# Regular fluctuations of surface ozone at Georg-von-Neumayer station, Antarctica

By P. WINKLER, Deutscher Wetterdienst, Meteorologisches Observatorium Hamburg, Frahmredder 95, D-2000 Hamburg 65, Germany, S. BRYLKA, Alfred-Wegener-Institut für Polar und Meeresforschung, Columbusstraβe, D-2850 Bremerhaven, Germany and D. WAGENBACH, Institut für Umweltphysik der Universität Heidelberg, Im Neuenheimer Feld 366, D-6900 Heidelberg, Germany

(Manuscript received 15 April 1991; in final form 17 July 1991)

#### ABSTRACT

It is shown, that surface ozone at the Georg-von-Neumayer (GvN) station in Antarctica exhibits long frequency fluctuations with periods at 30, 18, and 14 days. In some years, these periods are also visible in the temperature. Periods of 25–30 days have been established for antarctic radon-222 in the literature in several investigations, however, radon-222 measurements at GvN do not show the 30-day period which may be suppressed there by the meteorological situation. A hypothesis is proposed, explaining how the periodic fluctuations of ozone and of radon-222 at GvN and other sites could be caused by long-range transport phenomena. The mechanism which induces the periodic fluctuations remains unknown.

#### 1. Introduction

The antarctic winter is known as a "coreless winter" which means that the temperature does not fall below a certain value although the radiation budget remains negative (Schwerdtfeger, 1984). The reason for this phenomenon is the more or less continuous transport of heat into the continent due to the activity of lows. Normally one would assume that the storm and heat transport follows an irregular pattern, however, Schwerdtfeger already stated, that the yearly course of the temperature shows low-frequency fluctuations with a period of about 30 days during winter.

Surface ozone at the coastal Georg-von-Neumayer station (70°37′S, 8°22′W) has been measured since 1982 with the wet chemical KJ method (Attmannspacher, 1971). In view of the state of knowledge to day, the aim of this study was to find out whether ozone also shows regular fluctuations of the same period since it is also subject to transport and not produced locally. Ozone in the remote troposphere is of mainly stratospheric origin and it is destroyed either in clouds (Lilieveld and Crutzen, 1990) or at the

earth surface. Destruction on the snow surface appears to be negligible in comparison to other sinks (Galbally and Roy, 1980). Especially in winter, when the Antarctic Ocean is ice covered, advection should dominate the local ozone budget.

In 1970, Lambert et al. investigated the frequency spectrum of radon-222 (222Rn) measured at Dumont d'Urville and found a pronounced maximum at 28-day intervals but they could not explain this behavior clearly. A similar periodicity for <sup>222</sup>Rn has been described by Pereira (1990) for a series measured at Ferraz at the Antarctic Peninsula. He established a period of 25 days for the 1986 data and of 30 days for the 1987 data, but could not give an explanation either. <sup>222</sup>Rn mainly originates from the emanation on continents. A minor oceanic source also exists which is 2-3 orders of magnitude smaller (Peng et al., 1979). The evasion at marine <sup>222</sup>Rn, however, constitutes substantially to the mean atmosphere <sup>222</sup>Rn level at subantarctic sites (Heimann et al., 1990) whereas the ice covered antarctic continent is not important. The periodicity of the dominant <sup>222</sup>Rn peaks at antarctic stations should therefore be caused by a periodic advection of air which had recent contact with continents.

In this paper, we try to find out whether O<sub>3</sub> at Georg-von-Neumayer also shows periodic fluctuations as was established for <sup>222</sup>Rn. Such a behavior is to be expected especially during the austral winter when in absence of radiation photochemical production or destruction in the troposphere should be minimal (Liu et al., 1987) and fluctuations should be caused by transport phenomena. Temperature and <sup>222</sup>Rn which have

been measured contemporarily will be used for confirming the results obtained for ozone.

