

We thank the Reviewers for the insightful and helpful comments and for the careful reading of the manuscript.

Response to the comments made by Earle Williams, the Reviewer #1.

The main goal of this study, the assessment of the seasonal variation of the global electrical circuit from a polluted continental location, is very worthwhile and strongly appreciated, and from that standpoint every effort should be made to get the work published. The true seasonal variation of the DC global circuit is still not firmly established, even in measurements of the ionospheric potential (with contradictory seasonal variations from Muhleisen/Fishcer and from Markson). My main concern, already communicated to the second author, is the limited approach taken here: the measurement of condensation nuclei to characterize the atmospheric medium, rather than the measurement of air conductivity with a pair of Gerdien tubes. In the following, suggestions are made for improving the present approach, but in the end we suggest a conductivity approach that rests on observations readily available to the authors at the same Świder location (by Marek Kubicki).

Summary: Consider for publication after major revisions (and possible inclusion of conductivity measurements)

Reply: We thank the Reviewer for the careful and critical reading of the manuscript and the constructive comments made. After completing the digitisation of all Świder atmospheric electric 1965-2005 data, we were also able to present new results that include analysis of the annual variation of the conductivity measured at Świder (by the Gerdien tube). More details and a new plot are given in an added section of the manuscript.

Response to substantive issues:

(1) Characterization of the medium with a CN counter

For reasons of time, the authors have been reluctant to get involved with the Świder conductivity measurements, and instead have chosen to rely on a CN counter. (The distinction between large and small ions of atmospheric electricity is not mentioned.) If the large ion population is reliably measured with the CN counter, then the air conductivity can be inferred (though this is not the best approach to obtaining air conductivity, as it is an indirect one). Unfortunately, the documentation on what is being measured with the Scholz counter is thin, even to the point of not disclosing what supersaturation value is achieved. It would also be valuable to know the instrument response to clean oceanic air but that is of course not easily obtained. In any case, a big improvement in the characterization of the Scholz counter is essential here.

Reply: In this study we wanted to concentrate rather on PG and aerosol number concentration as the PG is the most commonly observed atmospheric electric parameter worldwide, and there are more and more observations of aerosol concentrations, so other stations could make a similar analysis to investigate the results at their location.

We admit the information about the instrumentation for aerosol concentration measurements is limited as it is very scarce in the observatory yearbooks. We would like to add more details about the measuring apparatus which we found in the literature, in the archive materials of the Institute and from the observatory staff. A Scholz counter is a type of condensation counter constructed by Scholz as an improvement of the Aitken counter (McMurry 2000), designed to measure the concentration of condensation and nuclei nearly the total concentration of aerosol. At Świder, the main part of the small Scholz counter was a brass cylindrical chamber with a volume of 102 cm³ and a height of 4 cm,

with which the adiabatic expansion ratio of 1:1.25 could be achieved. The Scholz counter allows measurements in a wide range of CN concentrations from 5 to 960 000 particles in cm^3 . We have not found any information on the experimental error, however, it should not be higher than the experimental error of the Aitken counter which is about 10%. The Scholz counter was used by the end of 1982 and after which a photoelectric counter was used as a more convenient (automatic) replacement. It was based on the construction of a Verzář counter which had the condensation chamber of 680 cm^3 in the volume.

(2) The conductivity model used here

Section 6 describes a conductivity model, but without sufficient details to thoroughly check its viability and origin. Equation (1) represents this model, but this is not an equation found in Tinsley and Zhou (2006). It may be an equation taken from Israel's text, but that is not identified. I for one do not recognize equation (1) from available references, though the inverse relationship between conductivity and N is reasonable. In addition, all parameters used here should be properly quantified and justified. One piece of evidence that this conductivity model is not working properly (even if equation (1) is taken at face value) is a simple check on Ohm's Law and air-earth current. One need only check equation (2) numerically (though it should be born in mind that the GEC air-earth current may vary annually). For winter, a value $\sigma = 2.28 \times 10^{-15}$ and $E = 370 \text{ V/m}$, $J = 0.84 \text{ pA}$, too small by at least a factor of two. For summer, $\sigma = 1.76 \times 10^{-15}$ and $E = 370 \text{ V/m}$, $J = 0.65 \text{ pA}$, and so too small by a factor greater than three. The evidence here is that the conductivity model is giving too small a conductivity, and that inference is backed up by the large values of N coming out of the CN counter (with values per cc larger than ones typically reported in the literature, even for cities, see Chalmers (1967). The authors should make these points and these calculations. They also need to take a careful look at their conductivity model.

