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A new Himalayan ice core CH_4 record: possible hints at the preindustrial latitudinal gradient

S. Hou[1], J. Chappellaz[2], D. Raynaud[2], V. Masson-Delmotte[3], J. Jouzel[3], P. Bousquet[3], and D. Hauglustaine[3]

[1]Key Laboratory of Coast and Island development of Ministry of Education, School of Geographic and Oceanographic Sciences, Nanjing University, Nanjing 210093, China
[2]UJF – Grenoble 1/CNRS, Laboratoire de Glaciologie et Géophysique de l'Environnement (LGGE), UMR5183, Grenoble, 38041, France
[3]IPSL/Laboratoire des Sciences du Climat et de l'Environnement (LSCE), UMR8212,CEA/CNRS/UVSQ-IPSL, 91191 Gif-sur-Yvette, France

Correspondence to: S. Hou (shugui@nju.edu.cn)

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Abstract. Two ice cores recovered from the Himalayan East Rongbuk (ER) Glacier on the northeast saddle of Mt. Qomolangma (Everest) (28°01′ N, 86°58′ E, 6518 m above sea level) give access to a tentative record of past Himalayan atmospheric mixing ratio of CH_4 spanning the past 1200 yr. The major part of the record is affected by artifacts probably due to in situ production. After selecting what may represent the true atmospheric mixing ratio, an average of 749 ± 25 ppbv of CH_4 is estimated for the late preindustrial Holocene, which is $\sim 36 \pm 17$ ($\sim 73 \pm 18$) ppbv higher than the atmospheric levels recorded in the Greenland (Antarctic) ice cores. A comparison of these new data with model simulations of the CH_4 latitudinal gradient suggests either that the models do not get a correct balance between high and low latitude CH_4 sources, or that the filtered CH_4 profile from the ER cores remains biased by small artifacts.

1 Introduction

Methane records from polar ice cores provided a wealth of information about its natural variability over several climatic cycles (Loulergue et al., 2008). Natural methane variations are partly related to orbital and millennial variations in tropical and boreal wetland extents and CH_4 emissions. There is an ongoing debate about the cause of the preindustrial increase in atmospheric CH_4 mixing ratios, which started about 5000 yr ago. Early farming has been hypothesized as contributing to this increase in atmospheric CH_4 mixing ratios during the last 5000 yr (Ruddiman et al., 2008), which is also postulated to be of natural origin (Singarayer et al., 2011). High-resolution measurements of air samples from the Antarctic Law Dome ice cores and firn air provided a detailed record of CH_4 variability during the late preindustrial Holocene (LPIH), suggesting that LPIH CH_4 changes are mainly a response to changing climate, with small contributions from human activities (MacFarling-Meure et al., 2006). A new high-precision, high-resolution record of atmospheric CH_4 from the West Antarctic Ice Sheet (WAIS) Divide ice core covering 1000–1800 AD indicates that times of war and plague when large population losses could have reduced anthropogenic emissions are coincident with short periods of decreasing global CH_4 mixing ratios (Mitchell et al., 2011). Moreover, two ice cores from Greenland provide a high-resolution $\delta^{13}C$ record of CH_4 during the last two millennia, confirming changes in pyrogenic and biogenic CH_4 sources due to both natural climate variability and changes in human population and land use (Sapart et al., 2012).

Records of CH_4 mixing ratios in the tropical or subtropical atmospheres could provide an additional insight into latitudinal CH_4 gradients and further constrain the attribution of preindustrial variations. So far, few attempts have been made to reconstruct CH_4 levels from tropical glaciers. There, the integrity of the ice core records is indeed exposed to artifacts due to in situ production by methanogenic bacteria eventually activated by surface melt (Skidmore et al., 2000;

