Correlation between Cosmic Rays and Ozone Depletion

Q.-B. Lu

Department of Physics and Astronomy, University of Waterloo, Waterloo, ON, N2L 3G1, Canada

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This Letter reports reliable satellite data in the period of 1980–2007 covering two full 11-yr cosmic ray (CR) cycles, clearly showing the correlation between CRs and ozone depletion, especially the polar ozone loss (hole) over Antarctica. The results provide strong evidence of the physical mechanism that the CR-driven electron-induced reaction of halogenated molecules plays the dominant role in causing the ozone hole. Moreover, this mechanism predicts one of the severest ozone losses in 2008–2009 and probably another large hole around 2019–2020, according to the 11-yr CR cycle.

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There is interest in studying the effects of galactic cosmic rays (CRs) on Earth's climate and environment, particularly on global cloud cover in low atmosphere (≤ 3 km) [1–5] and ozone depletion in the stratosphere [6–16]. The former has led to a different scenario for global warming, while the latter has provided an unrecognized mechanism for the formation of the O_3 hole. The discovery of the CR-cloud correlation by Svensmark and Friis-Christensen [1] has motivated the experiments to investigate the physical mechanism for the correlation [3–5]. In contrast, the CR-driven electron reaction mechanism for O_3 depletion was first unexpectedly revealed from laboratory measurements by Lu and Madey [6,7]. Then the evidence of the correlation between CRs, chlorofluorocarbon (CFC) dissociation, and O_3 loss was found from satellite data by Lu and Sanche [8]: the O_3 hole is exactly located in the polar stratosphere and at the altitude of ~ 18 km where the CR ionization shows a maximum. CRs are the only electron source in the stratosphere, while halogen (Cl, Br)-containing molecules are long known to have extremely large cross sections of dissociative attachments of low-energy electrons [9]. The latter reaction will be greatly enhanced when halogenated molecules are adsorbed or buried at the surfaces of polar molecular ice, relevant to polar stratospheric cloud (PSC) ice in the winter polar stratosphere, as firstly discovered by Lu and Madey [6,7] and subsequently confirmed by others in experiments and theoretical calculations [10–16]. For example, the dissociative attachment cross section at ~ 0 eV electrons for CF_2Cl_2 adsorbed on the surface of water ice has been measured to be $\sim 1 \times 10^{-14}$ cm^2, which is about 1×10^6 times the photolysis cross section of CF_2Cl_2 [6,10]. As many challenges as the CR-cloud model has received [4,5], however, the CR-related O_3 depletion mechanism has also been the subject of strong debate [17–23].

The current focus in debate is whether there exists a time correlation between CR intensity and O_3 loss over an 11-yr CR cycle [19–23]. NASA TOMS Satellite data showed a time correlation between the annual mean total O_3 at latitudes 0–65S and the CR intensity in the single CR cycle of 1981–1992 [8]. However, it has been argued that no such a correlation would exist beyond one CR cycle [19], or no correlation between CRs and O_3 loss in the polar region would exist [19,21,22]. It was thus suggested that no further studies of the CR-driven mechanism for O_3 depletion should be motivated [22]. However, the widely accepted photochemical model predicted that the total O_3 over 60°S to 60°N and the Antarctic springtime O_3 would recover (increase) by 1% to 2.5% and 5% to 10% between 2000 and 2020, respectively [24], which are clearly not consistent with the observed data. Moreover, recent findings have indicated large discrepancies between the photochemical model and the observed O_3 loss—at least 60% of O_3 loss at the poles seems now to be due to an "unknown" mechanism [25,26]. In fact, the large fluctuations of the O_3 data in Refs. [19,21,22] did not allow us to prove or disprove the CR-O_3 correlation, as demonstrated in Ref. [23]. A careful examination of this time correlation is therefore of critical significance to determine a correct mechanism for O_3 depletion. In this Letter, reliable CR and O_3 data covering two full 11-yr CR cycles up to 2007 are reported. These data have extended the observation of the time correlation between CRs and global O_3 depletion for *over two CR cycles*, and for the first time, the time correlation between CRs and total O_3 in the Antarctic area in springtime (October) and annually was observed. These results cannot be explained by the photochemical model; instead, they provide strong evidence of the CR-driven reaction as the dominant mechanism for causing the O_3 hole. Moreover, *new predictions* on future trends of the O_3 hole are given and are important to further test the proposed mechanism.