## 2. Calculation of spectra

Daily mean values of surface ozone at GvNstation in Antarctica were investigated: (1) 13 March-23 November 1983 and (2) 17 March-

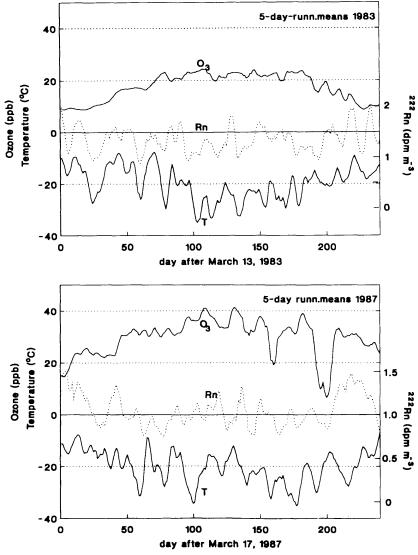


Fig. 1. Time series of 5-day running means of the concentration of surface ozone, temperature and radon at Georg-von-Neumayer station during 1983 (upper part) and 1987 (lower part). For the Rn-record, a linear trend of  $-0.0006 \,\mathrm{dpm/m^3}$  (1983) and of  $-0.0013 \,\mathrm{dpm/m^3}$  (1987) times day number has been removed.

27 November 1987. These two time periods were selected because the data sets were complete. We avoided the use of summer data because own unpublished studies as well as those of Barrie et al. (1988) indicate a photochemical destruction of ozone during the sunlit period, at least in polar latitudes. The detection of periodic fluctuations,

although they may be present also during summer, would have been made difficult by using such data. Fig. 1 shows 5-days-running mean values of ozone for the two years and the selected time periods. One can see that the short-term fluctuations of ozone are smaller and the development with time is smoother in 1983 as compared with 1987. It

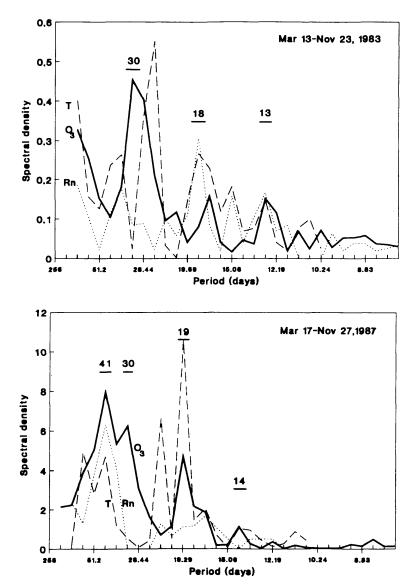


Fig. 2. Periodograms of ozone concentration, temperature and  $^{222}$ Rn as depicted in Fig. 1. The spectral density (y-axis) is the Fourier transformate of the auto-correlation function, which corresponds to the squared amplitude of the sinusoids. When a maximum appears the corresponding period has a higher frequency than neighboured periods. Upper part: 1983 data, lower part: 1987 data. The maxima at 40, 30, 18 and 13 days are marked by the heavy line.

may be concluded that a different meteorological development occurred in the two years, which was the main reason for the selection of the two time periods. The identification of any periodic fluctuations in meteorologically different years is an important hint for their general occurrence. In Fig. 1, 5-days-running means of temperature (2 m) (AWI, 1990, private communication) and Radon are shown for comparison. The <sup>222</sup>Rn record has been derived from the  $\alpha$ -activity of short lived decay products attached to aerosol particles. Here, a secondary equilibrium is assumed between radon and its products. This equilibrium may not have been reached during times with intensive scavenging, i.e., heavy snowfall or heavy snow drift, resulting in an underestimation of 222Rn. A similar situation can arise during extremely clean situations leading to a significant fraction of unattached Rn-decay products. As far as such artificial minima originating from the measurement method of <sup>222</sup>Rn have been recognized, they were eliminated and replaced by interpolated values (definitely 5 values in both periods). Thus, artificial periods in the subsequent Fourier analysis will be avoided.

The power spectra were calculated using the Fast Fourier Transformation (FFT) (Press et al., 1986). In order to avoid a jump introduced by unsteady transitions at the beginning and endpoints of the periodically repeated series, the linear regression was calculated for the selected period and the linear trend subtracted. In order to eliminate short-term fluctuations, 5-day moving averages were calculated.