Campen et al., 2003). Very few subtropical glaciers may have the potential to provide a reliable CH_4 preindustrial value. Warm summer temperatures and elevated concentrations of impurities are the main caveats of most alpine glacier settings, precluding access to an undisturbed methane record in air bubbles. The first record obtained from an Himalayan ice core, i.e., the Dasuopu ice core (28°23′ N, 85°43′ E, 7100 m a.s.l.), gave an average CH_4 preindustrial mixing ratio of 825 ppbv (later modified to 782 ± 40 ppbv; Li et al., 2010) during the last two millennia (Yao et al., 2002). This value is ~ 120 ppbv (or ~ 160 ppbv) higher than its Greenland (or Antarctic) counterparts. While such a difference is comparable to the modern inter-polar CH_4 gradient maximum of ~ 140 ppbv (Dlugokencky et al., 1994), simulations of preindustrial CH_4 suggest much lower methane mixing ratios in the subtropical areas than in the higher northern latitudes (Houweling et al., 2000). The Dasuopu record is also characterized by a very large methane variability at small depth scales, which cannot reflect the true atmospheric evolution, as gas diffusion and trapping smooth out any subdecadal variability (e.g., Buizert et al., 2012). Therefore the average CH_4 mixing ratio deduced from the Dasuopu record is clearly biased. Detailed CH_4 measurements along the last interglacial section of the NEEM core (Greenland) and along specific sections of the Dye 3 core (Greenland) have shown large peaks associated with melt layers (NEEM community members, 2013). The Dasuopu record is most probably similarly affected by artifacts generating local peaks and artificially increasing the average CH_4 mixing ratios observed along the core. Here we present new CH_4 results spanning the last 1200 yr, obtained from two new Himalayan high-elevation ice cores, in an attempt to re-evaluate the CH_4 mixing ratio difference between the Himalayas and polar sites during the LPIH.

2 The ice cores

We recovered one ice core to bedrock (117.06 m deep, hereinafter referred to as "Core2001") in 2001, and two more cores to bedrock (108.83 m and 95.80 m deep, respectively) in 2002 from the East Rongbuk (ER) Glacier (27°59′ N, 86°55′ E, 6518 m a.s.l.) on the northeast saddle of Mt. Qomolangma (Everest) using an electromechanical drill in a dry hole. The CH_4 measurements were performed on the Core2001 and only the 108.83 m core of 2002 (hereinafter referred to as "Core2002"). Diameters are 6.9 cm and 9.4 cm for Core2001 and Core2002, respectively. Ice layers are horizontal along the cores, suggesting negligible ice dynamical deformation. After the drilling was finished, the ice cores were packed into the insulated boxes, which were hauled down the mountain by yaks to the base camp (more details in the Supplement), where the ice cores were transferred into the refrigerators. The core temperature inside the insulated boxes was tried to be kept as low as possible ($< -5\,^\circ$C) in order to accommodate for gas analysis, but slight warming of the ice compared to the natural existing temperatures in the glacier ($< -8.9\,^\circ$C) could not be prevented. However, this slight warming condition was retained for less than 20 h. A potential increase of CH_4 due to this slight warming might be negligible, given the fact that the ~ 1 cm-thick veneer ice was removed when preparing the samples for the CH_4 measurements (Bereiter et al., 2009). We maintained the core temperature well below $-8.9\,^\circ$C at other times.

The ER Glacier covers an area of 48.45 km^2 with a length of 14 km. Its equilibrium line is at 6250 m a.s.l., and is amongst the highest on Earth. A repeating survey with a Sokkia GSS1A Global Positioning System (accuracy of ± 5 mm over distances up to 10 km) in 1998 and 2002 did not identify horizontal movement at the drill site. The average annual net accumulation is ~ 400 mm water equivalent, as determined by snow pit and ice core studies (Kaspari et al., 2008). Borehole temperatures in the 108.83 m core ranged from $-8.9\,^\circ$C at 10 m to a minimum of $-9.6\,^\circ$C at 20 m, then warmed slightly to $-8.9\,^\circ$C at the bottom. However, snow melting occurs at the ER drilling site in summer and percolates downwards. Refreezing of meltwater speeds up the later stages of snow transformation, resulting in low gas content in the summer strata, which is used as an indicator of summer temperature trends (Hou et al., 2007).

