9 September 2021

To: Editor, ACP PEEX Part II special issue

This a cover letter for a R1-R2-R3 reply of manuscript to ACP PEEX Special Issue by Hanna K. Lappalainen, Tuukka Petäjä, Timo Vihma, Jouni Räisänen, Alexander Baklanov, Sergey Chalov, Igor Esau, Ekaterina Ezhova, Matti Leppäranta, Dmitry Pozdnyakov, Jukka Pumpanen, Meinrat O. Andreae, Mikhail Arshinov, Eija Asmi, Jianhui Bai, Igor Bashmachnikov, Boris Belan, Federico Bianchi, Boris Biskaborn, Michael Boy, Jaana Bäck, Bin Cheng, Natalia Ye Chubarova, Jonathan Duplissy Egor Dyukarev, Konstantinos Eleftheriadis, Martin Forsius, Martin Heimann, Sirkku Juhola, Vladimir Konovalov, Igor Konovalov, Pavel Konstantinov, Kajar Koster, Elena Lapsina, Anna Lintunen, Alexander Mahura, Risto Makkonen, Svetlana Malkhazova, Ivan Mammarella, Stefano Mammola, Stephany Buenrostro Mazon, Outi Meinander, Eugene Mikhailov, Victoria Miles⁶, Stanislav Myslenko⁵, Dmitry Orlov⁵, Jean-Daniel Paris²⁶, Roberta Pirazzini, Olga Popovicheva, Jouni Pulliainen, Kimmo Rautiainen, Torsten Sachs, Vladimir Shevchenko, Andrey Skorokhod, Andreas Stohl, Elli Suhonen, Erik S. Thomson, Marina Tsidilina, Veli-Pekka Tynkkynen, Petteri Uotila, Aki Virkkula, Nadezhda Voropay, Tobias Wolf, Sayaka Yasunaka, Jiahua Zhang, Yubao Qiu, Aijun Ding, Guo Huadong, Valery Bondur, Nikolay Kasimov, Sergej Zilitinkevich, Veli-Matti Kerminen and Markku Kulmala "Recent advances on the understanding of the Northern Eurasian environments and of the urban air quality in China - Pan Eurasian Experiment (PEEX) program perspective. The manuscript introduces the recent scientific results from the Northern Eurasia regions and China and summarize recent progress in the understanding of the land – atmosphere – ocean systems feedbacks. 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.

We checked and corrected point by point the corrections indicated by the R1, R2 and R3. Find here below our detailed replies.

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

Sincerely yours,

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RC1: <u>'Comment on acp-2021-341'</u>, Anonymous Referee #1, 22 Jun 2021 reply

General Comments

This paper summarizes the results of a number of projects under the PEEX program, and pulls them into a single overview manuscript. The paper explores linkages between the various results and their connection to impacts, for example on human health. Overall, the paper contains a lot of potentially useful information with many associated references. However, I had a hard time understanding who the intended audience was. The paper could be a useful reference for scientists looking for information and related papers on environmental parameters in the Russian Arctic. However, in order to achieve this, some reorganization would be needed to help the reader find what they are looking for. Furthermore, some of the descriptions of the work lack detail. The specific comments below give examples of this, for the introduction, atmospheric composition, and synthesis sections.

We thank the reviewer for the apropos remarks. This paper, as the PEEX program (2012 -), is a multidisciplinary 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 that 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

Line 87 (abstract): Include a clear statement of why a review paper is needed. Consolotation/linkages between datasets collected over large spatial area across multiple research themes?

We edited the abstract so that it will better describe the scope and context of the paper. We also added a "table of content together" with several cross-references.

Line 87 (abstract) and Line 106 (introduction): Why are urban megacities in China included in the paper? Would this fit better in a separate accompanying paper?

Chinese megacities and air quality have been identified as major environmental challenges in the PEEX Science Plan / program. We added the following text to clarify this aspect: "*The Pan-Eurasian Experiment (PEEX) Science Plan, released in 2015, addressed a need for a holistic system understanding and outlining the most urgent research needs for fast chancing in the Arctic-boreal region. Air quality in China together*

with long-range transport of the atmospheric pollutants were also indicated as one of the most crucial topics of the research agenda. These two geographical regions, the Northern Eurasian arctic–boreal region and China, especially the megacities, were identified as a "PEEX region". It is also important to recognize that the PEEX region science-based policy actions will have significant impacts in a global scale."

Line 128: What is the Silk Road Economic Belt Program?

We added the following text and reference:" *initiated by the President XI Jinping in 2013*" (Dave and Kobayashi 2018)

Line 152: I am a bit confused about the study area. Can a map be produced that summarizes the region of interest?

