

**RC2: 'Comment on acp-2021-341', Anonymous Referee #2, 22 Jun 2021 reply**

I reviewed, and hence the following comments are targeted for, the abstract and the atmospheric component of this manuscript. For a lengthy review paper like this one, a table of contents would make it easier for the reader to navigate. The abstract needs to be rewritten. All it shows right now is what was done in this paper, followed by a generic sentence “although the scientific knowledge in these regions has increased, there are still gaps in our understanding of large-scale climate-Earth surface interactions and feedbacks”, which tells nothing. You can put a sentence like this in many review papers notwithstanding the topics. The authors need to show a couple of most noteworthy advancements and gaps in knowledge of understanding the key processes in the Arctic-boreal regions. With the impressively long list of authors, the review naturally includes a great ensemble of studies spanning different spheres. However what is lacking is the connection that integrates the cited studies and demonstrates how those studies serve to advance our knowledge of the atmospheric processes in the Arctic-boreal region.

We thank the reviewer for the apropos remarks. This paper, as the PEEX program (2012 - ), is a multi-disciplinary research framework. This type of a framework is a relevant baseline if we, as a scientific community, aim to understand and find new feedbacks and interactions in the land-ocean-atmosphere continuum. For the future it is important to make perspective papers, where results from different disciplines are introduced to a wider scientific audience also with attempts to provide more holistic views on large-scale environmental challenges. The section structure of the paper follows the research agenda structure (land / atmosphere / aquatic / society systems and feedback & interactions) of the PEEX Science Plan. The result are reflected to this structure.

The geographical region discussed in this paper covers the Northern Eurasian region, in this case the boreal (taiga) forest zone, the Eurasian Arctic and China. China is identified as a relevant source area of the atmospheric pollution effecting the Arctic – boreal region, but also as one of a region of interest when discussing the global-scale environmental challenges and large-scale feedbacks. Referring to the so-called “Valeriepieris circle” map (2013) demonstrating that more people is “living inside a circle than outside it” (Danny Quah, London School of Economics and Political Science) concretizes the importance of China for the global climate change and air pollution challenge.

In addition to our responses to the specific comments, we have re-edited the abstract, added “table of contents” and a short description of our literature strategy to help the reader to understand better the chosen structure of the paper and our approach reporting the recent research results of the PEEX program. We frame our overview of the resent results by the PEEX community (including our co-authors), by the papers published in the ACP PEEX Part I special issue and by other relevant sources such as PEEX collaborating projects.

### **Specific comments**

PEEX tackles the Arctic-boreal region (lines 89-90), and the manuscript was supposed to summarize “results obtained during the last five years in the Northern Eurasian region” (lines 91 – 92).

To clarify the geographical scope of the paper, we added the definition of the PEEX region in the abstract and in the section “1. Introduction”.

What is the authors’ definition of “northern Eurasian region”? The one monitoring site and some of the air quality studies from China cited in the manuscript took place in a city of ~32°N latitude. Is that counted as within the “northern Eurasian region”?

We added the definition of the PEEX region in the abstract and in the section “1. Introduction”. *“The PEEX study region consists of the Northern Eurasian Arctic and boreal (taiga) environments, thus the major geographical part of the environments is located in the Russian territory. China was added to the study area*

*in 2013 as it was seen as locally and globally consequential region for climate change, air quality and long-term transport of atmospheric pollutants (Kulmala et al., 2015 a,b; Lappalainen et al., 2016, 2018)."*

That city is in a different atmospheric circulation regime from those northern European and Russian cities and monitoring sites. I agree wholeheartedly that air quality in China and their influence on the Arctic should be studied. However, the inclusion of work from a monitoring site from 32°N latitude in East China seems more like a happenstance than a strategic choice as the inclusion of studies from other locations and areas in the review.