High-quality data of cosmic rays are available from measurements at several stations [27], while NASA TOMS and OMI satellite datasets have so far provided the most reliable and widely used data for global and polar-area total ozone since 1979 [28]. Figure 1 shows the long-term data for both annual mean CR intensity and total ozone of the southern hemisphere (0°–60° S) during 1980–2007, covering two full CR cycles. These

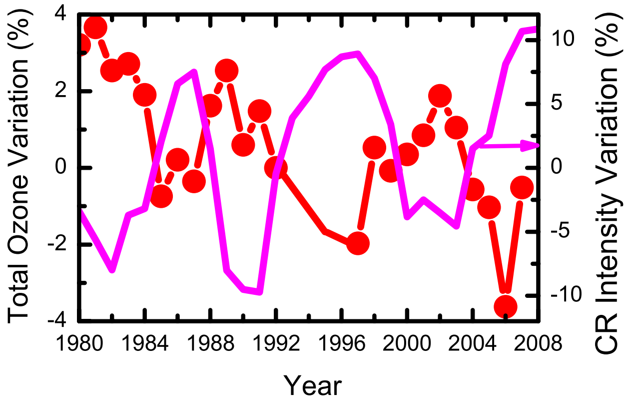

FIG. 1 (color online). Percentage variations of CR intensity and annual mean total ozone with latitudes 0–60° S during the period of 1980–2007. The CR data (solid line) were averaged from 3 station measurements (McMurdo, Thule, and Newark) of the Bartol Research Institute [27], while ozone data (solid circles + line) were obtained from NASA TOMS/OMI satellite datasets [28]. The ozone data were relative to the value at 1992.

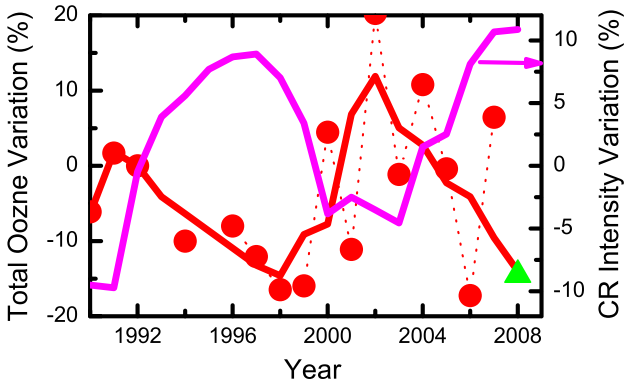

FIG. 2 (color online). Percentage variations of CR intensity and monthly mean total ozone in October Antarctic with latitudes of 60–90° S during the period of 1990–2007. The CR data are the same as in Fig. 1, while O_3 data (solid circles + line) were obtained from NASA TOMS/OMI satellite datasets with a nominal uncertainty of 1% [28], relative to the value at 1992. The 2002 O_3 data point is multiplied by a factor of 0.75 to fit in the plot; the solid line passing through the data points is a smoothed curve to aid the eyes. The total O_3 data for October 2008 (upper triangle) is a predicted value (see text).

high-quality (low-noise) data now clearly establish the correlation of O_3 loss with CR intensity variation: the total ozone is exactly in reverse phase with the CR intensity. This observation indeed provides most convincing evidence of the time correlation between CR intensity and global O_3 depletion.

Most interestingly, although the monthly mean O_3 data have generally much larger fluctuations than the annual mean data, the observed polar O_3 loss (hole) in October over Antarctica with latitudes of 60°–90° S in the period of 1990–2007, obtained from NASA TOMS and OMI datasets [28], also visibly shows a time correlation with the CR intensity, as shown in Fig. 2. A similar correlation with CRs can also be seen for the total ozone data in September or November (the hole period). In particular, the O_3 hole in 2002 is the smallest, corresponding to the CR-intensity minimum. Nevertheless, there are other effects such as atmospheric dynamics and meteorological conditions leading to the unusually small hole in 2002, as well reviewed in the literature [24]. In September 2002, a major stratospheric warming in the southern hemisphere split the polar vortex and O_3 hole for the first time in the record history. Atmospheric dynamics could also lead to short-term large fluctuations of total O_3 in the polar hole from year to year, but its effect on the long-term trend of O_3 loss is limited.

Furthermore, the monthly mean total O_3 data in the polar hole period (September or October) obtained from ground-based Antarctic stations [29] have large fluctuations by up to 20%[19,21,22]. This made it impossible to observe the time correlation between O_3 loss and CR-intensity variation (within 10%), as demonstrated recently [23]. However, the annual mean total O_3 data obtained from these stations should have much smaller fluctuations than the monthly data. This is indeed observed in Fig. 3, which again exhibits a clear correlation between CR intensity and annual mean total O_3 over Antarctica in the period of 1990–2007. It can be seen that the annual mean total O_3 in the polar region has an oscillation amplitude of about 5%. This value lies between the modulation amplitudes of $2 \sim 3\%$ for the annual mean total O_3 in the southern hemisphere if the mean decreasing trend is removed (Fig. 1) and of $\sim 12\%$ for the Antarctic O_3 hole in October (Fig. 2).