In Fig. 2, the periodograms of ozone, temperature and <sup>222</sup>Rn are plotted for the two time periods. Ozone shows maxima at 30 and 18 days for the first and at slightly over 40, 30, and 18 days for the second period. Less pronounced maxima occur around 13 and 14 days, respectively. Fig. 2 also shows the spectra for temperature and <sup>222</sup>Rn. It is noteworthy that the 18- and 13-day period are also visible for these two parameters. In 1983 a peak at 26 days is found for the temperature which is slightly shorter than that of ozone, while <sup>222</sup>Rn shows a less pronounced peak at around 40 days, coinciding with another small peak in the temperature spectrum. A 30-day period is missing for <sup>222</sup>Rn. In 1987, the 19 day peak is very prominent in the temperature spectrum but less pronounced for <sup>222</sup>Rn. At a period of 41 days all three parameters show marked peaks. Only ozone shows another secondary peak at again 30 days. The temperature shows another peak at 23 days again coinciding with a small <sup>222</sup>Rn maximum and an ozone minimum.

When cross-correlation spectra (not depicted) are calculated, the coherence is best pronounced at around 18–19 days and 13–14 days. At longer periods coherence maxima appear around 28–30 days in 1983 and around 44 days in 1987. It may be important to emphasize that maxima of different parameters at a similar period appear to be more essential for our consideration than the absolute height of the peaks in the power spectra.

#### 3. Discussion

Ozone at Georg-von-Neumayer station seems to have a similar long frequency period of around 28–30 days as found for <sup>222</sup>Rn at other sites, at least in some years. In other years, the period may be longer. Periods of this length have also been reported for several islands in the Indian Ocean for <sup>222</sup>Rn (Balkanski and Jacob, 1990), for Ferraz (Pereira, 1990), and Dumont d'Urville (Lambert et al., 1970). In the following, we try to investigate meteorological large scale transport mechanisms which simultaneously influence ozone, <sup>222</sup>Rn, and temperature.

With respect to the general circulation and long range transport there are several interesting aspects which should be considered:

(1) As mentioned in the introduction, <sup>222</sup>Rn above average concentrations in Antarctica originate mainly from the continents. Lambert et al. (1990) showed that <sup>210</sup>Pb, a long-lived decay product of <sup>222</sup>Rn, and the parent substance <sup>222</sup>Rn have similar atmospheric lifetimes (7 and 5.5 days, respectively) and thus undergo similar depletion during transport. However, following Lambert et al. (1990), a pronounced minimum of the concentration of <sup>210</sup>Pb is obvious at 50°-60°S with higher concentrations north and south of that latitude. It must be concluded that the transport of <sup>210</sup>Pb and consequently <sup>222</sup>Rn does not proceed along the surface but rather goes through the upper troposphere. Indeed, the modelling of longrange transport of <sup>222</sup>Rn (Heimann et al., 1990) shows concentration maxima in a simulated zonalheight cross section between 900 and 500 hPa at 43° and 51°S. Convection above the continents transports the radon-rich air rapidly from the boundary layer to upper levels, where transport is more efficient. These levels then reach higher <sup>222</sup>Rn concentrations than the air in the marine boundary layer (Feichter and Crutzen, 1990). Considering the large scale circulation of air around and into Antarctica it must be established that the air enters the continent in upper levels, descends downwards above the pole and leaves the content in lower altitudes (Wexler et al., 1960; Bromwich, 1978). Apart from climatological differences in the aerosol residence time, this largescale exchange mechanism may explain why <sup>210</sup>Pb reaches higher concentrations at Antarctica than in the boundary layer of the surrounding ocean.