Reply: We apologise for giving the wrong reference to Eq. 1. Tinsley and Zhou (2006) in their Eq. 3 give a differential equation, the solution of which is our Eq. 1. This equation can also be found in Schonland (1932), Makino and Ogawa (1985), among others. The input parameters for the conductivity model are also taken from other published models of the air electrical conductivity (which use the same Eq. 1), such as Makino and Ogawa (1985), Sapkota and Varshneya (1990), Tinsley and Zhou (2006), Kulkarni (2022). Every parameter used is based on empirical models (e.g. ion production) or other experimental data. The ion mobility of $1.5 \text{ cm}^2 \text{ s}^{-1} \text{ V}^{-1}$ seems a realistic value for both positive and negative atmospheric ions at altitudes of up to 15 km according to Swider (1988). The ion recombination coefficient of $1.4 \times 10^{-6} \text{ cm}^3 \text{ s}^{-1}$ was used by Kulkarni (2022), similarly to Makino and Ogawa (1985). Ion production by cosmic rays of the order of $1.0 \text{ s}^{-1} \text{ cm}^{-3}$ is appropriate for the production at the ground level, and of the order of $1.0 \text{ s}^{-1} \text{ cm}^{-3}$ for the production by radioactivity from radon ($8.6 \text{ s}^{-1} \text{ cm}^{-3}$ in Makino and Ogawa (1985) over land). In the correction we would like to update the ion production value to $2.2 \text{ s}^{-1} \text{ cm}^{-3}$ which gives more realistic values of the conductivity at Świder. We also need to correct ion recombination coefficient α since our calculations were made with $\alpha = 1.3 \times 10^{-7} \text{ cm}^3 \text{ s}^{-1}$. We leave the values of the beta coefficient of ion attachment rate calculated following the method of Tinsley and Zhou (2006) which uses the aerosol parameters of Hess et al. (1998). Hess et al. (1998) use constant model distributions of the aerosol constituents (soot, water soluble, insoluble and others) while in reality these distributions may also vary depending on seasons and day. In general, it is still a simple conductivity model which still needs to rely on several input parameters for various processes present such as the ion production, the rates of ion recombination and ion attachment to aerosol particles. For these parameters we use representative values used in other conductivity models while in reality these values may vary over a wider range, and in fact to have realistic model of local conductivity we need more observations to

derive the required parameters. We modified further the model as compared to our first reply.

(3) The arbitrary CN threshold of 10,000 per cc

This threshold in CN is mentioned repeatedly (lines 8, 45, 99, 103, 111, in captions for Figures 7 and 8, lines 177, 341 and 347), but it is not made clear why this value was selected. No reference is given for justification. The suggestion is that the authors are seeking a characterization of the medium in cleaner conditions, but again the best parameter for that purpose is the Gerdien-measured conductivity, since this quantity is dominated by small ions with mobilities orders of magnitude larger than those for large ions/CN). In the end, the selection of this threshold is not resolving the main troublement at present (lines 13-14 of the Abstract and lines 360-362 of the Conclusions).

Reply: We have given additional references (Landsberg 1938, Schonland 1953, Mohnen and Hidy 2010).

(4) The main troublement of the paper

The authors' main troublement, linked directly to the important interest in the annual variation in the source of the global electrical circuit, is that the seasonal variation in N (and conductivity inferred from the model that is N-dependent) is quite small in comparison with the potential gradient, leaving the impression that NH winter is still dominating the DC global circuit, as Lord Kelvin had inferred (probably incorrectly) more than a century ago. Based on the weight of the evidence now available, something is wrong with using the CN measurements to infer the true air conductivity.

