AGU PUBLICATIONS

Geophysical Research Letters

RESEARCH LETTER
10.1002/2014GL062814

Key Points:
- High-frequency measurements of halocarbon have been made in Japan
- These data combined with model suggest large Japanese emissions in 2011
- The extraordinary halocarbon emissions are likely due to the Tohoku earthquake

Supporting Information:
- Figure S1
- Figure S2
- Table S1
- Table S2
- Text S1

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Citation:
Saito, T., X. Fang, A. Stohl, Y. Yokouchi, J. Zeng, Y. Fukuyama, and H. Mukai (2015), Extraordinary halocarbon emissions initiated by the 2011 Tohoku earthquake, *Geophys. Res. Lett.*, *42*, 2500–2507, doi:10.1002/2014GL062814.

Received 9 DEC 2014
Accepted 10 MAR 2015
Accepted article online 12 MAR 2015
Published online 6 APR 2015

Extraordinary halocarbon emissions initiated by the 2011 Tohoku earthquake

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Abstract The Tohoku earthquake of 11 March 2011, with moment magnitude $M_w = 9.0$, and subsequent tsunami caused catastrophic structural damage in east Japan. Using high-frequency atmospheric monitoring data, we show that emissions of halocarbons, potent greenhouse gases and stratospheric ozone-depleting substances, dramatically increased shortly after the earthquake and that annual emissions were significantly higher in 2011 than in other years. We estimate that the sum of earthquake-related emissions of the six studied halocarbon species (CFC-11, HCFC-22, HCFC-141b, HFC-134a, HFC-32, and SF_6) was 6.6 (5.2–8.0) Gg, which is equivalent to ozone depletion potential-weighted emissions of 1.3 (1.1–1.6) Gg with a global warming potential equivalent to 19.2 (15.8–22.5) Tg of carbon dioxide. These extraordinary halocarbon emissions are likely due to destruction of building components containing halocarbons, such as air conditioners, foam insulation, and electrical equipment.

1. Introduction

Halocarbons such as chlorofluorocarbons (CFCs), hydrochlorofluorocarbons (HCFCs), and hydrofluorocarbons (HFCs) have been widely used for a variety of applications such as refrigerants in refrigerators and air conditioners and as blowing agents in foam manufacture. However, CFCs and HCFCs are regulated under the Montreal Protocol on the Substances that Deplete the Ozone Layer because of their contributions to stratospheric ozone depletion [*Montzka and Reimann*, 2011]. Although CFCs reside in certain products (e.g., building insulation foam) and HCFC are still used, HFCs are currently used in developed countries as major substitute gases for the ozone-depleting substances and HFCs emissions are rapidly increasing [*Velders et al.*, 2009]. HFCs are powerful greenhouse gases and are therefore included in the Kyoto Protocol to the United Nations Framework Convention on Climate Change together with other greenhouse gases (e.g., sulfur hexafluoride (SF_6) and perfluorocarbons (PFCs)) [*Forster et al.*, 2007].

On 11 March 2011, the Tohoku earthquake and tsunami brought extensive damage to northeastern Honshu Island. Destruction of building components containing halocarbons may be expected to release these substances into the atmosphere. However, there is little information about the impact of the earthquake on halocarbon emissions. In this study, we present in situ measurements of atmospheric halocarbons at three Japanese stations (Hateruma Island, Cape Ochiishi, and Ryori). By combining the atmospheric measurements with an atmospheric transport model and an inverse method, we estimate halocarbon emissions from Japan.

2. Experiments

Halocarbons including CFCs, HCFCs, HFCs, PFCs, halons, and SF_6 have been measured at Hateruma and Ochiishi, and ozone-depleting substances including CFCs have been measured at Ryori. Hourly measurements of halocarbons have been performed at Hateruma (24.1°N, 123.8°E) and at Ochiishi (43.1°N, 145.3°E) as part of the National Institute for Environmental Studies halocarbon monitoring project. Ambient air is analyzed with a fully automated preconcentration/gas chromatography/mass spectrometry system. Halocarbons are quantified with gravimetrically prepared standard gases, which are analyzed in the same manner as the ambient air samples after every five air analyses [*Enomoto et al.*, 2005; *Yokouchi et al.*, 2006; *Saito et al.*, 2010]. At the Ryori Global Atmospheric Watch regional station (39.3°N, 142.3°E), halocarbons including CFC-11 have been

