vary across the surface”

Page 30429, line 4: You mention Monte Carlo simulations here, but don’t appear to talk about them much again (further discussion is on MD simulations).

Page 30430, line 5: change “Molecular dynamics” to MD since you already defined it above.

Page 30431, line 4: what does the “mW” model stand for, is this an acronym for anything?

Page 30432, line 18: “has since long been” is awkward wording

Page 30432, line 20: “…with time in situ and from this the surface area…” awkward wording

Page 30433, line 3: You mention that SEM can help to infer where chemical species are located, but just be clear to point out that you can’t get molecular level info, only elemental, and that SEM is not suitable for all elements (especially for light elements which we care most about in many cases).

Page 30433, line 16: “…as this is an intrinsic property of all crystals.” What exactly does “this” stand for here?

Page 30441, line 6: “…how impurity-induced changes in ice surface properties…”

Page 30433, starting line 24: You discuss the “two regimes” of gas-ice interaction, then proceed to describe the first in detail. The second regime doesn’t start to be described until much later in the paragraph, and gets a bit lost in the subsequent text. I would suggest mentioning up front what those two regimes are … then describe them in turn.

Page 30446, line 25: what is “jump migration”?

Page 30447, line 9: what are “Bjerrum defects”?

Page 30448, line 9: what are “adlayers”?

Page 30448, line 25: “…corroborating the experimental finding…”

Page 30449, line 13: “…self-diffusion on the ice surface…” (also on lines 15 and 30, say ‘the ice surface’)

Page 30449, line 19: “Further MD work by Pfalzgraff et al (2010, 2011); Gladich et al (2011 confirmed…” Do you mean to say: “Furthering the MD work by Pfalzgraff…Gladich et al confirmed…”?

Page 30450, line 14: what exactly is a “long-lasting uptake”?

Page 30451, line 12: “…measure individual processes…”

Page 30451, line 14: what is meant by “re-analysed existing data thereby more reliably the measurements”?

Page 30452, line 9: what are “short-circuits”?

Page 30452, line 19: “…in a confined reservoir”

Page 30452, line 21: “…desorption has also been used…”

Page 30453, line 21: “One way to assess…”

Page 30456, line 30: Should it say “…given the complexity of the involved systems”?

Page 30457, line 12: “…much more rapidly than…”

Page 30459, line 6: “…environmentally relevant…”

Page 30461, line 29: “…precipitates at 250 K…”

Page 30462, line 13: “gradient in the brine, with heavier brine lying above lighter seawater.”

Page 30463: the bromine explosion is not a term everyone may know

Page 30464, line 11: “It takes into account…”

Page 30465, line 23: “…give no conclusive picture.” Picture of what?

Page 30467, line 19: “…6 year period...”

Page 30469, line 19: “…it slightly increased…”

Page 30470, line 21: “…shifts in ice core signals…”

Page 30471, line 26: “…change on a scale of seconds or less…”

Page 30472, line 4: “…of solute that gets expelled…”

Page 30481-30482: The sentence beginning “this change in local pH…” is long and hard to follow. Also, there are missing brackets around references at the end I think, which makes it difficult to follow as well.

Page 30482: “…in ice compare to aqueous solution…”

Page 30486, line 18: “Specifically, recent models have…”

Page 30487, line 2: “…treating them analogously to…”

Page 30489, line 11: “…geometric constraints…”

Page 30490: What is a “4 stream” radiative transfer code? Describe in brief, or don’t get so specific unless it is an important caveat of the model that needs to be mentioned.

Section 4.4, point #3: Parameterizing based on aqueous phase is justified, unless of course the absorption spectra change when frozen…

Page 30493, line 12: “…has not yet been…”

Some of section 5 and 6 seem redundant … these sections may need to be merged, or otherwise better separated to avoid recurring discussion points (e.g. you mention a few times the need to include metamorphism and compaction in models, etc.)