3 Measurements and results

The CH_4 measurements were performed at LGGE (Laboratoire de Glaciologie et Geophysique de l'Environnement) in 2003 using an automated wet-extraction method. The analytical method is described in detail by Chappellaz et al. (1997). In brief, an ice sample ($\sim$ 40–50 g) is melted under vacuum in a glass container sealed with a viton O-ring and then refrozen from the bottom with an alcohol bath at $-50\,^\circ$C to expel gas dissolved in the water (extraction efficiency of 99 %). The gas sample is then expanded into an evacuated sample loop and injected into the gas chromatograph (Varian 3300) with a multi-position Valco valve. N_2+O_2+Ar and CH_4 are separated on a Porapak-N column (3 m, 1.8 mm ID) and detected with a thermal conductivity detector (TCD) and a flame ionization detector (FID), respectively. Each sample was measured three times. Calibration is performed against a standard gas (Air Liquide) with 499 ppbv of CH_4 in air. Linearity of the FID was checked by measuring CSIRO standard gases with CH_4 mixing ratios in air ranging from 394 to 1679 ppbv. Blanks of the experimental procedure, determined by adding the 499 ppbv gas standard to artificial bubble-free ice (totally degassed ice produced from ultra-pure water), amount to 20 ± 13 (2σ) ppbv and are substracted from all results.

We analyzed 77 and 112 samples from Core2001 and Core2002, respectively. All the samples are taken below the bubble close-off depth (28.4 m and 26.3 m for Core2001 and Core2002, respectively) to the bottom. The CH_4 profiles are

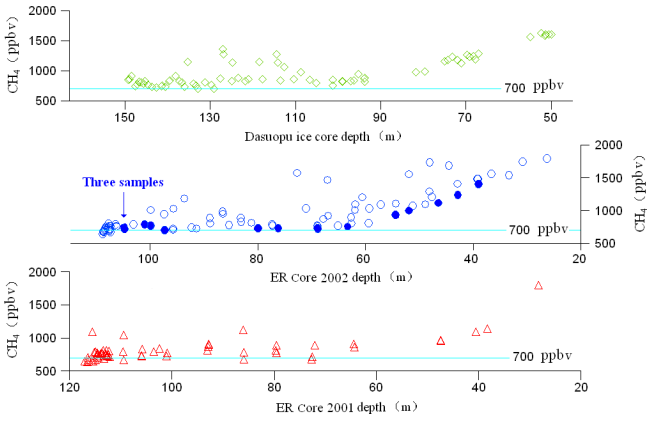

Fig. 1. CH_4 profiles from three Himalayan ice cores. The average 700 ppbv preindustrial CH_4 level depicted by polar ice cores is indicated with a horizontal light blue line. The solid circles in the ER Core2002 panel stand for the filtered 15 samples that are used for the discussion.

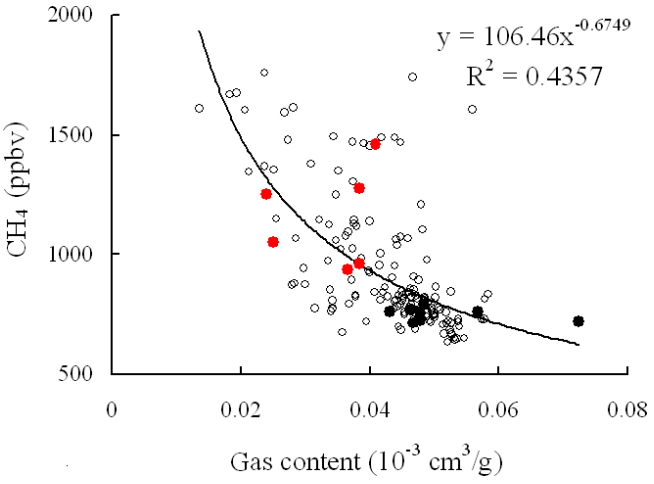

Fig. 2. Distribution of CH_4 mixing ratios against gas content of the same samples in ER ice cores, with an exponential fit. Samples with CH_4 mixing ratios over 1800 ppbv are excluded. The solid circles stand for the filtered 15 samples that are used for the discussion, with the red solid circles for the industrial records and the black solid circles for the preindustrial records.