measured by the Japan Meteorological Agency (JMA) using a gas chromatograph equipped with an electron capture detector [*World Meteorological Organization*, 2012]. For Hateruma and Ochiishi, the measurements are reported on the NIES-08 calibration scale, whereas for Ryori, the JMA calibration scale was used. Intercomparison experiments between the calibration scales for CFC-11 measurement showed agreement within ±1%. For the inversions, we ignored this small difference.

The inversion method used in this study is based on backward Lagrangian particle dispersion model calculations from the measurement stations using FLEXible PARTicle (FLEXPART) model [*Stohl et al*., 2005], which establishes source-receptor relationships between emissions and atmospheric concentrations [*Stohl et al*., 2009]. A detailed gridded a priori emission data set was developed by spatially disaggregating recent reported global emissions and their trends [*Keller et al*., 2011]. For disaggregation of global emissions to different countries, for HFC-134a, HFC-32, and SF_6, emissions at country level including Japan were taken from the United Nations Framework Convention on Climate Change data set [*United Nations Framework Convention on Climate Change* (*UNFCCC*), 2014]. The remaining emissions in all other countries were disaggregated according to the Population Density Grid Future Estimates v3 [*Consortium for International Earth Science Information Network*, 2005]. This data set was also used for within-country gridding of all national emissions. A Bayesian optimization technique was used to estimate both emission strength and distribution over Japan as well as other countries. The algorithm optimizes the model agreement with the measurements while also considering a priori emissions and the uncertainties in the emissions, observations, and the model simulations. The inversion for CFC-11 used Hateruma, Ochiishi, and Ryori measurements, while inversions for the other species were performed with Hateruma and Ochiishi. We repeated the inversions by independently setting the a priori emissions and uncertainties to 50%, 100%, 150%, or 200% of their original values. The definition methods for a priori uncertainty, model uncertainty, baseline, and other terms are the same as those described in *Stohl et al.* [2009]. Detailed information for the FLEXPART backward simulation and inversion algorithm is available in the supporting information.

3. Results and Discussion

The Tohoku earthquake occurred on 11 March 2011 off the Pacific coast of Japan, and the earthquake and subsequent tsunami brought extensive damage to northeastern Honshu Island. Air that had resided over the affected region 1–2 days after the earthquake traveled northeastward and arrived at Ochiishi on 13–14 March (see Figure 1a). In this air mass, we observed substantially enhanced mixing ratios for the halocarbon species CFC-11, HCFC-22, HCFC-141b, HFC-134a, HFC-32, and SF_6 (Figure 1b). Reasoning that the relative enhancement ratio of two species reflects their emission ratio in the source region (tracer ratio method) [*Palmer et al*., 2003], we compared the relative enhancements of mixing ratios of the five halocarbons other than HFC-134a (C_x) to that of HFC-134a ($C_{HFC\text{-}134a}$)—the best reference tracer for quantifying halocarbon emissions in Japan [*Li et al*., 2011]—in east Japanese pollution events before and after the earthquake. The east Japanese pollution events at Ochiishi during the period of 2009–2012 were identified by an air mass back trajectory analysis (see supporting information). The resulting monthly $\Delta C_x/\Delta C_{HFC\text{-}134a}$ ratios in March 2011 were highest among all months in 2009–2012 for all studied ratios. The mean values in March 2011 beyond the 95% upper prediction limits are calculated from data before the earthquake (January 2009 to February 2011) for all studied ratios other than $\Delta C_{CFC\text{-}11}/\Delta C_{HFC\text{-}134a}$ (Figure 2), although the associated P values are low only for $\Delta C_{HFC\text{-}32}/\Delta C_{HFC\text{-}134a}$ ($P = 0.01$), moderate for $\Delta C_{HCFC\text{-}141b}/\Delta C_{HFC\text{-}134a}$ ($P = 0.08$) and $\Delta C_{SF6}/\Delta C_{HFC\text{-}134a}$ ($P = 0.10$), and high for $\Delta C_{HCFC\text{-}22}/\Delta C_{HFC\text{-}134a}$ ($P = 0.18$). This result suggests that emissions of the halocarbons were extraordinarily enhanced relative to HFC-134a emissions shortly after the earthquake.