- (2) Ozone does not have a dominant continental source like 222Rn if we disregard the ozone production from biomass burning, which is confined mainly to the dry months of October and November and tropical latitudes. In the pristing atmosphere tropospheric ozone stems from the stratosphere due to stratospheric-tropospheric exchange. Maxima of this exchange were derived for 20°S, 38°S and to a minor degree for 70°S (Fabian and Pruchniewicz, 1977) from ozone observations. Arpe et al. (1986) calculated vertical and meridional cross sections of the eddy kinetic energy, which in the tropopause level should be proportional to tropospheric-stratospheric exchange. The highest contribution originates from eddies with wave numbers between 4 and 9 at a latitude between 45° and 50°S. Their result was obtained by integration over real meteorological fields with the model of the European Center for Medium Range Weather Forecasts over one year. Thus, elevated concentrations of ozone at GvN must be caused by weather pattern where an increase in stratospheric-tropospheric exchange occurs in midlatitudes. When ozone fluctuates at GvN, such weather pattern must occur also regularly.
- (3) As mentioned above, <sup>222</sup>Rn and O<sub>3</sub> have different sources and sinks. Nevertheless, it seems likely that the periodic fluctuations of <sup>222</sup>Rn and O<sub>3</sub> are caused by the same large scale meteorological phenomenon. Kidson (1986) investigated variations of the zonal wind in the southern hemisphere and found a period of 27 days for the winter 1979. He also showed that the winds in the

500 hPa level at the latitudes 48.75 and 67.5°S varied opposingly. He defined an index based on the wind difference in these latitudes and deduced that during low index situations a blocking high is favored near and to the east of New Zealand while during high index situations the high in the South Pacific is well developed. This demonstrates that periodic changes of favored states of the circulation take place and that these periodic fluctuations may induce periodic transport of air of continental origin to Antarctica and that during this transport ozone is also enriched. As we have already seen, the transport is predominantly expected to go via the upper troposphere and during meteorological situation stratospherictropospheric exchange must be stimulated leading to the enrichment of ozone. We cannot decide whether an air mass with high 222Rn is enriched with O<sub>3</sub> by this exchange process or whether the exchange is stimulated by a transport pattern with rapid meridional air mass exchange. In the first case both tracers would arrive in phase, in the second case out of phase in Antarctica. We will discuss that point further under item (5).

Kidson (1986) also mentioned a period of 16–18 days in zonal wind variations which we also found at GvN for ozone and temperature and, less pronounced, for <sup>222</sup>Rn. A small maximum in Pereiras (1990) periodogram for <sup>222</sup>Rn was observed at 16–18 days for the 1987 but not for the 1986 data. Thus this oscillation in the circulation may only sometimes be strong enough to cause long range transport of <sup>222</sup>Rn but seems to influence ozone more regularly.

(4) Taking into account that 222Rn and O3 have a ground- and a stratospheric source, respectively, and considering the fact that at GvN-station the temperature and ozone show oscillations with similar periods, we can test our hypothesis, that the periodic transport of <sup>222</sup>Rn and O<sub>3</sub> is caused by the same meteorological phenomenon, by a correlation between temperature and ozone. Since 222Rn in Antarctica originates from low latitudes (i.e., S-America or S-Africa) it should correlate with warm air masses, i.e. comparably high temperatures. Ozone, in contrast, with its stratospheric source, should correlate with low temperatures because, as is known from investigations on stratospherictropospheric exchange (e.g., WMO, 1988), surface 38 P. WINKLER ET AL.

ozone increases behind cold fronts. Thus we expect an anticorrelation between ozone and temperature which is confirmed by Fig. 3, derived from data of the GvN-station. We have only used data of the winter months (April-September 1982–1986) to avoid a masking by the yearly cycles of both parameters. Another test by a correlation between temperature and ozone in which we used deviations of both parameter from their yearly cycle confirms this result although the correlation coefficients for both years are not very high.

(5) A correlation between 222Rn and temperature should be positive as it must be expected that air masses with temperatures above average originate from low latitudes and this transport advects <sup>222</sup>Rn enriched air as well. This can only be confirmed with the data of 1983 but not of 1987. Several reasons may be responsible for this unexpected behavior in 1987: (a) The temperature in a flat surface layer is sometimes strongly influenced by outgoing longwave radiation. Thus the 2m-temperature may sometimes not be representative enough to identify air masses of subtropical origin. (b) In occluding lows, the warm (and radon-enriched) air arrives only at higher levels but not at the surface, while ozone may be elevated near the ground because the cold airmass advects at the surface. This situation may be more frequent at stations in high latitudes like GvN than in lower

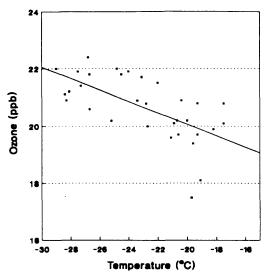


Fig. 3. Scattergram between 2 m-temperature and ozone at Georg-von-Neumayer-Station (average values for 10-sectors of wind direction).