shown as a function of depth in Fig. 1, and the CH_4 profile of 75 samples from the Himalayan Dasuopu ice core that were measured by the same set of instruments at LGGE is included for comparison (Yao et al., 2002; Li et al., 2010). Similar features are observed for all three Himalayan ice cores: CH_4 values measured on neighboring samples exhibit a large scatter that can exceed hundreds of ppbv, incompatible with normal firn air diffusion and gas-trapping processes, and pointing to local artifacts causing anomalously high CH_4 values, as recently observed on a shallow ice core from the North Greenland Eemian project (NEEM-2011-S1) by continuous CH_4 measurements (Rhodes et al., 2013), NEEM interglacial ice and Dye 3 ice sections (NEEM community members, 2013). In the case of melting, atmospheric gases diffuse into the meltwater, which is subsequently refrozen within the glacier with a methane–air solubility ratio of 1.9 for water at 0 °C in equilibrium with the atmosphere. Similarly to the last NEEM interglacial ice, a reverse relationship is apparent between CH_4 and gas content measurements that were performed simultaneously by gas chromatography on the same set of samples (Hou et al., 2007; Fig. 2), indicating that the low CH_4 mixing ratios correspond to samples with relatively high gas content, i.e., from winter ice layers whose air content was rarely impaired by melting (Hou et al., 2007; Li et al., 2010). Although methanogens that could be cultured oligotrophically at low temperatures (0.3 to 4 °C) were identified from the debris-rich basal ice layers of high Arctic glaciers (Skidmore et al., 2000), relatively low concentrations of culturable bacteria were observed in the ER ice core (Zhang et al., 2007). On the other hand, Rhodes et al. (2013) have shown that local methane peaks are associated with organic-rich ice layers. The number of bacteria may thus not be the limiting factor in producing CH_4 anomalies, but instead the amount of organic substrate may play a more important role. In the following, we attempt to filter out the ER CH_4 series from artifacts, in order to tentatively reconstruct a preindustrial and regional atmospheric CH_4 level using the baseline CH_4 profile of ER Core2002.

Corrections may be required to account for changes in firn thermal and gravitational diffusion. The magnitude of this gravitational fractionation is proportional to the thickness of the firn diffusive zone, which can be estimated using $^{15}N/^{14}N$ measurements in N_2 ($\delta^{15}N_2$). Twenty samples randomly collected in the depth range of 102–117 m of Core2001 yield a $\delta^{15}N_2$ maximum of 0.089 ‰, resulting in a negligible correction of about 0.4 ppbv for the LPIH samples (calculation following Sowers et al., 1989). Thermal fractionation is expected to have only a minor impact on the composition of bubble air in tropical glaciers (Campen et al., 2003). We conclude that the magnitude of the gravitational and thermal fractionation is negligible, and their effect was ignored.

4 Dating

Core2002 was annually dated back to 1534 AD at a depth of 98.0 m using seasonal variations of δD and soluble ions, and the time scale was verified by identifying large volcanic horizons. Below 98.0 m, annual layer counting is not possible due to layer thinning. Thus, prior to 1534 AD the core was dated using an ice-flow model (Kaspari et al., 2008). Dating uncertainties are estimated to be negligible at 1963 AD, based on a volcanic Bi horizon from the Agung eruption, and ± 5 yr at 1534 AD based on repeated annual layer counting (Kaspari et al., 2008).

We estimate a present gas–ice age difference of ∼ 30 yr, based on the firn gas measurements at the Himalayan Dasuopu Glacier and calculations from the present accumulation rate and temperature (Hou et al., 2007). The gravitational signal in the isotopic composition of atmospheric air in the firn can be used to infer information about the past thickness of the diffusive column (Caillon et al., 2001). $\delta^{15}N_2$ measurements along the ER cores give an average of 0.035 ± 0.025 ‰ (1σ) and a maximum of 0.089 ‰, resulting in ∼ 8 to 20 m of diffusive column thickness with a firn temperature of −8.9 °C. Although there might be limitations in this approach due to the existence of melt layers along the Himalayan ice cores (Hou et al., 2007; Li et al., 2010), the small δ values imply a relatively constant firn thickness that controls the gas–ice age difference. Thus we simply assume that the gas age is 30 years younger than its corresponding ice age for the studied period.

Accurate dating of Core2001 is not available yet. Therefore we only discuss below the CH_4 record of Core2002 covering the last 1200 yr.