We found another remarkable feature in the time series of CFC-11 mixing ratios in 2011 (Figure 3). Before the earthquake, CFC-11 pollution plumes were detected almost exclusively at Hateruma, which occasionally receives air masses influenced by CFC-11 emissions in developing countries in continental Asia. Such pollution plumes have, however, rarely been observed at the Cape Ochiishi and Ryori sites which are mainly influenced by emissions in Japan. After the earthquake, however, pollution events began to be observed frequently at Ochiishi and Ryori. Both the frequency and magnitude of the pollution events were most pronounced at Ryori, which is located in the disaster area. A Kolmogorov-Smirnov nonparametric test showed that the enhanced concentration values above baselines at Ryori after the earthquake (14 May

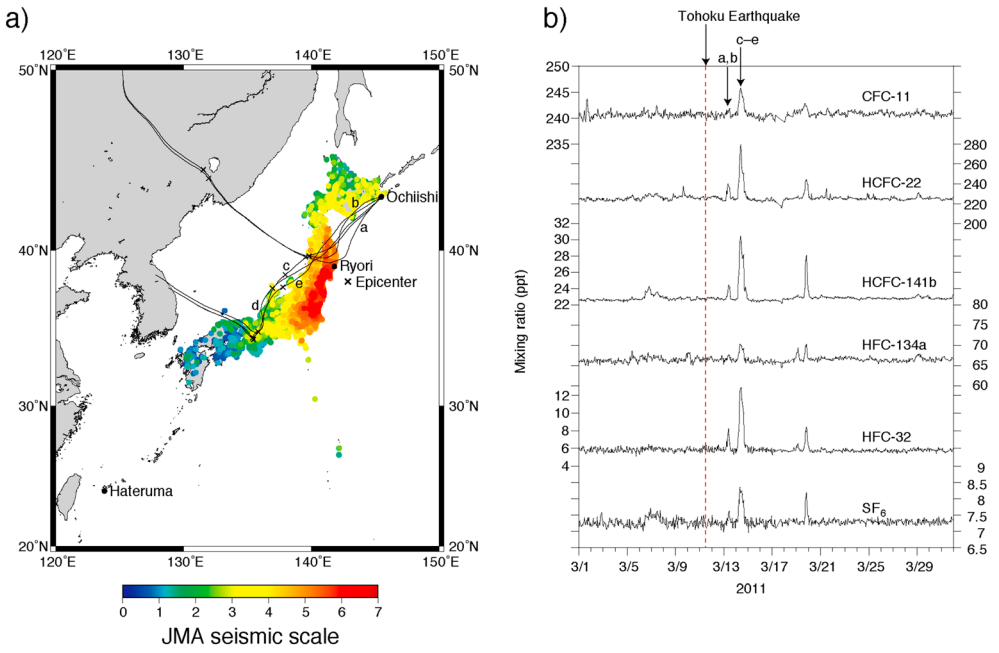

Figure 1. Atmospheric measurements of halocarbons in Japan. (a) Map showing the locations of atmospheric monitoring stations and the distribution of seismic intensity of the 11 March 2011 Tohoku earthquake (Japan Meteorological Agency scale) [*Japan Meteorological Agency*, 2012]. The lines represent 3 day back trajectories for air masses arriving at 500 m altitude over Cape Ochiishi on 13 March 2011 at (a) 06:00 LT and (b) 09:00 LT and on 14 March 2011 at (c) 06:00 LT, (d) 09:00 LT, and (e) 12:00 LT (calculated by Meteorological Data Explorer trajectory model, METEX) [*Zeng et al.*, 2010]. The 24 h intervals are labeled with "*cross*" symbols. (b) Time series of six halocarbons measured at Ochiishi in March 2011. The arrows indicate the starting time of the back trajectories.