latitudes like islands in the subantarctic ocean or at the antarctic peninsula. (c) The oceanic <sup>222</sup>Rn source contributes significantly at GvN during stormy situations where the ocean is stirred intensively stimulating gas exhalation. Thus, not any <sup>222</sup>Rn maximum must originate from continents, but could be caused by enhanced oceanic exhalation. During winter when the ocean is ice covered, such disturbances are not expected to occur in the direct neighborhood of GvN-station.

(6) In the literature two kinds of reasons have been discussed as responsible for the production of periodic fluctuations: pure terrestrial origin; astrophysical origin.

Balkanski and Jacob (1990), investigating the transport of continental air to the subantarctic Indian Ocean by means of a general circulated model, found a period of 28 days for <sup>222</sup>Rn at 3 islands. The authors claim that this period is produced by the model and therefore cannot the triggered by astrophysical mechanisms. However, a GCM also approximates nature only to some degree and it is not known, whether any periodicity is an artifact of the model.

An astrophysical influence was proposed by Lambert et al. (1970). They assumed that the synodic sun rotation (27 days) would influence the circulation not only in the high atmosphere but also in the troposphere. It is important to note in this context that the power spectrum of the radiation output of the sun shows a maximum at 23.4 days (Fröhlich, 1987) and not 27 days. This indeed excludes the sun as the direct origin for a periodicity in tropospheric air mass transport. It has also been shown by Xu (1990) that no relations exist between the stratospheric QBO and the tropospheric Southern Oscillation, although both have periods of 26 months, which also excludes a sun influence.

#### 4. Conclusions

Surface ozone and temperature at the Georg-von-Neumayer station in Antarctica show periodic fluctuations at 30–40, 18, and 13 days. It is likely that this result is not a randomly one, because these periods occur in two independent years. <sup>222</sup>Rn exhibits similar periods only in one year but not in the other at GvN station, while periods of

similar duration have been established by various authors (Lambert et al., 1970; Pereira, 1990). It is most likely that all the three parameters ozone, temperature, and 222Rn are influenced by periodically occurring large-scale phenomena, during which <sup>222</sup>Rn is advected from the continents simultaneously with warm air masses. Ozone is enriched due to stratospheric-tropospheric exchange which must be stimulated by the same large-scale transport process. Ozone arrives at GvN station together with cold air masses, i.e., with a certain phase shift to <sup>222</sup>Rn or temperature. It has been shown by Balkanski and Jacob (1990), that the development of a low at the tip of South Africa is important for the initiation of long-range transport of <sup>222</sup>Rn, and that this transport occurs periodically every 28 days.

From investigations of periodic fluctuations of a zonal index derived for 500 Hpa winds, Kidson (1986) stated that the fluctuations are restricted to a latitudinal belt between 48.7 and 67.5°S. Such a restriction may be a reason, why at GvN the 28 day period is less pronounced for <sup>222</sup>Rn than for ozone: GvN is located at the southern edge of this latitudinal belt and not each <sup>222</sup>Rn transport may reach the station. Additionally, radon-rich air

masses may arrive at the station frequently with occluding air masses only at higher levels while ozone arrives at ground level with cold air masses.

Although there is a large amount of evidence suggesting that large scale transport processes regularly occur in the SH and although tracers like <sup>222</sup>Rn and ozone provide signals for an independent confirmation of the periods derived from an investigation of the dynamics only, the true explanation for this periodicity is still unknown. Whether the position of the continents relative to the west wind belt stimulates a periodic formation of lows downstream of mountainous islands (e.g., New Zealand) or of South Africa, probably enhanced by a characteristic frequency of the atmosphere, has not been proved or disproved.

## 5. Acknowledgement

For the conduction of the measurements at the Georg-von-Neumayer-station in Antarctica we would like to express our gratitude to all winterers who have been active at the station. We would also like to thank to the Alfred-Wegener-Institut für Polar- und Meeresforschung for logistic support.