5 Discussion

A large part of the CH_4 profile of Core2002 (as well as those of Core2001 and Dasuopu) is affected by in situ artifacts. We thus attempt to define a criterion allowing us to exclude the measurements affected by these artifacts and to retain those that may reflect the atmospheric composition at the time of gas trapping. We first excluded 34 measurements with one standard deviation > 20 ppbv for Core2002, due to unsatisfactory chromatography, system leaks, or extremely low gas content caused by possible partial snow melting. There are 6 samples collected at depths of 26.3 m, 29.6 m, 48.0 m, 48.1 m, 83.1 m and 90.4 m, respectively, with CH_4 mixing ratios over 1800 ppbv (i.e., the modern CH_4 mixing ratio measured at Mt. Waliguan on the Tibetan Plateau – one of the CMDL baseline stations), which are thus removed. Such anomalously high mixing ratios of CH_4 were also observed in a Sajama ice core from the northern end of the Bolivian altiplano (18°6′ S, 68°53′ W, 6048 m a.s.l.), and suggested to be due to metabolizing microorganisms (Campen et al., 2003). Rhodes et al. (2013) identified abrupt (20–100 cm depth interval), high-amplitude (35–80 ppbv excess) CH_4 spikes in the Greenland NEEM-2011-S1 ice core, and suggested that these spikes may result from very localized biological in situ production in polar glacial ice.

We then filter out the remaining artifact by using a criterion on the deduced CH_4 annual variation rate. The CH_4 record from WAIS Divide, Antarctica, suggested a maximum growth rate of 1 ppbv yr^{-1} during the period from the preindustrial back to 1000 AD (Mitchell et al., 2011). We thus adopt ± 1 ppbv yr^{-1} as a maximum acceptable change between adjacent Core2002 samples, possibly reflecting atmospheric composition changes. We then reject the high CH_4 values when the growth rate calculated before or after them overruns this criterion. The procedure obviously lead to selecting low CH_4 values, as we assume that there is no identified process able to consume CH_4 preserved in the ice core bubbles. As to the industrial period, Etheridge et al. (1998) identified high CH_4 growth rates peaking at ∼ 17 ppbv yr^{-1} in 1981 AD. Thus we adopt this value as a maximum growth rate between adjacent Core2002 samples over the recent increasing part of the record. We applied the filtering from the top. For any comparison of the neighboring measurements, we just choose the lower value after satisfying a growth rate criterion. Once one measurement was removed, the criterion was further applied to samples that moved next to each other. Finally, 15 measurements (i.e., the solid circles of Figs. 1 and 2) were retained in the full record, satisfying the selection criterion and possibly reflecting the atmospheric composition at the ER site (Fig. 3). Among them, nine measurements date from the LPIH, providing an average CH_4 of 749 ± 25 ppbv. This is 76 ppbv (Yao et al., 2002) or 33 ppbv (Li et al., 2010) lower than its counterpart of the Dasuopu ice core, albeit ∼ 36 ± 17 (∼ 73 ± 18) ppbv higher than the GRIP (Law Dome) LPIH records. The higher CH_4 mixing ratios of the Dasuopu core may be due to its including more biased CH_4 measurements than the ER core. Core2002 and GRIP CH_4 measurements were performed in the same laboratory, with the same extraction and analysis technique and the identical standard gas, so that this CH_4 comparison can be considered in terms of absolute values.

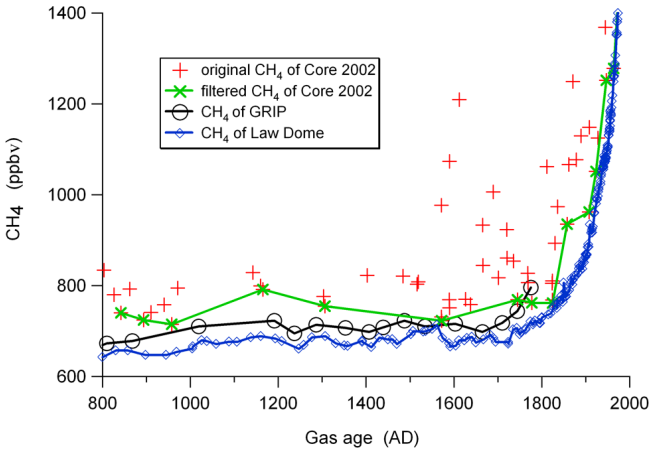

Fig. 3. The background CH_4 profiles of Core2002 with comparison to the GRIP (Chappellaz et al., 1997) and Law Dome ice core (MacFarling-Meure et al., 2006) records.