2011 to 11 March 2012; measurements at Ryori were not made during 11 March to 13 May 2011) were significantly higher than those before (14 May 2010 to 11 March 2011) ($P < 0.01$). This finding suggests that the earthquake strongly increased the CFC-11 emissions in the affected area. This is also suggested from CFC-11 pollution roses at Ryori (Figure 4), which shows that high CFC-11 mixing ratios (i.e., 75th and 95th percentiles) in 2011 were associated with west or southwest wind. The south-west sector with respect to Ryori contains the most affected areas along the Pacific coast of Japan.

To estimate Japanese halocarbon emissions, we used a Bayesian inverse modeling approach. The method modifies an a priori emission distribution to improve the agreement between simulated and observed mixing ratios but also keeps the a posteriori emissions constrained by the a priori emissions [*Stohl et al.*, 2009] (see Figure S1 in the supporting information for the time series of the measured and the simulated HCFC-22 mixing ratios). The inversion reduces the uncertainty from the a priori to the a posteriori simulation (see Figure S2 in the supporting information for the map of gridded uncertainty reduction between a priori and a posteriori HCFC-22 emissions). The time series of a posteriori monthly Japanese emissions, derived from monthly inversion over moving 3 month periods (Figure 5), show that the a posteriori emissions were consistently larger after the earthquake than before. Paired sample *t* tests also showed that the emissions of halocarbons were enhanced from March 2011 to February 2012 (2011#) (statistically significant at the 0.05 level) compared to 2010# (March 2010 to February 2011) for all studied species and compared to 2012# (March 2012 to February 2013) for all species other than HCFC-141b (see Table S1 in the supporting information). This too suggests that halocarbon emissions were altered by the earthquake.

The estimated annual emissions of all studied halocarbons were higher in 2011# than those in 2010# and 2012# (Table 1). To derive the extra emissions triggered by the 2011 Tohoku earthquake, we considered the excess of the 2011# emissions over the averaged 2010# and 2012# emissions. The relative increases were 10−111% for the six species studied. Our inversion setup likely leads to a conservative estimate of the true earthquake-related emissions for three reasons: (1) A first, probably strong, emission pulse on 11 March might

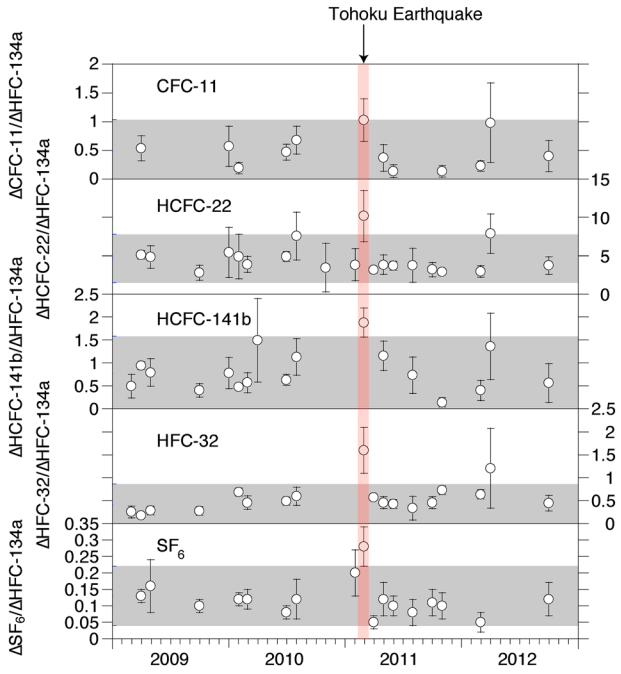

Figure 2. Monthly mean ratios (parts per trillion (ppt)/ppt) of ΔCFC-11/ΔHFC-134a, ΔHCFC-22/ΔHFC-134a, ΔHCFC-141b/ΔHFC-134a, ΔHFC-32/ΔHFC-134a, and $\Delta SF_6/\Delta$HFC-134a in air masses that traveled over east Japan before arriving at Ochiishi. The error bars represent the 95% confidence intervals from the monthly averages. The shaded areas represent the 95% prediction intervals calculated from data before the earthquake.

not have been fully captured by the measurement data. Measurements at Ryori were interrupted from 11 March to 13 May 2011; back trajectories show that air residing over Japan on 11 March 2011 was not sampled at Hateruma and that measurements at Cape Ochiishi only partially sampled such air. (2) The duration of the earthquake-related emission pulse might have been longer than a year (see supporting information). (3) The a priori emission values were kept constant for the period of 2009–2012 (see supporting information).