*This is also true. We have added a sentence in the abstract saying "It is also important to recognize that the PEEX geographical region is an area where science-based policy actions would have significant impacts on a global climate"*

I also think the statement of SORPES being the "first such station in China" in need of fact-checking. There are sites in Hong Kong that have been running for decades. There are long-term sites operated under China National Environmental Monitoring Center. There are some sites on city or regional levels such as the ones in Guangzhou (Liu et al., 2013, ACP), which showed data from 2010, and the Sichuan Ecological Environment Monitoring Center from the study by Zhao et al. (2019, Atmos. Pollu. Res.) focusing on Southwest China showing SO<sub>2</sub> and NO<sub>2</sub> concentration data from 2008 to 2018. It is likely that I have not exhausted the list of long-term monitoring sites preceding SORPSE in China.

*We agree with these views. Thus, to be more focused on the SMEAR concept relevant for this paper, we edited the text as follows: "In order to understand these feedbacks, Kulmala (2018) and Hari et al. (2016) emphasized the crucial role of continuous, comprehensive measurements on a network of flagship stations in tackling the air pollution problem in urban China and megacities elsewhere in the world. They also introduced a so-called "Stations for Measuring Atmospheric and Earth surface Relations" (SMEAR) concept, which consists of integrated atmospheric and ecosystem observations allowing the analysis of Earth surface – atmosphere feedbacks and interactions. The first SMEAR-type station in China, the SORPES station located in the Yangtze River Delta, has been operating since 2011 (Ding et al., 2016b)."*

The review is written often times in rather general terms with no key, specific findings from cited works. To make my point, here are a few examples. In lines 652-653, the result cited from Mikhailov et al. (2017) was that "i(I)n summer, precipitation is removing the pollutants from the air and leading to relatively clean atmospheric conditions this region". What is so revelatory here? The scavenging effect of precipitation is commonly known, or did they mean to emphasize the dominant effect of wet deposition of key soluble pollutants that caused smog in the region?

*We re-edited the text as follows: "Based on a five-year study by Mikhailov et al. (2017), it seems that the atmospheric pollution originating from the biomass burning and anthropogenic emissions is significantly affecting the Siberian region. However, in summer precipitation is removing the pollutants from the air and leading to relatively clean atmospheric conditions in this region.".* While the scavenging effect of precipitation is well-known, it is generally not known how this effect compares with other removal pathways or pollutant sources in different locations and seasons.

In the "Methodological and model developments" section (starting in Line 718), they cited Dada et al. (2018) for "a new classification method for atmospheric NPF", and cited Zaidan et al. (2018b) for "a mutual information approach to identify key factors contributing to the NPF", but never stated what those new approaches really were. I understand that a review needs to be succinct but I doubt there is absolutely no way to succinctly explain those new approaches.

*Related to Dada et al. (2018), we added some new information by writing: "The new method uses both ion and aerosol particle number concentration measurements in the size ranges of 2-4 nm and 7-25 nm,*

respectively, is complementary to the traditional event analysis, and can also be used as an automatic way of determining new particle formation events from large data sets.”

Related to Zaidan et al. (2018b), we added some new information by writing: “Zaidan et al. (2018b) used a mutual information approach for a variety of simultaneously monitored ambient variables, including trace gas and aerosol particle concentrations and several meteorological variables, in order to identify key factors contributing to atmospheric NPF.”

In lines 828 – 834: it is not clear to me what specific information I can gain from these generic statements.

Our purpose in this part of the paper is to pay readers’ attention to the several studies with a special relevance for the PEEX program, including a multi-scale modelling approach. The details of the models and used methods are found in the cited papers.

In line 872, the authors stated “the longest urban continuous record is from the SORPSE station in the Yangtze River Delta” and they cited Qi et al. (2015) for the work. The study presented a 2 year worth of dataset. Please explicitly state the length of the dataset for clarity. Following that statement, the authors reviewed the key results: “*NPF was in general the largest source of clusters and nucleation mode (<25 nm) particles, while traffic contributed to all the size ranges and dominated both cluster and nucleation modes on haze days. Aitken mode (25–100 nm) particles originated mainly from local emissions, with additional contributions from regional and transported pollution as well as from the growth of nucleation mode particles. Regional and transported pollution were identified as the main source of accumulation mode (>100 nm) particles*” (lines 875-880). Aren’t these all rather universal, basic knowledge for a megacity? Similar results have been shown in numerous papers over the past decades. What is unique pertaining to the location? What is original about these points? Immediately after, it was the same problem with the review of Bai et al. (2018a) on the PM and O<sub>3</sub> link, which stated that “the contribution in chemical and photochemical reactions was found to be prominent in summer”. It is not clear to me what readers gain from a statement like this.