We added the following text and references: "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., 2015b; Lappalainen et al., 2016, 2018)."

Line 533: In a number of places, more information/context is needed. For example, the discussion around line 533 indicates that wildfires caused CO2 increases. However, no information is provided on wildfire activity. Furthermore, it is not clear what methods are being used to detect increases over what averaging period.

We added the following text to clarify the situation: "There are tendencies of a significant growth or suppression of soil CO_2 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 CO_2 measurements during 2010-2017 and reported CO_2 concentration increases of 20 ppm in Tiksi at a coast of Laptev Sea and of 15 ppm at the Cape Baranov station. They also detected that wildfires in Siberia can lead to a parallel increase of the CO_2 concentration increased on average by 2.0 ppm yr⁻¹ during 2006-2013 in central Siberia, with a large inter-annual variations."

Line 566: Did wildfires impact CO concentrations?

Ratikin et al. (2018) concluded in their paper "Such pattern of changes in atmospheric composition especially in CO trends cannot be explained by growth of anthropogenic and/or wild-fires emissions. Possible reason of beginning of CO growth may be the change in the ratio of the natural sources and sinks with a significant role of atmospheric photochemical mechanisms."

Line 567: Why are CO and CH4 being compared and across different time periods? I'm having a hard time interpreting the results.

To avoid confusion, we removed information on CH₄ from this paragraph.

Line 574: What does 0.9-1.7% per year refer to? Background concentrations only? What about urban concentrations?

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

Line 581: What method was use to determine that monoterpenes are major contributors to O3 formation?

Berezina et al. (2019) evaluated the relative importance of VOCs in ground-level ozone formation by using ozone-forming potential (OFP) equation by Carter W. P. L. (1994). Development of ozone reactivity scales for volatile organic compounds. J. Air Waste Manage. Assoc., 44, 881–899.

Line 589: Discussion of stratospheric ozone should be in a separate paragraph and clearly identified as stratospheric to avoid confusion. Or perhaps in can be moved to the "UV variations" section?

We added cross-references between "2.2.1 Atmospheric composition and chemistry (Q4) / ozone" and "2.4.3 Natural hazards (Q12) / UV variation".

Line 598: Define threshold for NFP event days.

This is probably slightly too detailed information for this paper. We removed the two sentences related to this issue from the paper.

Line 627: Should the discussion on wildfire trends be included as a standalone section? I imagine that this is information that readers might be interested in (and want to find easily) for a variety of applications. Furthermore, this could then be cross-referenced from all sections discussing impact of wildfire trends on air quality.

We agree that the results / aspects related to "wildfires" or "fires" are included in several sections: "Black carbon and dust in the atmosphere and snow", "Anthropogenic emissions and environmental pollution in Russia", "Examples of air pollution episodes and related health effects" and "Future research needs from the system perspectives". The chosen approach builds on the PEEX research program structure. To avoid confusion, we added several cross-references between the relevant sections. In addition, we discuss the various perspectives and importance of fires in the section "*Future research needs from the system perspectives*"

Line 718: I feel like the section **''methodologies and model developments''** could be split up and moved into the relevant section. For example, readers interested in NFP may not think to read this part of the paper.

We would prefer to keep this section as it is. However, we understand the critics so, to make it easier for a reader to find relevant information, we added the "table of content" and cross-references between the sections "*Methodological and model developments related to atmospheric chemistry and physics*" and "2.2.2 Urban air quality and megacities".

Line 853: Is there trend information for NOx and SO2 emissions in the area?

We added information on NO₂ and SO₂ trends based on Wang et al. (2019).

Line 859: "relative frequent occurance" -- compared to what?

We modified this part of the text into the following form: "Compared with most other urban environments investigated so far, measurements in urban China demonstrated a relatively frequent occurrence of atmospheric new particle formation (NPF), and the observed NPF events were typically characterized by high particle formation rates and strongly size-dependent growth of newly-formed particles (Kulmala et al., 2016b; Wang et al., 2017b; Chu et al., 2019)."

Line 883: "... were analyzed for North China..." what method/data were used?

The data based on "monthly satellite and ground observed atmospheric constituents" were used.

Line 936: This paragraph and the ones after it contains a lot of information about air quality that is outside urban environments, as well as information on deposition and emissions. If a reader was seeking this information, they would not think to look under "**2.2.2 Urban air quality and megacities**" Reorganization of content is needed.

We added cross- references to clarify the content between these two sections.

Line 975: "... emissions of PM were observed..." what method was used?