We found that HCFC-22, used mainly as a refrigerant gas, was the largest contributor (~50%) to the total emissions arising from the Tohoku earthquake, likely due to emissions from damaged refrigerators and air conditioners. This result is consistent with an extensive survey conducted by the Ministry of the Environment of Japan and the Japan Refrigeration and Air Conditioning Industry Association in the affected areas. This survey found that HCFC-22 contributed ~80% of the total amount of refrigerant gases released from industrial refrigeration and air conditioning units due to earthquake damage [*Ministry of the Environment*, 2012a]. The refrigerant gases HFC-32 and HFC-134a also showed a spring peak in emissions. However, while HFC-32 and HCFC-22 showed a peak in March 2011, HFC-134a showed a broader peak with the highest emissions in April 2011. The delayed enhancement of HFC-134a is consistent with the high $\Delta C_x/\Delta C_{HFC\text{-}134a}$ ratios exclusively observed immediately after the earthquake (except for CFC-11). High emissions of SF_6 were likely caused by leaks from damaged high-voltage equipment containing SF_6 for electrical insulation, as Hitachi Cable, Ltd. reported that total SF_6 emissions in its installations doubled in 2011 compared to 2010 [*Hitachi Cable Ltd.*, 2012].

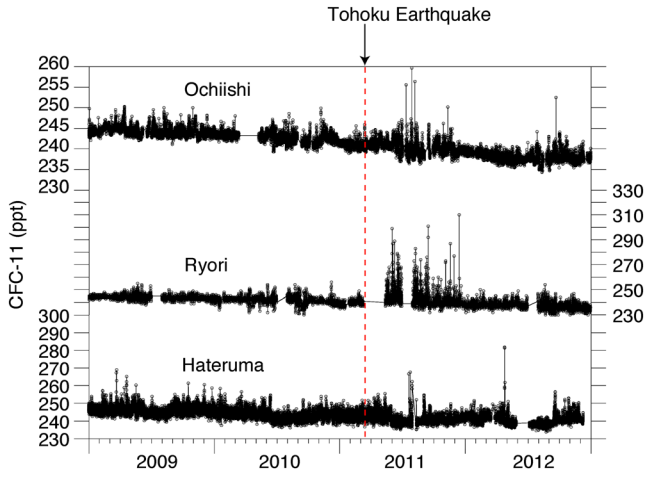

Figure 3. Time series of CFC-11 mixing ratios measured at three stations.

For CFC-11, emissions in 2011#, 2.7 (2.6–2.7) Gg/yr, were higher by 72% (53–91%) than the average for the other 2 years. CFC-11 had been used as a blowing agent for polyurethane insulation foam until its production in Japan was banned in 1996. It slowly escapes to the atmosphere from rigid closed-cell foams [*McCulloch et al.*, 2001]. Increased CFC-11 leak rates are likely due to structural damage to the insulation foams used in appliances and buildings. Unlike HCFC-22 and the other species, CFC-11 showed a distinct summertime maximum in emissions in 2011 (Figure 5), which might be the result of effective leakage from damaged foams at high temperature during the summer