Following the suggestion by the referee, we added the following after our citation to Qi et al. (2015): “..., covering almost a decade of measurements, ...”

Concerning the sources of particles in different size ranges, we do not fully agree with the referee. Although sub-micron particle sources have been discussed a lot in studies made in urban Europe and Northern America, very few studies discussing the origin of different size modes in megacity environment have been published. To our knowledge, the measurements by Zhou et al. (2020) cited here are the first reported study which covers the whole submicron size range down to sizes the cluster mode, ever made in a polluted megacity.

Related to the comment on Bai et al. (2018), we combined 2 sentences in the text to make the message clearer: “A photochemical link that related the production of fine PM and O<sub>3</sub> to VOCs was detected, and this mechanism was found to be prominent in summer.”

In lines 909-910, it was stated that “i(I)n line with the proposed mechanism, Shen et al. (2018) showed that aerosol optical properties evolve clearly during the development of multi-day pollution episodes in heavily polluted regions”. Again, what exactly in this result contributes to understanding the BL-PM link there? More importantly, how are all those results reviewed here contributing to understanding Arctic-boreal processes?

We agree that the connection between this information and our understanding on BL-PM link is rather weak. We removed the sentence including this statement from the manuscript.

#### More specific comments:

1. Lines 529 – 531: N<sub>2</sub>O came out of nowhere and no references were cited.

We removed the N<sub>2</sub>O results and corrected the text on lines 529 – 531 to the following format: “There are tendencies of a significant growth or suppression of soil CO<sub>2</sub> fluxes across different types of human impacts, such as forest fires, trampling, settlements, reindeer grazing and clearcuts on cryogenic ecosystems in Russia (Karelin et al., 2017). For example Ivanhov et al. (2019) analyzed ...”

2. There was spillover between sub-sections. Examples: under Northern Eurasian CO, they talked about CH<sub>4</sub> again (line 569). Before the review on black carbon starting in line 668, they already reviewed a bit about black carbon in previous subsections.

To avoid confusion, we removed information on CH<sub>4</sub> from this paragraph.

3. The authors had the tendency to use adjectives to describe results, such as “this amount had decreased remarkably in the Moscow urban environment” (line 675). How much is “this amount”? What amount qualifies as “remarkable”? Be quantitative.

We added the urban concentration (Moscow) “3.73%±0.39% per year”.

4. Lines 794 -: The authors started with stating “new atmospheric aerosol instruments have been deployed in the PEEX area”, but then went on talking about a new laboratory (AHL). They then merely mentioned that “the state-of-the-art instruments” were used. It was confusing. I associated “new” with “novel”, instrumental advancements. But none of the following information suggested that.

The formulation in the manuscript was not optimal as underlined by the referee’s comment. The deployed instrumentation is not new but rather state-of-the-art. The added value originates from deploying this set of equipment into a region that has not been explored with such capacity earlier. To clarify the message, we formulated the paragraph as follows: “Recently, a new atmospheric observation site equipped with state-of-the-art atmospheric aerosol instrumentation was deployed in Beijing, China (Liu et al. 2020). At the Beijing University of Chemical and Technology (BUCT), the Aerosol and Haze Laboratory (AHL) was established in 2018 - 2019, providing novel insights into air pollution in a comprehensive manner. The station hosts comprehensive instrumentation to concentrations of atmospheric trace gases, aerosol particle size distributions and mass concentrations, particle chemical composition on the levels from molecules, clusters and nanometer to micrometer sized aerosol particles. For example, the first results showed increased cluster mode particle number concentrations during NPF events, whereas during haze days accumulation mode particle number concentrations were high (Zhou et al., 2020). The observations have enabled to quantify number emission factors and underlined the importance of traffic (Kontkanen et al. 2020). Daytime sulfuric acid concentrations in Beijing were typically around  $4.9 \times 10^6 \text{ cm}^{-3}$  (Lu et al. 2019). During these measurements, an evidence was found on significant nighttime sulphuric acid production, yielding gaseous sulphuric acid concentrations of  $1.0$  to  $3.0 \times 10^6 \text{ cm}^{-3}$  (Guo et al., 2021). For further results, see also section 2.2.2 Urban air quality and megacities.”