Reply: Yes, the interest in the annual variation of the global electrical circuit was for us an important issue and one for the purpose of which we carried out the investigation, however, in the first place we wanted to investigate whether, having a long-term series of PG and CN measurements we would be able select the conditions of low aerosol concentration, and whether this could reveal a different character of the annual variation of the PG. It did not, however, since the PG still had a maximum over NH winter months, and we never said that corresponded to the annual maximum of the GEC. Perhaps we have not emphasised this conclusion of our paper enough. We agree that using only 3 hours from each day may not represent total seasonal or diurnal variation but it does represent some characteristic points that capture the variation of CN during a day (Fig. 6).

(5) The suggested resolution of the troublement

The good news here is that long-term Gerdien tube measurements of the air conductivity are also available at Swider and have been presented in earlier unreferenced work by Kubicki et al. (2007), in a work that is cited in the present manuscript (page 2) but not elaborated on in the present context. The air conductivity is always dominated by the small ions (whose mobilities are orders of magnitude larger because of their small size, but these are not the ions measured with typical CN counters). Figure 1b of Kubicki et al. shows that the wintertime conductivity of air is reduced by a factor of two in comparison with summer, and is inverse with the measured seasonal change in potential gradient, making the seasonal variation in air-earth current much less than either conductivity or PG. The most important point here is that the seasonal change in air conductivity is MUCH LARGER than the variation of CN, the main source of the authors' troublement.

One last aspect of Kubicki et al. (2016) that also has relevance is Figure 1a, showing that the seasonal variation of dust exceeds a factor of 5, and so shows the largest seasonal variation of all. It is

conceivable that the dust (however it is measured, and this is not explained in this brief abstract) is dominating the removal of small ions over the seasonal cycle. One must also be aware however (from Figure 6 of the reviewed work) that the true seasonal variation of CN is not captured by the 3-hour per day sampling of CN, a serious shortcoming.

Reply: The conductivity measurements have been carried at Świder, however the observatory yearbooks report only the value of the positive conductivity. By now we have digitised the whole 1956-2005 series and are able to present some analysis of the data. The annual variation of the conductivity confirms earlier results of Kubicki et al. with a minimum in NH winter months and twice higher values in the summer. There is also a distinct effect of conductivity to lowering the affecting CN concentrations but there is no very clear seasonal effect.

Summary: When air conductivity measurements at Świder are considered, and the authors can surely do that in a revised submission, the main troublement of the manuscript is removed. (This finding would not conflict with the Kubicki et al findings because the seasonal issue was not focused on in that work.) The revised findings would then support the general conclusions of Adlerman and Williams (1996) that the wintertime maximum in potential gradient is caused by the enhanced pollution expected in wintertime at extratropical locations such as Poland and the UK of Lord Kelvin.

Reply: Thank you for the detailed consideration given to our manuscript. We added results including an analysis of the conductivity for the purpose of investigating its annual variation and conclusions for the GEC annual variation. We concluded the conditions of low condensation nuclei conditions does not change the character of the annual variation of the PG at Świder.

Response to Reviewer #2

The paper contains a reinterpretation of historic data to investigate the relationship between aerosol concentration and atmospheric potential gradient. Many investigations similar to this have used proxies or estimates for aerosol concentrations, so the inclusion of real aerosol data makes this a useful addition to the literature.

In my view, the manuscript could aid the reader more by adding more technical information on the measurements and some additional statistical analysis. The site description may be included in the references, but it would benefit the manuscript to put pertinent information on the measurement site and places of interest nearby. For example, how near is the measurement site to major population centers, roads, the coast or industry. This would aid in understanding potential sources of aerosol.