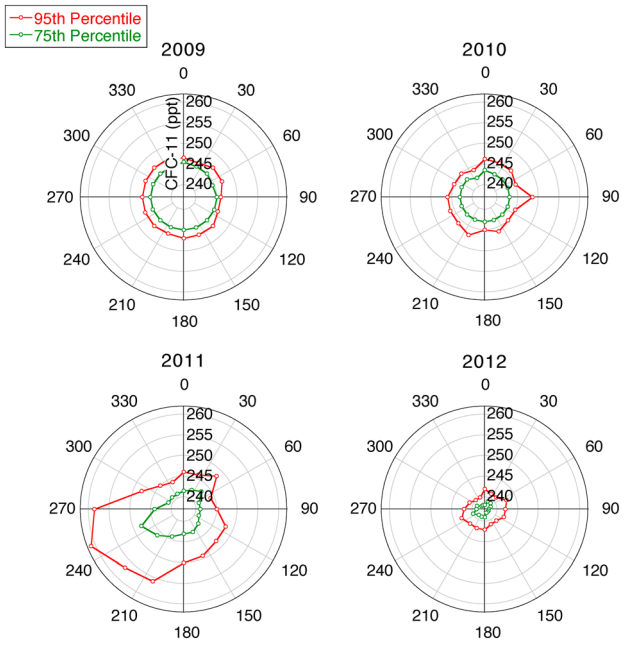

Figure 4. Annual pollution roses of CFC-11 mixing ratios (ppt) at Ryori.

[*Barnes et al.*, 2003; *Kondo et al.*, 2004]. We observed a similar seasonality with summer peaks in 2009 and 2010, although it is subtle compared to that in 2011 (Figure 5). Indeed, a calculation based on (1) a laboratory-determined temperature dependency of CFC-11 emissions from foams [*Kondo et al.*, 2004], (2) the local mean temperatures in the disaster area (3.8°C and 24.9°C in March and August 2011, respectively, at Sendai), and (3) the CFC-11 emission estimate for March 2011 suggests that CFC-11 emissions would be ~8 Gg/yr in August 2011, which exceeds our estimate. Treatment of debris is another possible candidate for the seasonal source of CFC-11, because demolition of damaged buildings and shredding of debris would dramatically enhance its release [*Kjeldsen and Jensen*, 2001]. According to the Ministry