Added references:

Liu, Y.C., Yan, C., Feng, Z., Zheng, F., Fan, X., Zhang, Y., Li, C., Zhou, Y., Lin, Z., Guo, Y., Zhang, Y., Ma, L., Zhou, W., Liu, Z., Dada, L., Dällenbach, K., Kontkanen, J., Cai, R., Chan, T., Chu, B., Du, W., Yao, L., Wang, Y., Cai, J., Kangasluoma, J., Kokkonen, T., Kujansuu, J., Rusanen, A., Deng, C., Fu, Y., Yin, R., Li, X., Lu, Y., Liu, Y., Lian, C., Yang, D., Wang, W., Ge, M., Wang, Y., Worsnop, D.R., Junninen, H., He, H., Kerminen, V.-M., Zheng, J., Wang, L., Jiang, J., Petäjä, T., Bianchi, F. and Kulmala, M. (2020) Continuous and comprehensive atmospheric observation in Beijing: a station to understand the complex urban atmospheric environment, Big Earth Data 4, 295-321.

Kontkanen, J., Deng, C., Fu, Y., Dada, L., Zhou, Y., Cai, J., Daellenbach, K.R., Hakala, S., Kokkonen, T.V., Lin, Z., Liu, Y., Wang, Y., Yan, C., Petäjä, T., Jiang, J., Kulmala, M. and Paasonen, P. (2020) Size-resolved particle number emissions in Beijing determined from measured particle size distributions, Atmos. Chem. Phys. 20, 11329-11348.

Lu, Y., Yan, C., Fu, Y., Chen, Y., Liu, Y., Yang, G., Wang, Y., Bianchi, F., Chu, B., Zhou, Y., Yin, R., Baalbaki, R., Garmash, O., Deng, C., Wang, W., Liu, Y.C., Petäjä, T., Kerminen, V.-M., Jiang, J., Kulmala, M. and Wang, L. (2019) A proxy for atmospheric daytime gaseous sulfuric acid concentration in urban Beijing, Atmos. Chem. Phys. 19, 1971-1983.

5. Lines 811-813: Were the authors suggesting that human influence suppressed NPF?

Not necessarily. Human influence may certainly suppress NPF due to higher pre-existing aerosol loadings, but it may also favor it by providing SO<sub>2</sub> which produces sulfuric acid – a major precursor for atmospheric NPF. We do not currently know which of these effects is more important e.g. in Siberia, but our new PEEX-related measurements will definitely provide new insight into this.

6. Lines 820 – 823: Did this development improve model simulation of aerosol-radiation and -cloud interactions?

We do not yet have concrete data on this. In principle, a more accurate representation of the particle number size distribution, together with size resolved particle emissions into the atmosphere, should enhance the accuracy of large-scale model simulations.

7. Some references are missing, such as Wang et al. (2017a, 2019)

We do not fully understand this comment, as these two references appear both in text and in the reference list. Maybe there is confusion due to the fact that in the original reference list, references starting with either V or W were mixed. We fixed this problem by putting all these references in a correct alphabetical order.

8. Lines 947 – 952: references are needed

We removed the general statement in the beginning of this paragraph. The revised version now reads: *“The Russian part of the Barents Euro-Arctic region includes severe emission ‘hot spots’ for air pollutants. The Kola Peninsula, despite the presence of areas with undisturbed nature in the eastern part, is the most industrially developed and urbanized region in the Russian Arctic. The main polluters are...”*

With the revised text, we think that the references already given in this paragraph are sufficient.

9. The manuscript can use a good amount of editing.

We have carefully checked the language and grammar of the latest version of the manuscript.