Reply: Indeed, we could add additional information about the measurement site. The PAS Geophysical Observatory in Świder is located in the central part of Poland in the Warsaw suburban area, about 25 km south-east. Świder used to be a popular holiday and health resort village located on the Świder river. The distance to the nearest urban centre, which is the district town of Otwock, is 2.5 km. There is no major industry in the area but there are local anthropogenic sources of air pollution from heating, very typical for these suburban conditions. The architecture is dominated by residential buildings and mainly includes single and multifamily houses. The Observatory is located in a less populated area nearer the river and covers about 7 ha. It includes the main office and three observation pavilions, two residential buildings are in a distance. The entire area is surrounded by trees (predominantly pine trees), with several clearings. In one of these clearings of an area of

approximately 1 ha, one pavilion and the station's instruments for atmospheric electricity and meteorology observations are located.

The measurement instrumentation is listed but more detail on the instruments used would also be helpful. Of particular importance is the measurement range, sensitivity and error of the aerosol measurement. I believe more detail of the aerosol instrumentation needs to be repeated within the manuscript, as the references are not easily available. Please could you include details of what kind of aerosol counter is used, the size range that are counted and details of inlet tubing.

Reply: We admit the information on the details of the apparatus is scarce as it is also very scarce in the observatory yearbooks, especially in regard to the technical details of the counters. We would like to add more details about the measuring apparatus which we found in the literature, in the archive materials of the Institute and from the observatory staff. We also add some details of the methodology and schedule of observations.

Measurements of concentration of condensation nuclei (CN) in Geophysical Observatory in Świder between 1965 and 2005 were carried out using two counters: a small Scholz counter and a photoelectric CN counter built in the observatory and using chamber of a Verzár counter as a base. The measurement method used in both counters was the process of condensation of water vapor on atmospheric aerosol particles present in the measurement chamber, followed by a quantitative analysis of the resulting mist droplets. We intend to add literature references where these counters are described in more detail, e.g. McMurtry (2000). Observations were performed three times a day between 5:50 GMT and 6:20 GMT (till 1971), 6:10 GMT and 6:30 GMT (afterwards), then between 11:00 GMT and 11:30 GMT, and between 19:00 GMT and 19:30 GMT (till 1971), 18:10 UT – 18:30 UT afterwards. These three periods of observations are referred to in the yearbooks as 06, 12, and 18 GMT or UT, respectively.

At Świder, the main part of the small Scholz counter was a brass cylindrical chamber with a volume of 102 cm³ and a height of 4 cm, and the adiabatic expansion ratio of 1:1.25 could be achieved. Measurements were performed within the clearing of the meteorological station using the suction method, an air sample with a volume of 1 cm³ was taken at a height of 1 m above the ground surface. Measurements with a Scholz counter should be repeated a few times, and one measurement takes several minutes. The Scholz counter allows measurements in a wide range of CN concentrations from 5 to 960 000 particles in cm³. We have not found any information on the experimental error, however, it should not be higher than the experimental error of the Aitken counter which is about 10%.

Since January 1983, the measurements have been performed with the photoelectric CN counter that was placed inside a measurement pavilion. The air samples were collected from the outside of the building at a height of 1 m above the ground. The suction of air was made through a 1 m long rubber pipe using an electric rotational pump. According to the yearbooks the counter enabled measurement of the concentration of CN whose radius ranged from 0.005 to 10 µm. The measuring range of the counter was 4500 to 850 000 CN in 1 cm³ of the air. The basic measurement of the number of CN took place in a cylindrical chamber filled with the tested air sample of the volume equal to 680 cm³. Estimates of the number of droplets were obtained using a photoelectric counter system by measuring the extinction of light. The electronic circuit system was built (also patented) by Stanisław Warzecha. The measurement accuracy was 15%.

In section 2.3, who assessed the criteria for fair weather, was this done by observatory staff, or more recently?

The criteria for fair weather were assessed by the observatory staff on an ongoing basis. The information is also included in the yearbooks.

Finally, I would suggest that the paper could be strengthened by adding some more statistical testing to the analysis. For example, it is claimed that limiting the condensation nuclei concentrations does not significantly change the distributions of corresponding potential gradient values – was a test used to prove the difference is not significant? When comparing the shape of distributions, could you consider using a non-parametric distribution test, eg, Kolmogorov-Smirnov test?