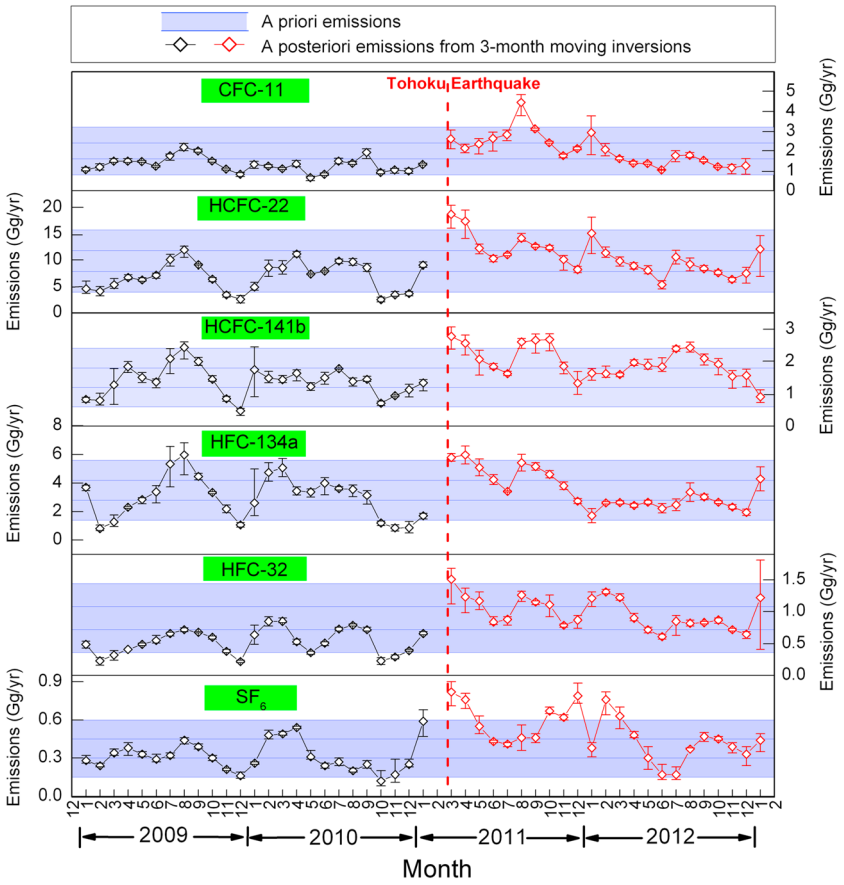

Figure 5. A posteriori emissions (Gg/yr) for Japan from monthly inversions for moving 3 month periods. The observation data before 11 March 2011 were excluded from the inversion for March 2011 (February–April 2011) to avoid combining data from before and after the earthquake. The error bars indicate ranges of values given by the inversions using a priori emissions of 50%, 100%, 150%, or 200% of their original a priori values.

Table 1. Estimates of Emissions in 2010#, 2011#, and 2012# and Emissions Caused by the Tohoku Earthquake

	Halocarbon Emissions (Gg/yr)						
	2010#	2011#	2012#	Relative Increase[a] (%)	Tohoku Earthquake Emissions (Gg)	ODP-Weighted Tohoku Earthquake Emissions (Gg)	GWP-Weighted Tohoku Earthquake Emissions (Tg CO_2eq)
CFC-11	1.6 (1.5–1.8)	2.7 (2.6–2.7)	1.5 (1.3–1.6)	72 (53–91)	1.1 (0.9–1.3)	1.1 (0.9–1.3)	5.3 (4.3–6.2)
HCFC-22	8.5 (8.3–8.8)	11.6 (11.1–11.9)	8.2 (7.9–8.5)	38 (29–47)	3.2 (2.5–3.8)	0.2 (0.1–0.2)	5.8 (4.5–6.9)
HCFC-141b	1.6 (1.6–1.7)	2.2 (2.1–2.2)	2.0 (1.8–2.1)	21 (10–30)	0.4 (0.2–0.5)	0.04 (0.02–0.06)	0.1 (0.0–0.1)
HFC-134a	3.1 (3.0–3.3)	4.1 (4.0–4.3)	2.4 (2.1–2.6)	49 (35–65)	1.4 (1.1–1.7)	0 (0–0)	1.9 (1.5–2.4)
HFC-32	0.57 (0.56–0.59)	0.94 (0.89–0.96)	0.58 (0.54–0.62)	63 (46–76)	0.4 (0.3–0.4)	0 (0–0)	0.2 (0.2–0.3)
SF_6	0.24 (0.22–0.26)	0.54 (0.53–0.55)	0.32 (0.29–0.36)	91 (73–111)	0.3 (0.2–0.3)	0 (0–0)	5.8 (5.2–6.6)
Total					6.6 (5.1–8.0)	1.3 (1.1–1.6)	19.2 (15.8–22.4)

[a]"Relative increase" is the factor by which a posteriori emissions estimated from inversions for 2011# are larger than the average of values from inversions for 2010# and 2012#.

of the Environment of Japan [*Ministry of the Environment*, 2012b, 2013], about 18.8 million tons of disaster waste (~10 times the normal yearly amount of waste in the disaster areas) was generated in the disaster areas in Tohoku, and (except for debris contaminated with radioactivity in Fukushima) the debris was transferred by August 2011 to makeshift yards where it was roughly sorted with heavy machinery and manual labor.

To quantify the total impact on ozone depletion and global warming, we weighted the emissions of each of the six measured halocarbon with its respective ozone depletion potential (ODP) [*United Nations Environment Programme Ozone Secretariat*, 2009] and global warming potential (GWP) integrated over a 100 year horizon [*Forster et al*., 2007], respectively. The ODP- and GWP-weighted earthquake emissions are estimated to be 1.3 (1.1–1.6) Gg CFC-11 equivalent and 19.2 (15.8–22.5) Tg CO_2 equivalent, respectively. If only the six species for which we have actually determined emissions are considered, the extra ODP-weighted and GWP-weighted emissions from the Tohoku earthquake amount to 38% (29–45%) and 36% (29–43%) of the total Japanese emissions of these six gases in 2011, respectively. CFC-11 was the largest and third-largest contributor in terms of ozone depletion (84% (77–89%) of the total) and global warming (28% (21–35%)), after HCFC-22 and SF_6, respectively. HFC-134a emissions also made large contributions, whereas contributions for HCFC-141b and HFC-32 were negligible. If we assume that the ODP-weighted emissions of other halocarbons did not change much during the period of 2008–2011, based on the emission estimates in Japan in 2008 [*Li et al*., 2011], the extra ODP-weighted and GWP-weighted emissions due to the Tohoku earthquake are 30% (23–36%) and 24% (20–31%) of the Japanese total halocarbon emissions in 2011, respectively. The Tohoku earthquake emissions account for approximately 4% (for SF_6) or less (2% for CFC-11 and HFC-32 and 1% for HCFC-22, HCFC-141b, and HFC-134a) of the global emissions for 2011 [*Rigby et al*., 2014].