### REFERENCES

- Arpe, K., Brankovic, C., Oriol, E. and Speth, P. 1986. Variability in time and space of energetics from a long series of atmospheric data pronounced by ECMWF. Contr. Phys. Atm. 69, 321–356.
- Attmannspacher, W. 1971. A simple wet-chemical instrument with small inertia for measuring surface-near ozone in the atmosphere. (In German) *Meteorol. Rdsch.* 24, 183–188.
- Balkanski, Y. J. and Jacob, D. J. 1990. Transport of continental air to the subantarctic Indian Ocean. Tellus 42B, 62-75.
- Barrie, L. A., Bottenheim, J. W., Schnell, R. C., Crutzen, P. J. and Rasmussen, R. A. 1988. Ozone destruction and photochemical reactions at polar sunrise in the lower Arctic atmosphere. *Nature 334*, 138-141.
- Bromwich, D. H. 1978. Some considerations in deriving poleward water vapor transport values for coastal east Antarctica. A.J.U.S. 13, 196–197.
- Fabian, P. and Pruchniewicz, P. G. 1977. Meridional distribution of ozone in the troposphere and its seasonal variation. *J. Geophys. Res.* 82, 2063–2073.
- Feichter, J. and Crutzen, P. J. 1990. Parameterization of vertical tracer transport due to deep cumulus convec-

- tion in a global transport model and its evaluation with <sup>222</sup>Rn measurements. *Tellus 42B*, 100–117.
- Fröhlich, C. 1987. Variability of the solar "constant" on time scales of minutes to years. *J. Geophys. Res.* 92, 796–800.
- Galbally, I. E. and Roy, C. R. 1980. Destruction of ozone at the earth surface. Q.J.R. Meteor. Soc. 106, 599-620.
- Heimann, M., Monfray, P. and Polian, G. 1990. Modeling the long-range transport of <sup>222</sup>Rn to subantarctic and antarctic regions. *Tellus 42B*, 83–99.
- Kidson, J. W. 1986. Index cycles in the southern hemisphere during global weather experiment. Month. Weather Rev. 114, 1654-1662.
- Lambert, G., Polian, G. and Taupin, T. 1970. Existence of periodicity in radon concentrations and in the large-scale circulation at lower altitudes between 40° and 70° south. *J. Geophys. Res.* 75, 2341–2345.
- Lambert, G., Ardouin, B. and Sanak, J. 1990. Atmospheric transpsort of trace elements toward Antarctica. Tellus 42B, 76-82.
- Lilieveld, J. and Crutzen, P. 1990. Influences of cloud photochemical processes on tropospheric ozone. *Nature 343*, 227–233.

- Liu, S. C., Trainer, M., Fehsenfeld, F. C., Parrish, D. D.,
  Williams, E. J., Fahey, D. W., Hübler, G. and Murphy,
  P. C. 1987. Ozon production in rural troposphere and
  the implication for regional and global ozone distributions. J. Geophys. Res. 92, 4191-4207.
- Peng, T. H., Broecker, W. S., Mathieu, G. C. and Li, Y. H. 1979. Radon evasion rates in the Atlantic and Pacific Oceans as determined during the GEOSECS program. J. Geophys. Res. 84, 2471-2486.
- Pereira, E. B. 1990. Radon-222 time series measurements in the Antarctic peninsula (1986-1987). *Tellus 42B*, 39-45
- Press, W. H., Flannery, B. P., Teukolsky, S. A. and Vetterling, W. T. 1986. Numerical recipes, the art of scientific computing. Cambridge University Press.

- Schwerdtfeger, W. 1984. Weather and climate of the Antarctic. *Developments in Atmospheric Science 15*, Elsevier, Amsterdam, 28.
- Wexler, H., Moreland, W. B. and Weyant, W. S. 1960. A preliminary report on ozone observations at Little America, Antarctica. *Mon. Weath. Rev.* 88, 43-54.
- WMO 1988. Ozone 1985. Global ozone research and monitoring project. World Meteorological Organization. Report No. 16. Geneva, Switzerland.
- Xu, J. S. 1990. On the relationship between the stratospheric QBO and the tropospheric SO. Report No. 48 Max-Planck-Institute for Meteorology, Hamburg, Germany.