The paragraph was updated as the following: "The annual yearbook "The State of Atmospheric Pollution in Cities on the Territory of Russia" for 2018 (Roshydromet and GGO, 2019) sates the highest atmospheric emissions of PM were observed in Siberian and Ural cities. In Novokuznetsk and Omsk, the observed PM was the highest (> 30 000 tons per year) while emissions from other cities such as Angarsk and Chelyabinsk were lower (< 20 000 tons per year). Note that in the 2015-2019 yearbooks, emissions from only stationary sources were provided due to revisions (approved and implemented in November 2019 by the Russian Ministry of Natural Resources and Ecology, MNRE) of methods applied for estimation of emissions into the atmosphere from mobile sources. Depending on a source type, different methods to calculate emissions are applied (MNRE, 2019). For the gaseous compounds, such as SO₂, the maximum emissions included very high from Siberian cities (e.g. Norilsk, Novosibirsk, Novokuznetsk, Omsk, Ufa, Irkutsk, Angarsk) and from North-West Russia cities (Zapolyarny, Nickel, Monchegorsk). High NO₂ emissions were observed in Novosibirsk, Omsk, Angarsk and Chelyabinsk. The CO integral urban emissions depend on a city size. These varied from less than 10 Gg yr⁻¹ (for small regional centers like Vladimir, Kursk, Samara) to 406 and 804 Gg yr⁻¹ for large metropolitan areas such as St. Petersburg and Moscow. As a whole, an analysis of spatio-temporal variation of trace gases in the boundary layer over Russian cities indicated significant emission variations between the urban environments and remote sites (Elansky et al., 2016)."

& 2 references were added:

Roshydromet and GGO (2019): The State of Atmospheric Pollution in Cities on the Territory of Russia for 2018. Yearbook. Federal Service on Hydrometeorology and Monitoring Environment, Roshydromet & A.I. Voeikov Main Geophisical Observatory, ISBN 978-5-9500883-8-4, St.Petersburg, 250 p., In Russian (http://voeikovmgo.ru/images/stories/publications/2019/ejegodnik_zagr_atm_2018+.pdf)

MNRE (2019): Methods for calculating emissions of pollutants into the atmosphe from stationary sources. Ministry of Natural Resources and Ecology. In Russian (https://www.mnr.gov.ru/docs/metodiki_rascheta_vybrosov_vrednykh_zagryaznyayushchikh_veshchestv_v_atmosfernyy_vozdukh_statsionarn/perechen)

Line 980: CO emissions are discussed. Why isn't this included (or cross-referenced) in the section describing ambient CO?

This paragraph discusses cities that are emission hot spots for different pollutants, including CO. Emission hotspots are a very different topic from ambient concentrations trends of the same pollutants (as discussed earlier in the paper in case of CO).

Line 1653: Should "UV variations" be linked to the discussion of stratospheric O3 (line 589)

We added cross- references between these sections.

Line 1665: "Air pollution and related health effects" - the discussion of air pollution in Bergen is very specific and also does not have any direct ties to human health. Perhaps this discussion should be moved into the section about atmospheric composition so that a reader interested in meteorological impacts can find it easily?

To clarify this, we modified the title of the paragraph as following "Examples of air pollution episodes...".

Line 1665: "Air pollution and related health effects" - the scope of this section seems vary narrow and focused on some example studies. What about the broad picture including all the species/locations monitored through the project? For example, there is no mention of urban air quality in megacities in this section.

Unfortunately, it has not yet been possible to carry out this type of a wide systematic approach under the PEEX framework. There is still a way to go towards a more coordinated observation system, which would allow us this type of reporting.

Line 1757: Why is BC highlighted as the most important air quality pollutant here for future study in relation to human health?

We fully agree. We removed "black carbon" from the text.

Line 1760: "... was found to vary from serious effects on population health..." I didn't see this evidence of health studies in the paper. Perhaps rephrase to something like "Air pollution was at concentrations that can be harmful to human health"?

We removed "population health" and edited the text according to this recommendation.

Technical comments

• Quite a few grammatical errors (recommend copy-editing).

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

• Citation: https://doi.org/10.5194/acp-2021-341-RC1

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

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 multidiciplinary 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 that 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 \sim 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 SO₂ and NO₂ 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's 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 O₃ 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_3 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: N₂O came out of nowhere and no references were cited.

We removed the N₂O results and corrected the text on lines 529 - 531 to the following format: "There are tendencies of a significant growth or suppression of soil CO₂ 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 CH₄ 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 CH₄ 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", instrumentational 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-ofthe-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 quatify number emission factors and underlined the importance of traffic (Kontkanen et al. 2020). Daytime sulfuric acid concentrations in Beijing were typically around 4.9×10^6 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 3.0×10^6 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 SO2 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 put putting all these references in a correct alphabetical order.