To obtain a quantitative statistical description of the correlation, the total O_3 variation is plotted versus the CR intensity, as shown in Fig. 4. The data obtained from

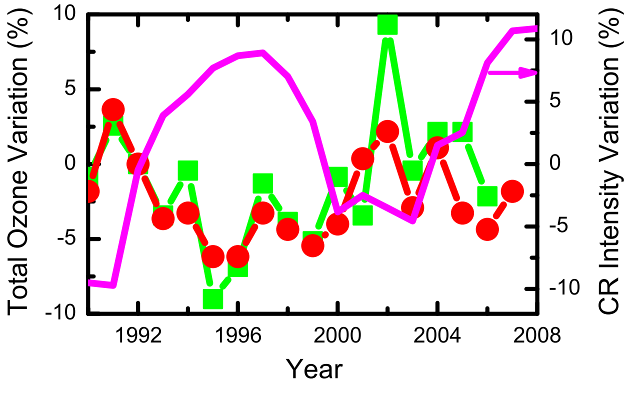

FIG. 3 (color online). Percentage variations of CR flux (solid line) and annual mean total O_3 (data points + lines) measured at two Antarctic stations, Faraday/Vernadsky (65°15′S, 64°16′W) and Halley (75°35′S, 26°36′W), during the period of 1990–2007. The CR data are the same as Fig. 1, while O_3 data (solid circles from Faraday station; solid squares from Halley) were obtained from the British Antarctic Survey [29], relative to the value at 1992. The data point at Halley in 2002 is multiplied by a factor of 0.7 to fit in the plot.

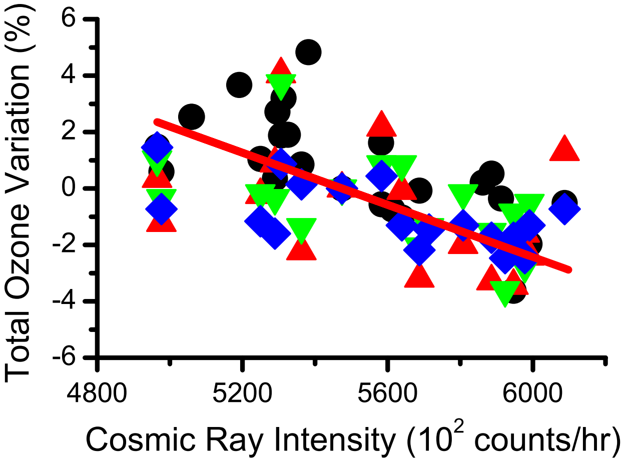

FIG. 4 (color online). Percentage variation of mean total ozone vs CR intensity with all the data from Figs. 1–3: solid circles for annual O_3 data with latitudes 0–60° S, upper triangles for October O_3 with latitudes 60–90° S, down triangles and diamonds for annual O_3 at Halley and Faraday/Vernadsky, respectively. To plot all these data together, the October and annual O_3 data for the polar region were reduced by factors of 5 and 2.5, respectively. The solid line is the best fit to the data, giving a linear equation (see text).

Figs. 1–3 indeed show that the total O_3 loss ($-\Delta(O_3)$) has a linear dependence on the CR intensity. This dependence can be quantitatively described by the best-fitted linear line, giving a linear equation: $-\Delta(O_3)/i \approx 25.3(1 - I/I_0)$. Here, $\Delta(O_3)$ is the percent variation of the total O_3 relative to the value at 1992 that corresponded to the average CR intensity $I_0 = 5474 \times 10^2$ counts/hr, I the CR intensity, and $i = 1$, 2.5, and 5 for the hemispheric annual mean total O_3, the annual and October mean total O_3 over Antarctica, respectively. This result shows that there is indeed a strong correlation between total O_3 variation and CR intensity, given the data dispersion.

The observed data in Figs. 1–4, especially the observed large oscillation amplitude (~12%) of the total ozone in the October Antarctic hole in an 11-yr cycle, cannot be explained by the so-called solar-cycle effect for the following reasons. First, the direct solar effect, in inverse phase with the CR effect, argued that when the solar UV radiation is stronger, more O_3 via the photolysis of O_2 would be formed in the upper stratosphere, so that the maximum O_3 level would occur at the maximum solar activity (i.e., the lowest CR intensity). This effect predicted small annual O_3 oscillations in the tropics and midlatitudes but not in the polar region and especially not in the springtime lower polar stratosphere where the O_3 hole is located [21]. Second, more remarkably, the solar-cycle effect should also predict that the higher solar intensity would lead to more destruction of CFCs and thus produce more active Cl to destroy O_3 [20]. Thus, the solar-cycle effect is ruled out for explaining the present data. In contrast, these data provide strong evidence of the CR mechanism [6,8]: the effect of CRs on ozone mainly occurs by stimulating the electron-induced reactions of halogen molecules on PSCs during the winter polar stratosphere, leading to the formation of the springtime O_3 hole. The appearance of the polar O_3 hole is known to have a consequent effect on annual total O_3 in the polar area and globally.

