Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC1
https://doi.org/10.5194/acp-2020-1202-RC1, 2021

Comment on acp-2020-1202

Anonymous Referee #2

Referee comment on "Wintertime sub-arctic new particle formation from Kola Peninsula sulphur emissions" by Mikko Sipilä et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-1202-RC1, 2021

Referee Comment:

General scientific comment:

The manuscript presents evidence on how important strong pollution sources emittting SO2 can influence or initiate nucleation events downwind of the sources in remote areas. The study is mostly based on case studies. One case study is presented in detail in the main manuscript. Three other case studies are presented in the supplemental part whereof in one case measured acids and ion clusters are missing, in a second one measured ion clusters are missing and in a third one measured ion clusters are missing and other explanations are listed for the respective NPF event.

The study delivers interesting results on pollution induced new particle formation, but some more details especially on statistics with respect to the whole measurement period are strongly recommended to include before final publication. The manuscript misses a detailed overview to put the case studies in the context of the full measurement period which was a couple of month in the winter period. An overview of all nuclation events, with regard to levels of SO2 concentration, sulfuric acid concentration (measured and modelled), wind direction, available UV radiation, quality of event (number of clusters and further growth, etc.) would place the events in a context which is needed the evaluate the abundance of natural and anthropogenic nucleation events during the measurement period. Right now the manusctipt delivers case studies within a larger measurement period and it is difficult to follow the significancee of the findings. What kind of types of NPF was observed in March? Were there periods when conditions for NPF were favourable based on pollutant emissions, but NPF did not take place? In addition, the paragraph on growth of observed particles and their contribution to CCN remains a bit vague and needs to be elaborated on.

Figure S1 shall be included in the main manuscript.

The manuscript requires a native speaker to improve the language!

Detailed scientific comments:

Abstract

-

Introduction

Page 1, line 25-26:

Comment: I would say that SO2 contributes to acidification of … by atmospheric aerosol and cloud formation … (check the sense of the sentence).

Page 2, line 32-33:

Comment: Can you give a reference here?

Page 2, line 45-47:

Comment : Check syntax of the sentence!

Page 3, line 68:

Better: … was published by …

Page 3, line 89:

Comment: 300 km is not so close in terms of distance between the observations and the emissions. I recommend to write the disctance explicitely here and also at other places in the manuscript (see abstract). 300 km gives some time for transport and corresponding processing!

2 Methods

2.1 Site and time of the study

-

2.2 Instrumentation

Pag4, line 103:

The part of the DMPS for ultrafine particle detection malfunctioned …

Pag4, line 109-114:

The paragraph is misunderstanding, write exactly when this instrument was used and since when with the switcher, etc.

2.3 Nucleation rate calculation

-

2.4 Sulfuric acid proxy calculation

This whole section leaves some questions. Because of missing data, the authors make a number of assumptions on the sulfuric acid concentration. Sulfuric acid is calculated using SO2 oxidation by OH which is proxied by global radiation; Criegee Intermediates proxied by monoterpene and ozone concentrations, condensation on pre-existing aerosol, assumption on monoterpenes, and global radiation assumption was used. This needs to be

verified and some general evaluation of this method using the campaign dataset should be discussed.

2.5 Trajectory analysis

-

3 Results and Discussion

3.1 New particle formation during the measurement period

General comment: As the study is mainly based on case studies, an overview table of these studies is needed stating the differences and similarities of the different events. The descrption here is otherwise confusing to evaluate which events follow certain patterns and which do not. What about situations where NPF would be expected to happen based on the general conditions, but it did not?

Page 6, line 170 - 179:

Comment: Please redo the figure, it is not possible to see specific days because of the overall scaling of the time axis. It might be an idea to add boxes where events have taken place and label these boxes with the respective event dates.

Page 6, line 177:

… consequent days? …

3.2 Case study 28th – 29th January 2020

-

3.2.1 Meteorological situation and trace gas concentrations

Page 7, line 204-205:

Comment: It must be possible with Hysplit to calculate boundary layer height aalong the trajectory. This is very useful to see that air masses even at low altitudes were not above the boundary layer height and air was not mixed in from above.

3.2.2 Aerosol precursors

Page 8, line 224:

Comment: Do you mean a gradient in temperature per meter?

Page 8, line 233:

Can you add a reference here?

3.2.3 New particle formation

Page 9, line 264:

The scale in the graph only shows up to 0.06 while the full peak is observed? Please correct the text here!