8. Lines 947 – 952: references are needed

We removed the general statement in the beginning 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 paragraphs 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.

Citation: https://doi.org/10.5194/acp-2021-341-RC

RC3: 'Comment on acp-2021-341', Anonymous Referee #3, 25 Jun 2021

This long review paper present the research progress of Pan Eurasian Experiment (PEEX) program, as well as that of urban air quality in China. It is an extremely comprehensive summary paper that includes the land ecosystem processes, thawing permafrost, ecosystem structural change, atmospheric composition and chemistry, urban air quality in megacities, weather and atmospheric circulation, changing water systems, snow, sea ice and ocean sediments, marine ecology, lake and rivers, anthropogenic and environmental impact on society, and natural hazards. Given its wide scope and my limited expertise, I focused mainly on the air quality part. In general, the review appropriately summarized the relevant work of PEEX, and it is well organized and presented. I only have some minor concerns. While it stressed the PEEX, however, some more information could be provided for **further explanation of changed air quality in mega cities. Relatively long-term trends in atmospheric composition should also be provided**.

We thank the reviewer for the apropos remarks. This paper, as the PEEX program (2012 -), is a multidiciplinary 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 that 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.

1. Section Northern Eurasian carbon monoxide (from line 566). Why was the CO elevated? As it is an indicator of energy efficiency, does it mean the energy efficiency went down in recent years?

Ratikin et al. (2018) concluded in their paper "Such pattern of changes in atmospheric composition especially in CO trends cannot be explained by growth of anthropogenic and/or wild-fires emissions. Possible reason of beginning of CO growth may be the change in the ratio of the natural sources and sinks with a significant role of atmospheric photochemical mechanisms."

2. Section Northern Eurasian Ozone (from Line 576). This paragraph stated the important chemical species of O3 formation in different regions. I would suggest the authors collect more information and provide a relatively long-term trend in O3 concentration in Northern Eurasia. Moreover, the driving forces of O3 change should also be analyzed.

We think this a relevant point, but such type of a trend analysis has not yet carried by the PEEX collaboration. We report and overview here only the existing results.

3. Section Black carbon and dust in the atmosphere and snow (from line 668). Similarly, could you reveal the long-term change of black carbon in Arctic and Northern Europe/Asia, and provide the main reasons for the changes.

Unfortunately, we do not have long-term black carbon data to make any trend analysis for the Russian part of the Arctic. BC trends for a few European and Northern America Arctic sites have been published in a scientific literature. We think it is not worth either repeating or trying to re-analyze those results here.

4. Section Air quality in China-recent observations (from line 846). There are a lot of studies stressing the changed air quality (i.e., reduced PM2.5 and increased O3) since 2013. However, very limited papers were selected in the review. The authors need to **explain their strategy in literature review**. Moreover, the reasons for the air quality change should also be well presented. For example, the implementation of national plan of air pollution control is considered to be the most important reasons for the improved air quality. It should be reviewed and presented here.

We have added a following description of our strategy in literature review: "For the literature material, we have combined literature searches with summaries the scientific approach by the PEEX community. We used the following sources for demonstrating the results: (i) individual input sent by the PEEX research community, (ii) content of the scientific papers published in Atmospheric Chemistry and Physics (ACP) PEEX special issue in 2016-2019 (www.atmos-chem-phys.net/special_issue395.html), (iii) scientific output from PEEX labeled projects <u>www.atm.helsinki.fi/peex/index.php/projects</u>]. For the individual input we asked the PEEX research community to identify the main published papers in peer reviewed journals for each question out of their own work and connect the work to one of the 15 science questions introduced in the PEEX science plan. Based on the abstracts we listed "addressed research themes" over last 5 years per PEEX key topical areas (Table 1), which we review in more detail in section 2."

5. The same section as Question 3. Why include only the NPF studies conducted at SORPES in YRD region? How about studies in other regions or sites? Is it because of the limited scope of PEEX?

The studies on NPF discussed in this paragraph have been conducted not only in SORPES and YRD, but also in Beijing. Furthermore, one of the studies made in PEEX (Chu et al., 2019), and also mentioned in this paragraph, reviewed practically all the NPF studies make in China prior to 2019.

6. Section anthropogenic emissions and environmental pollution in Russia (from 916). Given the different development stages and air pollution control plans between China and Russia, it would be interesting to compare the long-term trends in emissions and air quality for the two countries.

We think this a highly relevant point, but such analysis has not yet been carried out by the PEEX collaboration. It would need specific resources.

7. Some language errors need to be corrected. Line 501, section 3.1.1 or section 2.1.1? You don't need to give the full name of NPF as it appeared earlier.

We have corrected "section 3.1.1" to "2.1.1"

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

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