In the CR-driven mechanism, the O_3-depleting reactions depend on halogen concentrations, CR intensity, and PSC ice (to hold the electrons) in the stratosphere [6,8]. From 1992 up to now, the Antarctic O_3 loss has shown a clearest correlation with the CR intensity. This is because the total halogen amount of the stratosphere, particularly those of CFCs, is nearly constant in that period of time [30]; thus the regulating effect of CRs on O_3 loss becomes manifest. In contrast, such a time correlation is hardly seen in the enlarging spring polar O_3 loss during 1980s, since at that period of time, the halogen loading increased dramatically and thus ozone showed a drastic decreasing trend blurring the CR-O_3 loss correlation. And in the pre-1980s, no significant halogen loading was found in the stratosphere, and thus no significant O_3 loss was observed. This observation rules out not only the solar-cycle effect discussed above, but the possibility of the observed correlation being due to the mechanism of O_3 loss caused by the odd hydrogen (HO_x) and odd nitrogen (NO_y) species generated by cosmic rays in the polar stratosphere, similar to the production of NO_y species by solar particle events (SPEs) [24]. There may exist the other effects of charged particle precipitation on O_3 loss in the upper stratosphere [24]. However, SPEs and energetic electron precipitation are spontaneous frequent events without an 11-yr cycle, and that no significant effects of the recorded SPEs on total O_3 can be seen from the data in Figs. 1–3. Thus, it is also impossible to attribute the observed long-term oscillation in total O_3 with a periodicity of 11 years to SPEs and energetic electron precipitation.

One might argue that a simply time correlation does not guarantee a physical mechanism. However, there exist the following facts. (1) As discussed above, there is no alternative mechanism for the observed time correlation between polar ozone loss and CR intensity, which cannot be explained by the photochemical model predicting a monotonic recovery (increase) of the polar total ozone since 2000. (2) There is also a strong spatial correlation observed: the O_3 hole is exactly located in the lower polar stratosphere at ~18 km where the ionization rate of CRs producing electrons is the strongest [8,23]. (3) There are known PSC ice particles in the winter polar stratosphere. (4) Laboratory measurements and theoretical calculations have clearly demonstrated that ice surfaces can trap electrons and enhance the electron-induced reactions of halogenated species (inorganic and organic) at the surfaces by orders of magnitude, compared with corresponding gas-phase reactions [6–16]. (5) Even if one still assumes that the photochemical model were the dominant mechanism, then a small amplitude (10%) of the CR-intensity oscillation in an 11-yr cycle would cause a nonobservable variation of the polar total ozone. To illustrate this, assuming that the photochemical model accounted for 70% of O_3 loss in the polar stratosphere (i.e., only the rest of 30% were caused by the CR mechanism), then a variation of 10% in the CR intensity would cause only a 3% variation of the polar total ozone, which would be even lower than the fluctuation level of the ozone data (Fig. 2). This is contradictory to the observed modulation amplitude of about 12% in the polar total O_3 during an 11-yr cycle, as shown in Fig. 2. Thus, the above facts (1)–(5) force one to conclude that the CR-driven electron-induced reaction is the dominant mechanism for causing the polar O_3 hole. This mechanism must be input to remove the large discrepancy between the simulated results and the observed O_3 loss [25,26].

Finally, a correct mechanism should be able to not only explain the observed data, but also to predict future trends of the O_3 hole. Since the 11-yr cycle variation of the CR intensity is predictable, the CR-driven electron reaction mechanism leads to direct predictions of one of the severest O_3 losses (due to the CR peak) in 2008–2009, and of probably another maximum around 2019–2020 if a large halogen amount is still in the stratosphere. With $I = 6098 \times 10^2$ counts/hr for 2008 and the assumption that no significant decreases of halogen loading occur in the stratosphere (valid for a short time prediction), the best-fitted linear equation from Fig. 4 gives a variation $\Delta(O_3) \approx -14.5\%$ for the October mean total O_3 over Antarctica at latitudes of 60°–90° S, relative to the value of 219 DU at 1992 [28]. It follows that the monthly mean total O_3 over Antarctica in October 2008 is predicted to be about 187 DU, close to the measured value of 181 DU in 2006 (one of the deepest holes) [28]. Because of the accumulative effect of CR-driven electron reaction products, there may exist a time delay of about 1 year between the CR intensity maximum and the maximum ozone loss in the polar hole. For instance, the largest ozone holes were observed in 1987 and 1998, respectively, corresponding to the CR intensity maxima observed in 1986 and 1997. Although atmospheric dynamics and meteorological conditions could influence the CR effect and lead to large fluctuations of the O_3 hole from year to year, a long-term trend of the polar O_3 loss (hole) is predictable. It is interesting to examine these predictions.

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