Preindustrial CH_4 simulations with a three-dimensional chemistry–transport model (Houweling et al., 2000) provide a preindustrial annual average CH_4 mixing ratio of 711–717 ppbv for the grid box covering the Himalayas, and 733 ppbv for the Greenland grid box. Although problems may arise from the use of a coarse grid model, especially for the high Himalayas, the modeling results are broadly

consistent with the Greenland LPIH level observed in the GRIP core, but much lower than our estimation from the ER2002 data selection. Similar results have also been obtained using the LMDz-INCA climate–chemistry model (Kaplan et al., 2006). Present-day CH_4 simulations along the surface-air sampling sites of the NOAA Global Monitoring Network roughly distributed along latitudes (Fig. 4) perform well in comparison with CH_4 gradient observations (Dlugokencky et al., 1994). Methane retrievals from the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) suggest that the model underestimates CH_4 mixing ratios over the Himalayas and Tibetan Plateau, potentially the result of systematic errors related to the topography of the high altitudes and model deficiencies in the stratospheric CH_4 mixing ratios (Bergamaschi et al., 2009). The lower LPIH CH_4 mixing ratios than the ER ice core data selection (Fig. 4) may also result from an underestimation of the magnitude and development of boreal CH_4 sources in the LMDz-INCA (Kaplan et al., 2006). Alternatively, the ER 2002 CH_4 profile presented here after data selection may still be affected by local artifacts, although less pronounced than for the rejected data. The only way to ascertain this would be to run continous-flow CH_4 measurements with repeated analyses along parallel ice sticks (as performed on the NEEM-2011-S1 Greenland ice core by Rhodes et al., 2013) using Himalayan ice cores from different sites, or by applying discrete replicate sampling from the same ice core depth intervals. Replicable features in the resulting CH_4 record will bring the required level of confidence for the comparison with polar records. Recovering suitable ice cores is becoming an emergency, due to the rapid warming of high-altitude glaciers (Kehrwald et al., 2008).

6 Conclusions

The Himalayan ER ice cores allowed us to tentatively retrieve a background record of High Himalayan atmospheric CH_4 mixing ratios covering the last 1200 yr. Latitudinal gradients are expected to provide an independent observational constraint from subtropical regions for assessing the relative weight of different CH_4 sources with time, in addition to the temporal trend, interpolar gradient, and stable isotopic ratio evolution. After removing obvious outliers and attempting to select interpretable data based on the CH_4 rate of change, the resulting methane profile depicts a positive gradient between the Himalayan latitude and Greenland, which is not consistent with simulations by climate–chemistry coupled models. Such model–data mismatch may arise from model caveats related to the magnitude of boreal methane fluxes or from the possibility that the selected ER CH_4 data are still affected by artifacts which could not be removed with the applied criteria.

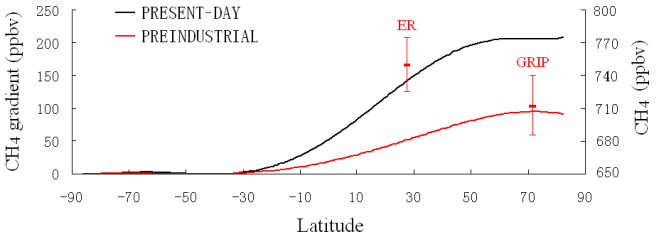

Fig. 4. CH_4 latitudinal gradients for the present-day and the preindustrial periods simulated with the LMDz-INCA climate–chemistry coupled model (left vertical axis), with comparison to the ER (this study) and the GRIP (Chappellaz et al., 1997) records (right vertical axis).

More ice core CH_4 records from high mountains (e.g., high areas in central Asia, the Alps, Caucasus, Alaska, the Andes) are necessary for investigating latitudinal gradients and obtaining robust features. It is becoming an urgent need, as alpine glaciers are warming fast and becoming less suitable for trace-gas investigations due to disturbances related to melting or in situ production. Future measurements should rely on continuous-flow techniques or discrete replicate sampling from the same depth intervals in order to pick up all details regarding the depth/CH_4 trends, the replicability of the records, and to eventually extract a suitable atmospheric reconstruction.

Supplementary material related to this article is available online at http://www.clim-past.net/9/2549/2013/cp-9-2549-2013-supplement.zip.

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