Reply: Thank you for pointing this out. We have tried to support our conclusions by using a non-parametric Kolmogorov-Smirnov (KS) and U Mann-Whitney (MW) tests. They have been used to calculate the statistical significance of differences between the PG distributions considering the decreasing number of CN. In regard to the distributions shown in Fig. 2 and in Fig. 7, both KS and MW tests indicated statistically significant differences (at the significance level of 0.05) between the PG-CN all and PG-CN<10000 distributions for the whole year as well as in the spring, autumn and winter. In case of the variations shown in Fig. 11, the KS test indicated statistically significant differences between the PG populations for all CNs and CN<10000, in January and February only. The MW test indicated such in April and November as well. When comparing PG and CN all with PG and CN<8000 the results of both tests indicate the statistically significant difference also in December, similarly to PG and CN<6000.

A few small corrections:

Section 2.1, there was a change if instrumentation, was any cross checking done to verify if there was a change in response?

Reply: We don't know exactly if any cross-checking has been done. In the yearbooks there is no information about it.

In Figure 1, is there any exclusion criteria before a data point can be included, for example, is there a minimum number of fair weather data points required for the month to be included?

Reply: No criterion regarding the minimum number of points for the months has been applied. Our intention was to use all the possible hourly values obtained during fair weather conditions.

In Figure 2 (and some subsequent figures) the values separated by commas are not clear which plot they belong too and this should be made explicit.

Reply: We added more description in the figures captions.

In Figure 6 can you clarify how long the measurement is that the means at 6h, 12h and 18h are?

Reply: As we mentioned, a single measurement lasted several minutes (about 5 min), and for any observation usually several single measurements were carried out and the final result was the average value calculated from these several measurements.

Line 47: in the statement 'less than 10,000' could you say what size these particle fall under?

Reply: The above statement refers to the number of CN below 10000 cm⁻³, we can suggest that there are particles from the whole measuring range (from 0.005 μm to 10 μm).

Line 228: What is the justification for the values given for the variable in equation 1, are they based on measured quantities?

Reply: Our intention was to use representative values of the necessary parameters, several of which are required for even such a simple conductivity model. Parameters of such order are used by various conductivity models which use values based on observational data or empirical models. These are the models of Makino and Ogawa (JGR 1985), Sapkota and Varshneya (1990), Tinsley and Zhou (2006), or Kulkarni (2022). Ion production by cosmic rays of the order of 1.0 s⁻¹cm⁻³ is appropriate for the production at the ground level, and of the order of 1.0 s⁻¹cm⁻³ for the production by radioactivity of radon. In the correction we would like to update the ion production value to 2.2 s⁻¹cm⁻³ which gives more realistic values of the conductivity. The ion mobility of 1.5 cm² s⁻¹ V⁻¹ seems a realistic value for both positive and negative atmospheric ions at altitudes of up to 15 km according to Swider (1988).

Line 230: What is the justification in assuming both mobilities are equal, as they are often not different to each other. Is it possible to run a sensitivity analysis to see what a difference would make?

Reply: As mentioned above the same value of (1.5 ± 0.3) cm² s⁻¹ V⁻¹ could be used for both positive and negative ions. A change of the mobility by 0.1 cm² s⁻¹ causes a change in the conductivity by about 5 × 10⁻¹⁷ S/m (5%) which is rather small in comparison with the effect of other parameters considered.

Line 242: Please provide more information on where and when these size distributions were measured.

The information is taken from Kubicki et al. (2016), and the distributions were measured at the Świder Observatory after 2010. The data have not been published, however.

Line 254: Why was relative humidity excluded?

Reply: The effect of humidity would complicate the conductivity model even more, and we are unable at the moment to use a practical conductivity model that takes it into account. The aerosol concentration distribution parameters of Hess et al. (1998) used for the calculation of the ion attachment rate are given for the relative humidity of 50%. We consider it is appropriate for average fair weather conditions, even though observational values may cover a wider range.

Some smaller changes:

Line 133, change "except of" to "except in"

Line 176, superscript should be -3

Line 273: Should this read 1.3 times by?

Reply: Thank you for the detailed consideration given to our manuscript. We have corrected the mistakes.