This study suggests that halocarbon emissions in Japan substantially increased due to the 2011 earthquake and tsunami. Our result is inconsistent with bottom-up emission estimates showing no abnormal emissions in 2011 (see Table S2 in the supporting information) [*Pollutant Release and Transfer Register* (*PRTR*), 2012, 2013a, 2014; *UNFCCC*, 2014], as these inventories did not take into account emissions associated with destruction of building components by the earthquake and tsunami [*Greenhouse Inventory Office*, 2013; *PRTR*, 2013b]. For instance, for CFC-11, emissions associated with disposal of building insulating foams were not accounted for in the Japanese government's Pollutant Release and Transfer Register inventory, which assumes that all CFC-11 are emitted over the lifetime of the foam (i.e., no disposal emission). This shows that top-down analyses based on atmospheric measurements are important, especially when natural disasters make it difficult to estimate emissions based on the bottom-up approach.

We emphasize that this is a case study aimed at identifying a new mechanism of emissions. Earthquakes are frequent, and the implications of our findings are global. In 2011, another major earthquake (Christchurch) caused massive destruction, and major earthquakes in populated areas happen almost every year. Furthermore, this mechanism of emissions likely extends to other natural disasters causing structural damage to infrastructure (floods, storms, etc.). According to an insurance report [*Munich Re*, 2012], losses from natural disasters in 2011 set a new record (US$380 bn, of which US$210 bn was from the Japanese earthquake/tsunami and US$18 bn was from earthquakes in New Zealand), and annual losses have exceeded US$100 bn in 13 of the years between 1992 and 2011, with a strong upward trend [*Munich Re*, 2012]. Thus, emissions such as those we have identified

are likely to occur repeatedly and are not limited to the 2011 Japan earthquake. In most cases, the extra emissions will be difficult to detect on the global scale, but their cumulative importance is great. Given that long-lasting reservoirs such as foams, refrigerators, and electrical equipment may store substantial amounts of halocarbons, the impact of natural disasters on halocarbon emissions is twofold. First, natural disasters may accelerate the release of halocarbons, while halocarbons without regulations to destroy them would be finally emitted regardless of whether disasters occur. Second, since repairing or replacing the destroyed equipment may need fresh halocarbons (although zero-ODP and low-GWP substitutes are likely to become more common in the future), large-scale natural disasters may increase the long-term total emissions of halocarbons and thus contribute to global warming and alter the recovery of stratospheric ozone.

Acknowledgments

Observation data are available (for daily mean values at Ryori and for hourly values at Hateruma and Ochiishi) at World Data Center for Greenhouse Gases (http://ds.data.jma.go.jp/gmd/wdcgg/wdcgg.html). Xuekun Fang acknowledges China Scholarship Council for providing financial support (no. 201206010235) during his 1 year visit to the Norwegian Institute for Air Research. This work was supported by the Global Environmental Fund (Ministry of the Environment of Japan). We thank Nobukazu Oda and Fujio Shimano of the Global Environmental Forum for their help in atmospheric observations at Hateruma Island and Cape Ochiishi and Shinya Takatsuji and the staff of JMA for their great contribution to measurements at Ryori and intercomparison experiments. We also thank R.L. Thompson at Norwegian Institute for Air Research for her helpful discussions and comments on the inversion results.

The Editor thanks two anonymous reviewers for their assistance in evaluating this paper.

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