Page 11, line 323-325:

Comment: Check syntax of this sentence!

Page 11, line 323:

… on that data …

Page 12, line 345:

… compared to what was measured here …

3.2.4 Particle growth and relevance for CCN-concentrations

General comment: This paragraph does not really give useful information on the availability of CCN related to pollution induced NPF events. There is a need for a more thorough analysis.

Page 12, line 365:

Comment: Here a more thorough explanation for the potential particle growth is needed. A literature review on potential VOCs in a similar environment during similar seasons would be sufficient here.

Page 12, line 367-368:

Comment: Check the sentence on nitric acid, this does not make sense!

Page 12, line 373-374:

Again, this explanation about the growth does not say anything. You say that some concentrations were measured, but too small, but in the vicinity probably higher, explaining the process is absolutely vague and with no real evidence on the process.

Page 13, line 397:

Why is March excluded here? If March does show other origin for NPF compared to the pollution transport, this could be well used to show the difference between anthropogenically and naturally initiated NPF. I do not understand why valuable data are omitted here?

Page 13, line 402-403:

Give detailed evidence for this statement! What is that based upon?

Page 14, line 423:

Comment: Shorten this sentence and split it in two, it is too long.

4 Conclusions

Page 14, line 410-418:

I am missing some statistics of how often NPF was observed with regard to certain wind directions and concentrations exceeding a threshold of SO2 occurred stating pollution was initiating the process, etc.. Also, such situations should be put into context when

conditions were favorable and NPF did not occur. See general comment above!

Language comments:

Introduction

Page 2, line 46:

… precipitates …

Page 2, line 62:

… exist …

Page 3, line 66:

Leave out … » of « …

2 Methods

2.1 Site and time of the study

Page 3, line 93:

The station …

Comment : As a general comment use the article « the » when describing things. I think it is not good grammar saying « Staion is located… ». This shall be « The station is located … ».

Page 4, line 97:

… the closest …

2.2 Instrumentation

-

2.3 Nucleation rate calculation

-

2.4 Sulfuric acid proxy calculation

-

2.5 Trajectory analysis

-

3 Results and discussion

3.1 New particle formation during the measurement period

Page 6, line 170:

… aerosol number size distribution …

Page 6, line 171:

… « observed » … instead of … « recorded » …

Page 6, line 174:

… took place …

Page 6, line 181:

… easterly winds …

Page 6, line 182:

…westerly winds …

Page 6, line 185:

…and were transported …

3.2 Case study 28th – 29th January 2020

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3.2.1 Meteorological situation and trace gas concentrations

-

3.2.2 Aerosol precursors

Page 8, line 230:

… are observed …

Page 8, line 231:

… remains …

Page 8, line 234:

… ends up …

3.2.3 New particle formation

Page 8, line 244:

… clusters …

Page 10, line 284:

… explain …

Page 11, line 341:

… nucleation rates …

Page 12, line 348:

… exist …

3.2.4 Particle growth and relevance for CCN-concentrations

Page 12, line 353:

… nucleation rates …

Page 12, line 363:

… than these …

Page 12, line 375:

… in the close environment of emission sources with high …. concentrations …

Page 13, line 377:

… diameters …

Page 13, line 379:

… of growing modes …

Page 13, line 381:

… few tens of …

Page 13, line 382:

… at diffèrent supersaturations …

Page 13, line 383:

… containing …

Page 13, line 384:

… concentrations …

Page 13, line 385:

… shows an increase …

Page 13, line 386:

The concentration of particles larger than …

Page 13, line 387:

… these events …

Page 13, line 390:

… a 1-week period of easterly winds …

Page 13, line 393:

… westerly winds …

Page 13, line 393:

… than the average …

Page 13, line 400:

… towards …

Page 13, line 400:

… an accurate …

Page 13, line 400:

… to CCN concentrations …

Page 13, line 402:

… towards …

4 Conclusions

Page 14, line 407:

… concentrations of sulfuric acid were high enough …

Page 14, line 420:

… few nm and larger …

Page 21, line 655:

… into the atmosphere.

… processing sites.

Page 22, line 660:

… aerosol number size distribution … (Comment: include size range)

Page 27, line 693:

…. easterly winds …

Page 27, line 694:

…. westerly winds …

Supplement

Page 1, line 7:

… linear diameter scale.

Page 1, line 10:

… intensive nucleation process …

Page 3, line 9:

… particle number size distribution …

Page 7, line 10:

… close to zero …

Page 8, line 2:

… particle number size distribution …